Edge-dependent reflection and inherited fine structure of higher-order plasmons in graphene nanoribbons

We investigate higher-order plasmons in graphene nanoribbons, and we present how electronic edge states and wave-function fine structure influence the graphene plasmons. Based on nearest-neighbor tight-binding calculations, we find that a standing-wave model based on nonlocal bulk plasmon dispersion is surprisingly accurate for armchair ribbons of widths even down to a few nanometers, and we determine the corresponding phase shift upon edge reflection and an effective ribbon width. Wider zigzag ribbons exhibit a similar phase shift, whereas the standing-wave model describes few-nanometer zigzag ribbons less satisfactorily, to a large extent because of their edge states. We directly confirm that also the larger broadening of plasmons for zigzag ribbons is due to their edge states. Furthermore, we report a prominent fine structure in the induced charges of the ribbon plasmons, which for armchair ribbons follows the electronic wave-function oscillations induced by intervalley coupling. Interestingly, the wave-function fine structure is also found in our analogous density-functional theory calculations, and both these and tight-binding numerical calculations are explained quite well with analytical Dirac theory for graphene ribbons.
Important role of screening the electron-hole exchange interaction for the optical properties of molecules near metal surfaces
Optical experiments on nanostructures such as molecules, one- or two-dimensional materials, are often performed with the nanostructures in close proximity of a substrate or some other polarizable media. In this case, the Bethe-Salpeter equation (BSE) can be used to calculate the optical excitations of the nanostructure by including the effect of the substrate via the screened electron-hole interaction. Here we show, that in such an approach, where the states of the substrate are not explicitly included in the BSE Hamiltonian but only enter through the screened Coulomb interaction, it is important also to screen the electron-hole exchange interaction. For the case of molecules like benzene physisorbed on the metallic Au(111) surface, the screening of the exchange interaction by the substrate redshifts the lowest optical transition by up to 300 meV. Furthermore, the screening of the exchange is essential in order to obtain the correct ordering of the size of quasiparticle and optical energy gap.
Benchmark Database of Transition Metal Surface and Adsorption Energies from Many-Body Perturbation Theory

We present an extensive set of surface and chemisorption energies calculated using state of the art many body perturbation theory. In the first part of the paper we consider 10 surface reactions in the low coverage regime where experimental data is available. Here the random phase approximation (RPA) is found to yield high accuracy for both adsorption and surface energies. In contrast, all the considered density functionals fail to describe both quantities accurately. This establishes the RPA as a universally accurate method for surface science. In the second part, we use the RPA to construct a database of 200 high quality adsorption energies for reactions involving OH, CH, NO, CO, N-2, N, O, and H over a wide range of 3d, 4d, and Sd transition metals. Due to the significant computational demand, these results are obtained in the high coverage regime where adsorbate-adsorbate interactions can be significant. RPA is compared to the more advanced renormalized adiabatic LDA (rALDA) method for a subset of the reactions, and they are found to describe the adsorbate-metal bond as well as adsorbate-adsorbate interactions similarly. The RPA results are compared to a range of standard density functional theory methods typically employed for surface reactions representing the various rungs on Jacob's ladder. The deviations are found to be highly functional, surface, and reaction dependent. Our work establishes the RPA and rALDA methods as universally accurate full ab initio methods for surface science where accurate experimental data is scarce. The database is freely available via the Computational Materials Repository (CMR).
Web of Science (2019): Indexed yes
BFI (2018): BFI-level 1
Web of Science (2018): Indexed yes
BFI (2017): BFI-level 1
Scopus rating (2017): CiteScore 4.58 SJR 2.135 SNIP 1.147
Web of Science (2017): Impact factor 4.484
Web of Science (2017): Indexed yes
BFI (2016): BFI-level 1
Scopus rating (2016): CiteScore 4.48 SJR 1.964 SNIP 1.195
Web of Science (2016): Impact factor 4.536
Web of Science (2016): Indexed yes
BFI (2015): BFI-level 1
Scopus rating (2015): CiteScore 4.68 SJR 1.886 SNIP 1.26
Web of Science (2015): Impact factor 4.509
Web of Science (2015): Indexed yes
BFI (2014): BFI-level 1
Scopus rating (2014): CiteScore 5.08 SJR 2.032 SNIP 1.447
Web of Science (2014): Impact factor 4.772
Web of Science (2014): Indexed yes
BFI (2013): BFI-level 1
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ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
BFI (2012): BFI-level 1
Scopus rating (2012): CiteScore 4.98 SJR 2.529 SNIP 1.461
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ISI indexed (2012): ISI indexed yes
Web of Science (2012): Indexed yes
BFI (2011): BFI-level 1
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Web of Science (2011): Indexed yes
BFI (2010): BFI-level 1
Scopus rating (2010): SJR 2.462 SNIP 1.362
Web of Science (2010): Impact factor 4.524
Web of Science (2010): Indexed yes
BFI (2009): BFI-level 1
Scopus rating (2009): SJR 2.158 SNIP 1.427
Web of Science (2009): Indexed yes
BFI (2008): BFI-level 1
Scopus rating (2008): SJR 1.883 SNIP 1.04
Web of Science (2008): Indexed yes
Web of Science (2007): Indexed yes
Web of Science (2006): Indexed yes
Web of Science (2005): Indexed yes
Web of Science (2004): Indexed yes
Web of Science (2003): Indexed yes
Web of Science (2002): Indexed yes
Web of Science (2001): Indexed yes
Web of Science (2000): Indexed yes

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Dissociation of two-dimensional excitons in monolayer WSe₂

Two-dimensional (2D) semiconducting materials are promising building blocks for optoelectronic applications, many of which require efficient dissociation of excitons into free electrons and holes. However, the strongly bound excitons arising from the enhanced Coulomb interaction in these monolayers suppresses the creation of free carriers. Here, we identify the main exciton dissociation mechanism through time and spectrally resolved photocurrent measurements in a monolayer WSe₂ p-n junction. We find that under static in-plane electric field, excitons dissociate at a rate corresponding to the one predicted for tunnel ionization of 2D Wannier-Mott excitons. This study is essential for understanding the photoresponse of 2D semiconductors and offers design rules for the realization of efficient photodetectors, valley dependent optoelectronics, and novel quantum coherent phases.
Janus MoSSe monolayers have been recently synthesized by replacing S by Se on one side of MoS$_2$ (or vice versa for MoSe$_2$). Due to the different electronegativities of S and Se, these structures carry a finite out-of-plane dipole moment. As we show here by means of density functional theory calculations, this intrinsic dipole leads to the formation of built-in electric fields when the monolayers are stacked to form N-layer structures. For sufficiently thin structures (N < 4), the dipoles add up and shift the vacuum level on the two sides of the film by ∼N·0.7 eV. For thicker films, the vacuum level shift saturates at around 2.2 eV due to compensating surface charges, which in turn leads to the formation of atomically thin n- and p-doped electron gases at the surfaces. The doping concentration can be tuned between 5 × 10$^{12}$ and 2 × 10$^{13}$ e/cm$^2$ by varying the film thickness. On the basis of band structure calculations and the Mott-Wannier exciton model, we compute the energies of intra- and interlayer excitons as a function of film thickness, suggesting that the Janus multilayer films are ideally suited for achieving ultrafast charge separation over atomic length scales without chemical doping or applied electric fields. Finally, we explore a number of other potentially synthesizable two-dimensional Janus structures with different band gaps and internal dipole moments. Our results open new opportunities for ultrathin optoelectronic components, such as tunnel diodes, photodetectors, or solar cells.
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BFI (2015): BFI-level 1
Scopus rating (2015): CiteScore 4.68 SJR 1.886 SNIP 1.26
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ISI indexed (2013): ISI indexed yes
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BFI (2012): BFI-level 1
Scopus rating (2012): CiteScore 4.98 SJR 2.529 SNIP 1.461
Web of Science (2012): Impact factor 4.814
ISI indexed (2012): ISI indexed yes
Web of Science (2012): Indexed yes
BFI (2011): BFI-level 1
Scopus rating (2011): CiteScore 4.92 SJR 2.339 SNIP 1.465
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ISI indexed (2011): ISI indexed yes
Web of Science (2011): Indexed yes
BFI (2010): BFI-level 1
Scopus rating (2010): SJR 2.462 SNIP 1.362
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Web of Science (2009): Indexed yes
BFI (2008): BFI-level 1
Scopus rating (2008): SJR 1.883 SNIP 1.04
Web of Science (2008): Indexed yes
Web of Science (2007): Indexed yes
Web of Science (2006): Indexed yes
Electron–phonon interaction and transport properties of metallic bulk and monolayer transition metal dichalcogenide TaS₂

Transition metal dichalcogenides have recently emerged as promising two-dimensional materials with intriguing electronic properties. Existing calculations of intrinsic phonon-limited electronic transport so far have concentrated on the semiconducting members of this family. In this paper we extend these studies by investigating the influence of electron–phonon coupling on the electronic transport properties and band renormalization of prototype inherent metallic bulk and monolayer TaS₂. Based on density functional perturbation theory and semi-classical Boltzmann transport calculations, promising room temperature mobilities and sheet conductances are found, which can compete with other established 2D materials, leaving TaS₂ as promising material candidate for transparent conductors or as atomically thin interconnects. Throughout the paper, the electronic and transport properties of TaS₂ are compared to those of its isoelectronic counterpart TaSe₂ and additional informations to the latter are given. We furthermore comment on the conventional superconductivity in TaS₂, where no phonon-mediated enhancement of T_C in the monolayer compared to the bulk state was found.

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BFI (2016): BFI-level 1
Scopus rating (2016): CiteScore 4.26 SJR 2.314 SNIP 0.915
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Emergent scale invariance of nonclassical plasmons in graphene nanoribbons

Using a nearest-neighbor tight-binding model we investigate quantum effects of plasmons on few-nanometer wide graphene nanoribbons, both for zigzag and armchair edge terminations. With insight from the Dirac description we find an emerging scale-invariant behavior that deviates from the classical model both for zigzag and armchair structures. The onset of the deviation can be related to the position of the lowest parabolic band in the band structure. Dirac theory is only valid in the parameter subspace where the scale invariance holds that relates narrow ribbons with high doping to wide ribbons with low doping. We also find that the edge states present in zigzag ribbons give rise to a blue shift of the plasmon, in contrast to earlier findings for graphene nanodisks and nanotriangles.
Fundamental limitation of electrocatalytic methane conversion to methanol

The electrochemical oxidation of methane to methanol at remote oil fields where methane is flared is the ultimate solution to harness this valuable energy resource. In this study we identify a fundamental surface catalytic limitation of this process in terms of a compromise between selectivity and activity, as oxygen evolution is a competing reaction. By investigating two classes of materials, rutile oxides and two-dimensional transition metal nitrides and carbides (MXenes), we find a linear relationship between the energy needed to activate methane, i.e. to break the first C-H bond, and oxygen binding energies on the surface. Based on a simple kinetic model we can conclude that in order to obtain sufficient activity oxygen has to bind weakly to the surface but there is an upper limit to retain selectivity. Few potentially interesting candidates are found but this relatively simple description enables future large scale screening studies for more optimal candidates.

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High-Throughput Computational Assessment of Previously Synthesized Semiconductors for Photovoltaic and Photoelectrochemical Devices

Using computational screening we identify materials with potential use as light absorbers in photovoltaic or photoelectrochemical devices. The screening focuses on compounds of up to three different chemical elements which are abundant and nontoxic. A prescreening is carried out based on information from the Inorganic Crystal Structure Database and Open Quantum Materials Database. The light absorption, carrier mobility, defect tolerance, and stability of the materials are assessed by a set of simple computational descriptors. The identified 74 materials include a variety of pnictogenides, chalcogenides, and halides. Several recently investigated light absorbers, such as CsSnI3, CsSnBr3, and BaZrS3, appear on the list.

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Source-ID: 2395556412
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Interlayer Excitons with Large Optical Amplitudes in Layered van der Waals Materials

Vertically stacked two-dimensional materials form an ideal platform for controlling and exploiting light-matter interactions at the nanoscale. As a unique feature, these materials host electronic excitations of both intra- and interlayer type with distinctly different properties. In this Letter, using first-principles many-body calculations, we provide a detailed picture of the most prominent excitons in bilayer MoS2, a prototypical van der Waals material. By applying an electric field perpendicular to the bilayer, we explore the evolution of the excitonic states as the band alignment is varied from perfect line-up to staggered (Type II) alignment. For moderate field strengths, the lowest exciton has intralayer character and is almost independent of the electric field. However, we find higher lying excitons that have interlayer character. They can be described as linear combinations of the intralayer B exciton and optically dark charge transfer excitons, and interestingly, these mixed interlayer excitons have strong optical amplitude and can be easily tuned by the electric field. The first-principles results can be accurately reproduced by a simple excitonic model Hamiltonian that can be straightforwardly generalized to more complex van der Waals materials.

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Web of Science (2017): Impact factor 12.08
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Scopus rating (2016): CiteScore 13.4
Web of Science (2016): Impact factor 12.712
Web of Science (2016): Indexed yes
BFI (2015): BFI-level 2
Scopus rating (2015): CiteScore 14.76
Web of Science (2015): Indexed yes
BFI (2014): BFI-level 2
Scopus rating (2014): CiteScore 14.04
Web of Science (2014): Impact factor 13.592
Web of Science (2014): Indexed yes
BFI (2013): BFI-level 2
Scopus rating (2013): CiteScore 14.23
Web of Science (2013): Impact factor 12.94
ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
BFI (2012): BFI-level 2
Scopus rating (2012): CiteScore 13.78
Web of Science (2012): Impact factor 13.025
ISI indexed (2012): ISI indexed yes
Web of Science (2012): Indexed yes
BFI (2011): BFI-level 2
Interlayer Trions in the MoS₂/WS₂ van der Waals Heterostructure

Electronic excitations in van der Waals heterostructures can have interlayer or intralayer character depending on the spatial localization of the involved charges (electrons and holes). In the case of neutral electron-hole pairs (excitons), both types of excitations have been explored theoretically and experimentally. In contrast, studies of charged trions have so far been limited to the intralayer type. Here we investigate the complete set of interlayer excitations in a MoS₂/WS₂ heterostructure using a novel ab initio method, which allows for a consistent treatment of both excitons and trions at the same theoretical footing. Our calculations predict the existence of bound interlayer trions below the neutral interlayer excitons. We obtain binding energies of 18/28 meV for the positive/negative interlayer trions with both electrons/holes located on the same layer. In contrast, a negligible binding energy is found for trions which have the two equally charged particles on different layers. Our results advance the understanding of electronic excitations in doped van der Waals heterostructures and their effect on the optical properties.

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Web of Science (2018): Indexed yes
BFI (2017): BFI-level 2
Scopus rating (2017): CiteScore 13.07
Web of Science (2017): Impact factor 12.08
Local Plasmon Engineering in Doped Graphene

Single-atom B or N substitutional doping in single-layer suspended graphene, realized by low-energy ion implantation, is shown to induce a dampening or enhancement of the characteristic interband $\pi$ plasmon of graphene through a high-resolution electron energy loss spectroscopy study using scanning transmission electron microscopy. A relative 16% decrease or 20% increase in the $\pi$ plasmon quality factor is attributed to the presence of a single substitutional B or N atom dopant, respectively. This modification is in both cases shown to be relatively localized, with data suggesting the
plasmonic response tailoring can no longer be detected within experimental uncertainties beyond a distance of approximately 1 nm from the dopant. Ab initio calculations confirm the trends observed experimentally. Our results directly confirm the possibility of tailoring the plasmonic properties of graphene in the ultraviolet waveband at the atomic scale, a crucial step in the quest for utilizing graphene’s properties toward the development of plasmonic and optoelectronic devices operating at ultraviolet frequencies.

**General information**

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Organisations: Department of Physics, Theoretical Atomic-scale Physics, Center for Nanostructured Graphene, SuperSTEM Laboratory, University of Leeds, Ömer Halisdemir University, Georg-August-Universität Göttingen, University of Limerick
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BFI (2017): BFI-level 2
Scopus rating (2017): CiteScore 14.29 SJR 7.203 SNIP 2.58
Web of Science (2017): Impact factor 13.709
Web of Science (2017): Indexed yes
BFI (2016): BFI-level 2
Scopus rating (2016): CiteScore 13.65 SJR 6.948 SNIP 2.604
Web of Science (2016): Impact factor 13.942
Web of Science (2016): Indexed yes
BFI (2015): BFI-level 2
Scopus rating (2015): CiteScore 13.55 SJR 6.712 SNIP 2.721
Web of Science (2015): Indexed yes
BFI (2014): BFI-level 2
Scopus rating (2014): CiteScore 12.49 SJR 5.981 SNIP 2.721
Web of Science (2014): Impact factor 12.881
Web of Science (2014): Indexed yes
BFI (2013): BFI-level 2
Scopus rating (2013): CiteScore 13.18 SJR 6.672 SNIP 2.735
Web of Science (2013): Impact factor 12.033
ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
BFI (2012): BFI-level 2
Scopus rating (2012): CiteScore 11.92 SJR 7.162 SNIP 2.685
Web of Science (2012): Impact factor 12.062
ISI indexed (2012): ISI indexed yes
Web of Science (2012): Indexed yes
Scopus rating (2011): CiteScore 11.05 SJR 6.282 SNIP 2.453
Web of Science (2011): Impact factor 11.421
ISI indexed (2011): ISI indexed yes
Web of Science (2011): Indexed yes
Scopus rating (2010): SJR 5.344 SNIP 2.069
Machine learning-based screening of complex molecules for polymer solar cells

Polymer solar cells admit numerous potential advantages including low energy payback time and scalable high-speed manufacturing, but the power conversion efficiency is currently lower than for their inorganic counterparts. In a Phenyl-C_{61}-Butyric-Acid-Methyl-Ester (PCBM)-based blended polymer solar cell, the optical gap of the polymer and the energetic alignment of the lowest unoccupied molecular orbital (LUMO) of the polymer and the PCBM are crucial for the device efficiency. Searching for new and better materials for polymer solar cells is a computationally costly affair using density functional theory (DFT) calculations. In this work, we propose a screening procedure using a simple string representation for a promising class of donor-acceptor polymers in conjunction with a grammar variational autoencoder. The model is trained on a dataset of 3989 monomers obtained from DFT calculations and is able to predict LUMO and the lowest optical transition energy for unseen molecules with mean absolute errors of 43 and 74 meV, respectively, without knowledge of the atomic positions. We demonstrate the merit of the model for generating new molecules with the desired LUMO and optical gap energies which increases the chance of finding suitable polymers by more than a factor of five in comparison to the randomised search used in gathering the training set.

General information
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Organisations: Department of Applied Mathematics and Computer Science, Cognitive Systems, Department of Energy Conversion and Storage, Atomic Scale Materials Modelling, Department of Physics, Theoretical Atomic-scale Physics
Contributors: Jørgensen, P. B., Mesta, M., Shil, S., García Lastra, J. M., Jacobsen, K. W., Thygesen, K. S., Schmidt, M. N.
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Web of Science (2018): Indexed yes
BFI (2017): BFI-level 2
Scopus rating (2017): CiteScore 2.5 SJR 1.252 SNIP 0.926
Web of Science (2017): Impact factor 2.843
Web of Science (2017): Indexed yes
BFI (2016): BFI-level 2
Scopus rating (2016): CiteScore 2.13 SJR 1.486 SNIP 0.964
Web of Science (2016): Impact factor 2.965
Web of Science (2016): Indexed yes
BFI (2015): BFI-level 2
Scopus rating (2015): CiteScore 1.98 SJR 1.255 SNIP 0.964
Web of Science (2015): Impact factor 2.894
Web of Science (2015): Indexed yes
Nano-imaging of intersubband transitions in van der Waals quantum wells

The science and applications of electronics and optoelectronics have been driven for decades by progress in the growth of semiconducting heterostructures. Many applications in the infrared and terahertz frequency range exploit transitions between quantized states in semiconductor quantum wells (intersubband transitions). However, current quantum well devices are limited in functionality and versatility by diffusive interfaces and the requirement of lattice-matched growth conditions. Here, we introduce the concept of intersubband transitions in van der Waals quantum wells and report their first experimental observation. Van der Waals quantum wells are naturally formed by two-dimensional materials and hold unexplored potential to overcome the aforementioned limitations they form atomically sharp interfaces and can easily be combined into heterostructures without lattice-matching restrictions. We employ near-field local probing to spectrally resolve intersubband transitions with a nanometre-scale spatial resolution and electrostatically control the absorption. This work enables the exploitation of intersubband transitions with unmatched design freedom and individual electronic and optical control suitable for photodetectors, light-emitting diodes and lasers.

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Scopus rating (2016): CiteScore 21.85 SJR 18.916 SNIP 7.649
Web of Science (2016): Impact factor 38.986
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BFI (2015): BFI-level 2
Scopus rating (2015): CiteScore 22.1 SJR 18.842 SNIP 8.019
Web of Science (2015): Impact factor 35.267
Web of Science (2015): Indexed yes
BFI (2014): BFI-level 2
Scopus rating (2014): CiteScore 21.76 SJR 17.177 SNIP 8.047
Web of Science (2014): Impact factor 34.048
Web of Science (2014): Indexed yes
BFI (2013): BFI-level 2
Scopus rating (2013): CiteScore 21.94 SJR 16.688 SNIP 7.784
Web of Science (2013): Impact factor 33.265
ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
BFI (2012): BFI-level 2
Scopus rating (2012): CiteScore 17.55 SJR 15.706 SNIP 7.569
Stacked Janus Device Concepts: Abrupt pn-Junctions and Cross-Plane Channels

Janus transition metal dichalcogenides with a built-in structural cross-plane (cp) asymmetry have recently emerged as a new class of two-dimensional materials with a large cp dipole. Using first-principles calculations, and a tailored transport method, we demonstrate that stacking graphene and MoSSe Janus structures result in record high homogeneous doping of graphene and abrupt, atomically thin, cross-plane pn-junctions. We show how graphene in contrast to metals can act as electrodes to Janus stacks without screening the cp dipole and predict a large photocurrent response dominated by a cp transport channel in a few-layer stacked device. The photocurrent is above that of a corresponding thin-film silicon device illustrating the great potential of Janus stacks, for example, in photovoltaic devices.

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Organisations: Theoretical Nanoelectronics, Department of Micro- and Nanotechnology, Center for Nanostructured Graphene, Department of Physics, Theoretical Atomic-scale Physics, Synopsys Denmark ApS
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Web of Science (2018): Indexed yes
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Scopus rating (2017): CiteScore 13.07
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We introduce the Computational 2D Materials Database (C2DB), which organises a variety of structural, thermodynamic, elastic, electronic, magnetic, and optical properties of around 1500 two-dimensional materials distributed over more than 30 different crystal structures. Material properties are systematically calculated by state-of-the-art density functional theory and many-body perturbation theory (and the Bethe–Salpeter equation for ~250 materials) following a semi-automated workflow for maximal consistency and transparency. The C2DB is fully open and can be browsed online (http://c2db.fysik.dtu.dk) or downloaded in its entirety. In this paper, we describe the workflow behind the database,
present an overview of the properties and materials currently available, and explore trends and correlations in the data. Moreover, we identify a large number of new potentially synthesizable 2D materials with interesting properties targeting applications within spintronics, (opto-)electronics, and plasmonics. The C2DB offers a comprehensive and easily accessible overview of the rapidly expanding family of 2D materials and forms an ideal platform for computational modeling and design of new 2D materials and van der Waals heterostructures.

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Organisations: Department of Physics, Theoretical Atomic-scale Physics, Center for Nanostructured Graphene, Neutrons and X-rays for Materials Physics, Plasma Physics and Fusion Energy
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Scopus rating (2017): CiteScore 6.05 SJR 2.813 SNIP 1.072
Web of Science (2017): Impact factor 7.042
Web of Science (2017): Indexed yes
BFI (2016): BFI-level 1
Scopus rating (2016): CiteScore 4.26 SJR 2.314 SNIP 0.915
Web of Science (2016): Impact factor 6.937
Web of Science (2016): Indexed yes
BFI (2015): BFI-level 1
Scopus rating (2015): CiteScore 5.89 SJR 4.602 SNIP 1.009
BFI (2014): BFI-level 1
BFI (2013): BFI-level 1
Original language: English
Keywords: Ab initio calculation, Opto-electroni properties, Database, Materials discovery, Materials design, 2D materials, Many-body perturbation theory
Electronic versions:
Haastrup_2018_2D_Mater._5_042002.pdf
DOIs:
10.1088/2053-1583/aacfc1

**Bibliographical note**
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Source-ID: 2436268867
Research output: Research - peer-review › Journal article – Annual report year: 2018

**Unraveling the not-so-large trion binding energy in monolayer black phosphorus**
Black phosphorus (bP) is a promising two-dimensional (2D) material for opto-electronic applications. Strongly bound excitons with binding energies up to 0.3 eV and remarkably large trion binding energies up to 100 meV have been observed for supported monolayer bP. Surprisingly, this trion binding energy is significantly larger than those found in other 2D materials (e.g. about 30 meV in transition metal dichalcogenides). This has previously been ascribed to the quasi-1D nature of bP. In this work we show, using first principles calculations, that the trion binding energy of bP is indeed large (80 meV) when referenced to the lowest bright exciton but only 30 meV when its energy is measured relative
to the lowest dark exciton. Our analysis thus shows that the trion binding energy in bP is not larger than in other 2D materials, and the previous conclusions have to be understood incorporating the large splitting between the dark and bright excitons in bP. We also explore the effect of substrate and in-plane strain of the exciton and trion binding energies and show that these effects do not change the main conclusions. Our results correct the misconception that trion binding energies in monolayer bP are particularly large due to its quasi-1D structure and contribute to the establishment of more a detailed understanding of optical properties of atomically thin semiconductors.

**General information**

State: Published

Organisations: Department of Physics, Theoretical Atomic-scale Physics, Center for Nanostructured Graphene

Contributors: Deilmann, T., Thygesen, K. S.

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Web of Science (2018): Indexed yes

BFI (2017): BFI-level 1

Scopus rating (2017): CiteScore 6.05 SJR 2.813 SNIP 1.072

Web of Science (2017): Impact factor 7.042

Web of Science (2017): Indexed yes

BFI (2016): BFI-level 1

Scopus rating (2016): CiteScore 4.26 SJR 2.314 SNIP 0.915

Web of Science (2016): Impact factor 6.937

Web of Science (2016): Indexed yes

BFI (2015): BFI-level 1

Scopus rating (2015): CiteScore 5.89 SJR 4.602 SNIP 1.009


BFI (2014): BFI-level 1

BFI (2013): BFI-level 1

Original language: English

Keywords: Black phosphorous, Trion, Optical spectra, Many-body perturbation theory

DOIs:

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Research output: Research - peer-review; Letter – Annual report year: 2018

**Band structure engineered layered metals for low-loss plasmonics**

Plasmonics currently faces the problem of seemingly inevitable optical losses occurring in the metallic components that challenges the implementation of essentially any application. In this work, we show that Ohmic losses are reduced in certain layered metals, such as the transition metal dichalcogenide TaS2, due to an extraordinarily small density of states for scattering in the near-IR originating from their special electronic band structure. On the basis of this observation, we propose a new class of band structure engineered van der Waals layered metals composed of hexagonal transition metal chalcogenide-halide layers with greatly suppressed intrinsic losses. Using first-principles calculations, we show that the suppression of optical losses lead to improved performance for thin-film waveguiding and transformation optics.

**General information**

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Organisations: Center for Nanostructured Graphene, Department of Physics, Theoretical Atomic-scale Physics

Contributors: Gjerding, M. N., Pandey, M., Thygesen, K. S.

Number of pages: 8
Band structure engineering in van der Waals heterostructures via dielectric screening: the GΔW method

The idea of combining different two-dimensional (2D) crystals in van der Waals heterostructures (vdWHs) has led to a new paradigm for band structure engineering with atomic precision. Due to the weak interlayer couplings, the band structures of the individual 2D crystals are largely preserved upon formation of the heterostructure. However, regardless of the details of the interlayer hybridisation, the size of the 2D crystal band gaps are always reduced due to the enhanced dielectric screening provided by the surrounding layers. The effect can be significant (on the order of electron volts) but its precise magnitude is non-trivial to predict because of the non-local nature of the screening in quasi-2D crystals. Moreover,
the effect is not captured by effective single-particle methods such as density functional theory. Here we present an efficient and general method for calculating the band gap renormalization of a 2D material embedded in an arbitrary vdWH. The method evaluates the change in the GW self-energy of the 2D material from the change in the screened Coulomb interaction. The latter is obtained using the quantum-electrostatic heterostructure (QEH) model. We benchmark the GΔW method against full first-principles GW calculations and use it to unravel the importance of screening-induced band structure renormalisation in various vdWHs. A main result is the observation that the size of the band gap reduction of a given 2D material when inserted into a heterostructure scales inversely with the polarisability of the 2D material. Our work demonstrates that dielectric engineering via van der Waals heterostructuring represents a promising strategy for tailoring the band structure of 2D materials.
materials are also discussed. A combined quantum/classical method is introduced and used throughout to account for dielectric screening and self-energy effects from substrates and van der Waals heterostructures including the difficult case of non-matching lattices.

**General information**

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Organisations: Center for Nanostructured Graphene, Department of Physics, Theoretical Atomic-scale Physics

Contributors: Thygesen, K. S.

Number of pages: 28

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Scopus rating (2017): CiteScore 6.05 SJR 2.813 SNIP 1.072

Web of Science (2017): Impact factor 7.042

Web of Science (2017): Indexed yes

BFI (2016): BFI-level 1

Scopus rating (2016): CiteScore 4.26 SJR 2.314 SNIP 0.915

Web of Science (2016): Impact factor 6.937

Web of Science (2016): Indexed yes

BFI (2015): BFI-level 1

Scopus rating (2015): CiteScore 5.89 SJR 4.602 SNIP 1.009


BFI (2014): BFI-level 1

BFI (2013): BFI-level 1

Original language: English

Keywords: 2D materials, Excitons, Plasmons, Density functional theory, GW method, van der Waals heterostructures

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Research output: Research - peer-review > Journal article – Annual report year: 2017

**Computational High-throughput Screening for Solar Energy Materials**

**General information**

State: Published

Organisations: Department of Energy Conversion and Storage, Department of Physics, Atomic Scale Materials Modelling, Theoretical Atomic-scale Physics

Contributors: Castelli, I. E., Thygesen, K. S., Jacobsen, K. W.

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Dark excitations in monolayer transition metal dichalcogenides

Monolayers of transition metal dichalcogenides (TMDCs) possess unique optoelectronic properties, including strongly bound excitons and trions. To date, most studies have focused on optically active excitations, but recent experiments have highlighted the existence of dark states, which are equally important in many respects. Here, we use ab initio many-body calculations to unravel the nature of the dark excitations in monolayer MoSe2, MoS2, WSe2, and WS(2). Our results show that all these monolayer TMDCs host dark states as their lowest neutral and charged excitations. We further show that dark excitons possess larger binding energies than their bright counterparts while the opposite holds for trions.
Effect of edge plasmons on the optical properties of MoS$_2$ monolayer flakes

Finite MoS$_2$ nanoparticles are known to support metallic edge states that are responsible for their catalytic activity. In this work we employ time-dependent density-functional theory (TDDFT) to study the influence of such edge states on the optical properties of triangular MoS$_2$ monolayer flakes. We find that the edge states support collective plasmon-like excitations that couple strongly to the optical field leading to pronounced absorption peaks below the onset of interband transitions on the basal plane. Additionally, structural relaxation of the flakes can significantly distort the edge states. Thus, we observe that while an evenly-spaced edge configuration supports one-dimensional (1D) plasmon modes similar to those of an ideal 1D electron gas, the relaxed structures show mixed plasmon and single-electron excitations in the low-energy response. Our findings illustrate the sensitivity of the optical response of MoS$_2$ nanostructures to the details of the edge configuration.

General information
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Organisations: Department of Physics, Theoretical Atomic-scale Physics, Center for Nanostructured Graphene, Aalto University
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Peer-reviewed: Yes

Publication information
Journal: Physical Review B
Volume: 96
Issue number: 15
Article number: 155407
ISSN (Print): 2469-9950
Ratings:
BFI (2019): BFI-level 1
Grid-Based Projector Augmented Wave (GPAW) Implementation of Quantum Mechanics/Molecular Mechanics (QM/MM) Electrostatic Embedding and Application to a Solvated Diplatinum Complex

A multiscale density functional theory-quantum mechanics/molecular mechanics (DFT-QM/MM) scheme is presented, based on an efficient electrostatic coupling between the electronic density obtained from a grid-based projector augmented wave (GPAW) implementation of density functional theory and a classical potential energy function. The scheme is implemented in a general fashion and can be used with various choices for the descriptions of the QM or MM regions. Tests on H2O clusters, ranging from dimer to decamer show that no systematic energy errors are introduced by the coupling that exceeds the differences in the QM and MM descriptions. Over 1 ns of liquid water, Born-Oppenheimer QM/MM molecular dynamics (MD) are sampled combining 10 parallel simulations, showing consistent liquid water structure over the QM/MM border. The method is applied in extensive parallel MD simulations of an aqueous solution of the diplatinum [Pt2(P2O5H2)4]- complex (PtPOP), spanning a total time period of roughly half a nanosecond. An average Pt-Pt distance deviating only 0.01 Å from experimental results, and a ground-state Pt-Pt oscillation frequency deviating by...
Interlayer Excitons and Band Alignment in MoS$_2$/hBN/WSe$_2$ van der Waals Heterostructures

van der Waals heterostructures (vdWH) are ideal systems for exploring light-matter interactions at the atomic scale. In particular, structures with a type-II band alignment can yield detailed insight into carrier-photon conversion processes, which are central to, for example, solar cells and light-emitting diodes. An important first step in describing such processes is to obtain the energies of the interlayer exciton states existing at the interface. Here we present a general first-principles method to compute the electronic quasi-particle (QP) band structure and excitonic binding energies of incommensurate vdWHs. The method combines our quantum electrostatic heterostructure (QEH) model for obtaining the dielectric function with the many-body GW approximation and a generalized 2D Mott-Wannier exciton model. We calculate the level alignment together with intra- and interlayer exciton binding energies of bilayer MoS$_2$/WSe$_2$ with and without intercalated hBN layers, finding excellent agreement with experimental photoluminescence spectra. A comparison to density functional
theory calculations demonstrates the crucial role of self-energy and electron-hole interaction effects.
Layered van der Waals crystals with hyperbolic light dispersion

Compared to artificially structured hyperbolic metamaterials, whose performance is limited by the finite size of the metallic components, the sparse number of naturally hyperbolic materials recently discovered are promising candidates for the next generation of hyperbolic materials. Using first-principles calculations, we extend the number of known naturally hyperbolic materials to the broad class of layered transition metal dichalcogenides (TMDs). The diverse electronic properties of the transition metal dichalcogenides result in a large variation of the hyperbolic frequency regimes ranging from the near-infrared to the ultraviolet. Combined with the emerging field of van der Waals heterostructuring, we demonstrate how the hyperbolic properties can be further controlled by stacking different two-dimensional crystals opening new perspectives for atomic-scale design of photonic metamaterials. As an application, we identify candidates for Purcell factor control of emission from diamond nitrogen-vacancy centers. Natural hyperbolic materials retain the peculiar optical properties of traditional metamaterials whilst not requiring artificial structuring. Here, the authors perform a theoretical screening of a large class of natural materials with hyperbolic dispersion among the family of layered transition metal dichalcogenides.
Probing the local nature of excitons and plasmons in few-layer MoS$_2$

Excitons and plasmons are the two most fundamental types of collective electronic excitations occurring in solids. Traditionally, they have been studied separately using bulk techniques that probe their average energetic structure over large spatial regions. However, as the dimensions of materials and devices continue to shrink, it becomes crucial to understand how these excitations depend on local variations in the crystal- and chemical structure on the atomic scale. Here we use monochromated low-loss scanning-transmission-electron-microscopy electron-energy-loss (LL-STEM-EEL) spectroscopy, providing the best simultaneous energy and spatial resolution achieved to-date to unravel the full set of electronic excitations in few-layer MoS$_2$ nanosheets over a wide energy range. Using first-principles many-body calculations we confirm the excitonic nature of the peaks at ~2eV and ~3eV in the experimental EEL spectrum and the plasmonic nature of higher energy-loss peaks. We also rationalise the non-trivial dependence of the EEL spectrum on beam and sample geometry such as the number of atomic layers and distance to steps and edges. Moreover, we show that the excitonic features are dominated by the long wavelength (q=0) components of the probing field, while the plasmonic features are sensitive to a much broader range of q-vectors, indicating a qualitative difference in the spatial character of the two types of collective excitations. Our work provides a template protocol for mapping the local nature of electronic excitations that open new possibilities for studying photo-absorption and energy transfer processes on a nanometer scale.

General information
State: Published
Organisations: Center for Nanostructured Graphene, Department of Physics, Theoretical Atomic-scale Physics, Trinity College Dublin, SuperSTEM Laboratory, Weizmann Institute of Science
Number of pages: 9
Publication date: 2017
Peer-reviewed: Yes
Simple vertex correction improves GW band energies of bulk and two-dimensional crystals

The GW self-energy method has long been recognized as the gold standard for quasiparticle (QP) calculations of solids in spite of the fact that the neglect of vertex corrections and the use of a density-functional theory starting point lack rigorous justification. In this work we remedy this situation by including a simple vertex correction that is consistent with a local-density approximation starting point. We analyze the effect of the self-energy by splitting it into short-range and long-range terms which are shown to govern, respectively, the center and size of the band gap. The vertex mainly improves the short-range correlations and therefore has a small effect on the band gap, while it shifts the band gap center up in energy by around 0.5 eV, in good agreement with experiments. Our analysis also explains how the relative importance of short- and long-range interactions in structures of different dimensionality is reflected in their QP energies. Inclusion of the vertex comes at practically no extra computational cost and even improves the basis set convergence compared to GW. Taken together, the method provides an efficient and rigorous improvement over the GW approximation.

General information
State: Published
Organisations: Department of Physics, Theoretical Atomic-scale Physics
Contributors: Schmidt, P. S., Patrick, C. E., Thygesen, K. S.
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Publication date: 2017
Peer-reviewed: Yes
The absence of inversion symmetry leads to a strong spin-orbit splitting of the upper valence band of semiconducting single-layer transition-metal dichalcogenides such as MoS$_2$ or WS$_2$. This permits a direct comparison of the electron-phonon coupling strength in states that only differ by their spin. Here, the electron-phonon coupling in the valence band maximum of single-layer WS$_2$ is studied by first-principles calculations and angle-resolved photoemission. The coupling strength is found to be drastically different for the two spin-split branches, with calculated values of $\lambda_{K}=0.0021$ and 0.40 for the upper and lower spin-split valence band of the freestanding layer, respectively. This difference is somewhat reduced when including scattering processes involving the Au(111) substrate present in the experiment but it remains...
significant, in good agreement with the experimental results.

**General information**
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Organisations: Department of Physics, Theoretical Atomic-scale Physics, Aarhus University, University of Trieste, Sincrotrone Trieste, Consiglio Nazionale delle Ricerche
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- Web of Science (2019): Indexed yes
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- Web of Science (2018): Indexed yes
- BFI (2017): BFI-level 2
- Scopus rating (2017): CiteScore 3.34 SJR 1.604 SNIP 1.04
- Web of Science (2017): Impact factor 3.813
- Web of Science (2017): Indexed yes
- Scopus rating (2016): CiteScore 3.16 SJR 2.339 SNIP 1.151
- Web of Science (2016): Impact factor 3.836
- Web of Science (2016): Indexed yes
- Scopus rating (2015): CiteScore 2.8 SJR 2.377 SNIP 1.13
- Web of Science (2015): Indexed yes
- Scopus rating (2014): CiteScore 3.3 SJR 2.762 SNIP 1.316
- Web of Science (2014): Impact factor 3.736
- Web of Science (2014): Indexed yes
- Scopus rating (2013): CiteScore 3.55 SJR 2.813 SNIP 1.326
- Web of Science (2013): Impact factor 3.664
- ISI indexed (2013): ISI indexed yes
- Web of Science (2013): Indexed yes
- Scopus rating (2012): CiteScore 3.57 SJR 3.173 SNIP 1.378
- Web of Science (2012): Impact factor 3.767
- ISI indexed (2012): ISI indexed yes
- Web of Science (2012): Indexed yes
- Scopus rating (2011): CiteScore 3.61 SJR 3.326 SNIP 1.423
- Web of Science (2011): Impact factor 3.691
- ISI indexed (2011): ISI indexed yes
- Web of Science (2011): Indexed yes
- Scopus rating (2010): SJR 3.318 SNIP 1.447
- Web of Science (2010): Impact factor 3.774
- Web of Science (2010): Indexed yes
- Web of Science (2009): Indexed yes
- Scopus rating (2008): SJR 2.923 SNIP 1.516
- Web of Science (2008): Indexed yes
- Scopus rating (2007): SJR 2.892 SNIP 1.588
Strong Plasmon-Phonon Splitting and Hybridization in 2D Materials Revealed through a Self-Energy Approach

We reveal new aspects of the interaction between plasmons and phonons in 2D materials that go beyond a mere shift and increase in plasmon width due to coupling to either intrinsic vibrational modes of the material or phonons in a supporting substrate. More precisely, we predict strong plasmon splitting due to this coupling, resulting in a characteristic avoided crossing scheme. We base our results on a computationally efficient approach consisting in including many-body interactions through the electron self-energy. We specify this formalism for a description of plasmons based upon a tight-binding electron Hamiltonian combined with the random-phase approximation. This approach is valid provided vertex corrections can be neglected, as is the case in conventional plasmon-supporting metals and Dirac-Fermion systems. We illustrate our method by evaluating plasmonic spectra of doped graphene nanotriangles with varied size, where we predict remarkable peak splittings and other radical modifications in the spectra due to plasmon interactions with intrinsic optical phonons. Our method is equally applicable to other 2D materials and provides a simple approach for investigating coupling of plasmons to phonons, excitons, and other excitations in hybrid thin nanostructures.
Sulfide perovskites for solar energy conversion applications: computational screening and synthesis of the selected compound LaYS$_3$

One of the key challenges in photovoltaic water splitting is to identify efficient semiconductors with band gaps of the order of $\sim$2 eV to operate as the large-band-gap component in water splitting tandem devices. Here, we address this challenge by extensive computational screening of ternary sulfides followed by synthesis and confirmation of the properties of one of the most promising materials. The screening focuses on materials with ABS3 composition taking both perovskite and non-perovskite structures into consideration, and the material selection is based on descriptors for thermodynamic stability, light absorption, charge mobility, and defect tolerance. One of the most promising candidates identified is LaYS$_3$. This material was synthesized directly in thin-film form demonstrating its stability, crystal structure, light absorption, and strong photoluminescence. These data confirms its potential applicability in tandem photoelectrochemical devices for hydrogen production.

General information
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Publication information
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Sustainable solar fuels and electricity through discovery and prototyping of new materials

**General information**

- **State:** Published
- **Organisations:** Department of Physics, Experimental Surface and Nanomaterials Physics, Silicon Microtechnology, Theoretical Atomic-scale Physics, Department of Micro- and Nanotechnology
- **Contributors:** Crovetto, A., Kuhar, K., Pandey, M., Thygesen, K. S., Jacobsen, K. W., Hansen, O., Seger, B., Vesborg, P. C. K., Chorkendorff, I.
- **Number of pages:** 1
- **Publication date:** 2017

**Sustainable solar fuels and electricity through discovery and prototyping of new materials**

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Research output: Research - peer-review → Journal article – Annual report year: 2017
The Atomic Simulation Environment - A Python library for working with atoms

The Atomic Simulation Environment (ASE) is a software package written in the Python programming language with the aim of setting up, steering, and analyzing atomistic simulations. In ASE, tasks are fully scripted in Python. The powerful syntax of Python combined with the NumPy array library make it possible to perform very complex simulation tasks. For example, a sequence of calculations may be performed with the use of a simple "for-loop" construction. Calculations of energy, forces, stresses and other quantities are performed through interfaces to many external electronic structure codes or force fields using a uniform interface. On top of this calculator interface, ASE provides modules for performing many standard simulation tasks such as structure optimization, molecular dynamics, handling of constraints and performing nudged elastic band calculations.

General information
State: Published
Organisations: Department of Physics, Theoretical Atomic-scale Physics, Department of Energy Conversion and Storage, Atomic Scale Materials Modelling, Department of Micro- and Nanotechnology, Theoretical Nanotechnology, University of Barcelona, University of Copenhagen, Malmö University, SINTEF, Aarhus University, Brown University, University of Wisconsin-Madison, University of Warwick, Carnegie Mellon University, Purdue University, Siminn, Karlsruhe Institute of Technology, Swiss Federal Institute of Technology Zurich, University of Freiburg
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Publication information
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Web of Science (2019): Indexed yes
BFI (2018): BFI-level 1
Web of Science (2018): Indexed yes
BFI (2017): BFI-level 1
Scopus rating (2017): CiteScore 2.37 SJR 0.875 SNIP 0.921
Web of Science (2017): Impact factor 2.617
Web of Science (2017): Indexed yes
BFI (2016): BFI-level 1
Scopus rating (2016): CiteScore 1.89 SJR 1.553 SNIP 0.91
Web of Science (2016): Impact factor 2.678
Web of Science (2016): Indexed yes
BFI (2015): BFI-level 1
Scopus rating (2015): CiteScore 1.65 SJR 1.043 SNIP 0.889
Web of Science (2015): Impact factor 2.209
Web of Science (2015): Indexed yes
BFI (2014): BFI-level 1
Scopus rating (2014): CiteScore 1.99 SJR 1.284 SNIP 0.987
Web of Science (2014): Impact factor 2.346
Web of Science (2014): Indexed yes
BFI (2013): BFI-level 1
We use density functional theory calculations to explore different polymorphs of a new class of 2D materials commonly known as MXenes, which are primarily carbides and nitrides of transition metals. The stability of the $M_2X$, $M_3X_2$, and $M_4X_3$ polymorphs in their bare and functionalized forms is assessed via the calculated standard heat of formation. We find that most of the MXenes are metallic, and we investigate their performance as electrocatalysts for the hydrogen evolution reaction (HER) using the free energy of hydrogen adsorption at equilibrium coverage as an activity descriptor. For a given
type of metal, we find that the hydrogen adsorption energy can vary by up to 0.5 eV depending on the number of metal layers in the structure, suggesting that the catalytic activity of MXenes can be tuned by controlling the layer thickness. On the basis of a combined stability and activity analysis of 72 different MXenes, we identify several new promising nonprecious HER electrocatalysts.
We explore the possibility of modulating the electronic band edges of the transition metal dichalcogenides (TMD) via alloying of different semiconductors within the same group (intra-group alloying). The stability of the ordered alloys is assessed from the calculated mixing enthalpy which is found to be close to zero for several alloys and below 20 meV/atom for all the alloys. We explore to what extent the electronic properties like the band gap and band edge positions of the alloy can be evaluated by taking the weighted average of the corresponding properties of the pristine systems. In general, this approach works well with the only exception being Cr containing compounds. Because the calculated properties of the alloys are very similar to the weighted averages, we expect that the trends observed for the ordered alloys will also hold for more realistic disordered alloys.

**General information**

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Organisations: Center for Atomic-scale Materials Design, Department of Physics, Center for Nanostructured Graphene
Contributors: Pandey, M., Jacobsen, K. W., Thygesen, K. S.
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Peer-reviewed: Yes

**Publication information**

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Volume: 120
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ISSN (Print): 1932-7447
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Web of Science (2019): Indexed yes
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Web of Science (2018): Indexed yes
BFI (2017): BFI-level 1
Scopus rating (2017): CiteScore 4.58 SJR 2.135 SNIP 1.147
Web of Science (2017): Impact factor 4.484
Web of Science (2017): Indexed yes
BFI (2016): BFI-level 1
Band Gap Tuning and Defect Tolerance of Atomically Thin Two-Dimensional Organic-Inorganic Halide Perovskites

Organic–inorganic halide perovskites have proven highly successful for photovoltaics but suffer from low stability, which deteriorates their performance over time. Recent experiments have demonstrated that low dimensional phases of the hybrid perovskites may exhibit improved stability. Here we report first-principles calculations for isolated monolayers of the
organometallic halide perovskites \((\text{C}_4\text{H}_9\text{NH}_3)_2\text{MX}_2\text{Y}_2\), where \(M = \text{Pb, Ge, Sn}\) and \(X,Y = \text{Cl, Br, I}\). The band gaps computed using the GLLB-SC functional are found to be in excellent agreement with experimental photoluminescence data for the already synthesized perovskites. Finally, we study the effect of different defects on the band structure. We find that the most common defects only introduce shallow or no states in the band gap, indicating that these atomically thin 2D perovskites are likely to be defect tolerant.
Defect-Tolerant Monolayer Transition Metal Dichalcogenides

Localized electronic states formed inside the band gap of a semiconductor due to crystal defects can be detrimental to the material's optoelectronic properties. Semiconductors with a lower tendency to form defect induced deep gap states are termed defect-tolerant. Here we provide a systematic first-principles investigation of defect tolerance in 29 monolayer transition metal dichalcogenides (TMDs) of interest for nanoscale optoelectronics. We find that the TMDs based on group VI and X metals form deep gap states upon creation of a chalcogen (S, Se, Te) vacancy, while the TMDs based on group IV metals form only shallow defect levels and are thus predicted to be defect-tolerant. Interestingly, all the defect sensitive TMDs have valence and conduction bands with a very similar orbital composition. This indicates a bonding/antibonding nature of the gap, which in turn suggests that dangling bonds will fall inside the gap. These ideas are made quantitative by introducing a descriptor that measures the degree of similarity of the conduction and valence band manifolds. Finally, the study is generalized to nonpolar nanoribbons of the TMDs where we find that only the defect sensitive materials form edge states within the band gap.
Efficient many-body calculations for two-dimensional materials using exact limits for the screened potential: Band gaps of MoS2, h-BN, and phosphorene

Calculating the quasiparticle (QP) band structure of two-dimensional (2D) materials within the GW self-energy approximation has proven to be a rather demanding computational task. The main reason is the strong q dependence of the 2D dielectric function around q = 0 that calls for a much denser sampling of the Brillouin zone (BZ) than is necessary for similar three-dimensional solids. Here, we use an analytical expression for the small q limit of the 2D response function to perform the BZ integral over the critical region around q = 0. This drastically reduces the requirements on the q-point mesh and implies a significant computational speedup. For example, in the case of monolayer MoS2, convergence of the G0W0 band gap to within similar to 0.1 eV is achieved with 12 x 12 q points rather than the 36 x 36 mesh required with discrete BZ sampling techniques. We perform a critical assessment of the band gap of the three prototypical 2D semiconductors, MoS2, h-BN, and phosphorene, including the effect of self-consistency at the G0W0 level. The method is
implemented in the open source code GPAW.

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Scopus rating (2008): SJR 2.923 SNIP 1.516
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Exciton ionization in multilayer transition-metal dichalcogenides

Photodetectors and solar cells based on materials with strongly bound excitons rely crucially on field-assisted exciton ionization. We study the ionization process in multilayer transition-metal dichalcogenides (TMDs) within the Mott-Wannier model incorporating fully the pronounced anisotropy of these materials. Using complex scaling, we show that the field-dependence of the ionization process is strongly dependent on orientation. Also, we find that direct and indirect excitons behave qualitatively differently as a result of opposite effective anisotropy of these states. Based on first-principles material parameters, an analysis of several important TMDs reveals WSe$_2$ and MoSe$_2$ to be superior for applications relying on ionization of direct and indirect excitons, respectively.

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Organisations: Center for Atomic-scale Materials Design, Center for Nanostructured Graphene, Department of Physics, Aalborg University, University of Delaware
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Excitons in van der Waals Heterostructures: A theoretical study

Van der Waals heterostructures (vdWHs) represent a novel and largely unexplored class of materials. Since 2013, when Geim and Grigorieva first conceived the stacking of 2D (two-dimensional) materials to create artificial layered structures with tailored properties, a number of promising (opto)electronics devices, e.g. light emitting diodes, solar cells, ultra-fast photodetectors, transistors etc., have been successfully fabricated. It is well established that for isolated 2D semiconductors and vdWHs the optical response is governed by excitonic effects. While it is understood that the reduced amount of electronic screening in freestanding 2D materials is the main origin of extraordinarily strongly bound excitons, a theoretical understanding of excitonic effects and of how the electronic screening is affected for the more complex case of multi-layer structures is still lacking due to the computational limitations of standard ab-initio methods.

In this thesis first-principles models that overcome the limitations of standard ab-initio techniques are developed for the description of dielectric, electronic and excitonic properties in isolated 2D materials and vdWHs. The main contribution is a multi-scale method that seemingly connects the excitonic effects in the monolayer limit to the more challenging case of multi-layered structures. The method is based on the analogy between vdWHs and the popular construction toy Lego. This analogy is much deeper than one would first expect: it is possible to predict the dielectric properties of a vdWH from the dielectric functions of the individual 2D layers, which represent the dielectric genome of the heterostructure. From the vdWH dielectric properties one evaluates the screened interaction between the electron and hole forming the exciton which can then be used in a generalized hydrogenic model to compute exciton binding energies in isolated, supported, or encapsulated 2D semiconductors. The non-locality of the dielectric screening is inherently included in our method and we can successfully describe the non-hydrogenic Rydberg series of low-dimensional systems. This multi-scale method also proves successful when combined with many-body perturbation techniques for accurate prediction of electronic band structure or with complex scaling techniques for exciton dissociation rates in vdWHs. The validity of our techniques is demonstrated through numerous comparison to experimental results. Ultimately this thesis puts forth a first-principles methodology that allows us to address scientific questions that are beyond the capability of existing state of the art techniques and enables 2D materials researcher to predict and design dielectric, electronic and excitonic properties of general vdWHs.

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Finite Bias Calculations to Model Interface Dipoles in Electrochemical Cells at the Atomic Scale

The structure of an electrochemical interface is not determined by any external electrostatic field, but rather by external chemical potentials. This paper demonstrates that the electric double layer should be understood fundamentally as an internal electric field set up by the atomic structure to satisfy the thermodynamic constraints imposed by the environment. This is captured by the generalized computational hydrogen electrode model, which enables us to make efficient first-principles calculations of atomic scale properties of the electrochemical interface.

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Organisations: Department of Energy Conversion and Storage, Department of Physics, Center for Nanostructured Graphene, University of Copenhagen
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First Principles Calculations of Electronic Excitations in 2D Materials

Since the first reported synthesis of graphene - an atomically thin carbon material - in 2004 there has been a surge of research in discovering other novel two-dimensional materials. The reason is clear: two-dimensional materials are thought to be able to lead to new fast and low-power ultra-thin electronics and high efficiency solar cells. Contrary to many other nano-materials, methods for large scale fabrication and patterning have already been demonstrated and the first real technological applications have already be showcased. Still the technology is very young and the number of well-studied 2D materials are few. However as the list of 2D materials is growing it is necessary to investigate their fundamental structural, electronic and optical properties. These are determined by the atomic and electronic structure of the materials that can quite accurately predicted by computational quantum mechanics methods.

One of these methods, Density Functional Theory (DFT), has been very successful at determining structural properties of 2D materials. It is however well-known that it less accurate when it comes to predicting the energy levels of excited states that are important in order to determine electronic transport, optical and chemical properties. On the other hand it has shown to be a great starting point for a systematic perturbation theory approach to obtain the so-called quasiparticle spectrum. In the GW approximation one considers the potential from a charged excitation as if it is being screened by the electrons in the material. This method has been very successful for calculating quasiparticle energies of bulk materials but results have been more varying for 2D materials. The reason is that the 2D confined electrons are less able to screen the added charge and some of the numerical methods that are efficient for bulk systems become invalid.

In this thesis I describe the study of a set of novel 2D materials and establish their electronic and optical properties using DFT and the GW approximation while taking the reduced screening properly into account as well as taking regard to other numerical problems that have often been neglected. Secondly I show how one can efficiently take the 2D nature into account in the GW approximation and thereby make future calculations require much less computational resources.

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Hubbard-U corrected Hamiltonians for non-self-consistent random-phase approximation total-energy calculations: A study of ZnS, TiO2, and NiO
In non-self-consistent calculations of the total energy within the random-phase approximation (RPA) for electronic correlation, it is necessary to choose a single-particle Hamiltonian whose solutions are used to construct the electronic density and noninteracting response function. Here we investigate the effect of including a Hubbard-U term in this single-particle Hamiltonian, to better describe the on-site correlation of 3d electrons in the transitionmetal compounds ZnS, TiO2, and NiO. We find that the RPA lattice constants are essentially independent of U, despite large changes in the underlying electronic structure. We further demonstrate that the non-selfconsistent RPA total energies of these materials have minima at nonzero U. Our RPA calculations find the rutile phase of TiO2 to be more stable than anatase independent of U, a result which is consistent with experiments and qualitatively different from that found from calculations employing U-corrected (semi)local functionals. However we also find that the+U term cannot be used to correct the RPA’s poor description of the heat of formation of NiO.

General information
Limitations of effective medium theory in multilayer graphite/hBN heterostructures

We apply effective medium theory (EMT) to metamaterials consisting of a varying number of consecutive sheets of graphene and hexagonal boron nitride, and compare this with a full calculation of the permittivity and the reflection based on the tight binding method and the transfer matrix method in order to study the convergence to EMT. We find that convergence is reached for both in-plane and out-of-plane directions already for five sheets but that for ≈30 sheets multiple reflection effects cause the reflection spectrum to differ from EMT. We show that modes that are evanescent in air are extremely sensitive to the electronic details of the sheets near the structure boundary and that EMT estimates poorly the reflection of these modes, causing an overestimation of the Purcell factor. Finally, we offer an improved EMT, which gives far better convergence in the low-energy regime.
Making the most of materials computations

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Organisations: Center for Atomic-scale Materials Design, Center for Nanostructured Graphene, Department of Physics
Simple Screened Hydrogen Model of Excitons in Two-Dimensional Materials

We present a generalized hydrogen model for the binding energies (EB) and radii of excitons in two-dimensional (2D) materials that sheds light on the fundamental differences between excitons in two and three dimensions. In contrast to the well-known hydrogen model of three-dimensional (3D) excitons, the description of 2D excitons is complicated by the fact that the screening cannot be assumed to be local. We show that one can consistently define an effective 2D dielectric constant by averaging the screening over the extend of the exciton. For an ideal 2D semiconductor this leads to a simple expression for EB that only depends on the excitonic mass and the 2D polarizability α. The model is shown to produce accurate results for 51 transition metal dichalcogenides. Remarkably, over a wide range of polarizabilities the binding energy becomes independent of the mass and we obtain $E_{2DB} \approx 3/(4\pi\alpha)$, which explains the recently observed linear scaling of exciton binding energies with band gap. It is also shown that the model accurately reproduces the nonhydrogenic Rydberg series in WS2 and can account for screening from the environment.

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Scopus rating (2015): CiteScore 5.76 SJR 4.656 SNIP 2.538
Stark shift and electric-field-induced dissociation of excitons in monolayer MoS$_2$ and hBN/MoS$_2$ heterostructures

Efficient conversion of photons into electrical current in two-dimensional semiconductors requires, as a first step, the dissociation of the strongly bound excitons into free electrons and holes. Here we calculate the dissociation rates and energy shift of excitons in monolayer MoS$_2$ as a function of an applied in-plane electric field. The dissociation rates are obtained as the inverse lifetime of the resonant states of a two-dimensional hydrogenic Hamiltonian which describes the exciton within the Mott-Wannier model. The resonances are computed using complex scaling, and the effective masses and screened electron-hole interaction defining the hydrogenic Hamiltonian are computed from first principles. For field strengths above 0.1 V/nm the dissociation lifetime is shorter than 1 ps, which is below the lifetime associated with competing decay mechanisms. Interestingly, encapsulation of the MoS$_2$ layer in just two layers of hexagonal boron nitride (hBN), enhances the dissociation rate by around one order of magnitude due to the increased screening. This shows that dielectric engineering is an effective way to control exciton lifetimes in two-dimensional materials.
Adiabatic-connection fluctuation-dissipation DFT for the structural properties of solids - The renormalized ALDA and electron gas kernels

We present calculations of the correlation energies of crystalline solids and isolated systems within the adiabatic-connection fluctuation-dissipation formulation of density-functional theory. We perform a quantitative comparison of a set of model exchange-correlation kernels originally derived for the homogeneous electron gas (HEG), including the recently introduced renormalized adiabatic local-density approximation (rALDA) and also kernels which (a) satisfy known exact limits of the HEG, (b) carry a frequency dependence, or (c) display a 1/k^2 divergence for small wavevectors. After generalizing the kernels to inhomogeneous systems through a reciprocal-space averaging procedure, we calculate the lattice constants and bulk moduli of a test set of 10 solids consisting of tetrahedrally bonded semiconductors (C, Si, SiC), ionic compounds (MgO, LiCl, LiF), and metals (Al, Na, Cu, Pd). We also consider the atomization energy of the H2 molecule. We compare the results calculated with different kernels to those obtained from the random-phase approximation (RPA) and to experimental measurements. We demonstrate that the model kernels correct the RPA's tendency to overestimate the magnitude of the correlation energy whilst maintaining a high-accuracy description of structural properties.

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Contributors: Patrick, C. E., Thygesen, K. S.
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Amongst the X(Sn,Pb)Y3 perovskites currently under scrutiny for their photovoltaic applications, the cubic B−α phase of CsSnI₃ is arguably the best characterized experimentally. Yet, according to the standard harmonic theory of phonons, this deceptively simple phase should not exist at all due to rotational instabilities of the SnI₆ octahedra. Here, employing self-consistent phonon theory, we show that these soft modes are stabilized at experimental conditions through anharmonic phonon-phonon interactions between the Cs ions and their iodine cages. We further calculate the renormalization of the electronic energies due to vibrations and find an unusual opening of the band gap, estimated as 0.24 and 0.11 eV at 500 and 300 K, which we attribute to the stretching of Sn-I bonds. Our work demonstrates the important role of temperature in accurately describing these materials.
Band-gap engineering of functional perovskites through quantum confinement and tunneling

An optimal band gap that allows for a high solar-to-fuel energy conversion efficiency is one of the key factors to achieve sustainability. We investigate computationally the band gaps and optical spectra of functional perovskites composed of layers of the two cubic perovskite semiconductors BaSnO3 and BaTaO2N. Starting from an indirect gap of around 3.3 eV for BaSnO3 and a direct gap of 1.8 eV for BaTaO2N, different layerings can be used to design a direct gap of the functional perovskite between 2.3 and 1.2 eV. The variations of the band gap can be understood in terms of quantum confinement and tunneling. We also calculate the light absorption of the different heterostructures and demonstrate a large sensitivity to the detailed layering.
We present calculations of the optical properties of a set of around 80 oxides, oxynitrides, and organometal halide cubic and layered perovskites (Ruddlesden-Popper and Dion-Jacobson phases) with a bandgap in the visible part of the solar spectrum. The calculations show that for different classes of perovskites the solar light absorption efficiency varies greatly depending not only on bandgap size and character (direct/indirect) but also on the dipole matrix elements. The oxides exhibit generally a fairly weak absorption efficiency due to indirect bandgaps while the most efficient absorbers are found in the classes of oxynitride and organometal halide perovskites with strong direct transitions.
We present a comprehensive first-principles study of the electronic structure of 51 semiconducting monolayer transition-metal dichalcogenides and -oxides in the 2H and 1T hexagonal phases. The quasiparticle (QP) band structures with spin-orbit coupling are calculated in the G(0)W(0) approximation, and comparison is made with different density functional theory descriptions. Pitfalls related to the convergence of GW calculations for two-dimensional (2D) materials are discussed together with possible solutions. The monolayer band edge positions relative to vacuum are used to estimate the band alignment at various heterostructure interfaces. The sensitivity of the band structures to the in-plane lattice constant is analyzed and rationalized in terms of the electronic structure. Finally, the q-dependent dielectric functions and effective electron and hole masses are obtained from the QP band structure and used as input to a 2D hydrogenic model to estimate exciton binding energies. Throughout the paper we focus on trends and correlations in the electronic structure rather than detailed analysis of specific materials. All the computed data is available in an open database.
Computational design of molecules for dye sensitized solar cells and nano electronics
The huge increase in computational power has enabled the use of high-throughput computational screening methods for many purposes. In combination with more detailed computational studies, this provides a powerful tool in the search for new materials and molecules useful for e.g. photovoltaics. This is illustrated in this thesis, where a high-throughput Density Functional Theory study of a total of 5145 porphyrin based dye molecules is presented. Initially, the structures of the dyes are optimized and the frontier energy orbital energies calculated. Following this, the dyes are scored for use in a dye sensitized solar cell (DSSC) in terms of a loss-less level alignment quality. This scoring only takes into account a simplified absorption spectrum of the dye in combination with the alignment between the molecular levels, the semi-conductor conduction band edge and the redox mediator. To improve on this, the effect of the free energy barrier associated with the dye regeneration reaction is included through extensive molecular dynamics simulations for a simple model dye, followed by an extrapolation of the result to the 5145 porphyrins. This model succeeds in rediscovering high efficiency dyes and suggests that the next generation of high efficiency porphyrin dyes may utilize a titanium metal center. Furthermore, the large internal reorganization energies calculated for the octahedral cobalt complexes, used as redox mediators, lead to the requirement of a large driving force for the regeneration reaction. Hence, using redox mediators with a lower internal reorganization energy would allow for a less constrained choice of dye, possibly boosting the efficiency. The obtained data is furthermore used to search for suitable pairs of porphyrins for a novel type of DSSC schemes, using two dyes in a molecular two-photon tandem approach. Here, a high current is sacrificed for a larger voltage. As a smaller current is however often associated with a better fill-factor, the proposed scheme may lead to an increase in the efficiency. Specific candidates for the different schemes are identified and the resulting setups have theoretically obtainable open-circuit voltages exceeding 1.5 V.

Creating a metal-molecule-metal junction allows tuning the conductance through the junction by manipulating the molecular energy levels. In this thesis a computational approach to model the conductance as a function of the applied bias voltage, shifting the molecular levels, for a redox active molecule is presented and compared to experimental results. Here, it is shown that shifting the molecular energy levels in and out of resonance with the Fermi level of the electrodes, allows for a standard tuning of the conductance. In addition to this, reversibly changing the redox state, allows for switching quantum interference on and off, shifting the conductance by an order of magnitude. The simple computational model used is furthermore qualitatively found to be in very good agreement with experiments. A different way of tuning the conductance through a molecular junction, is by controlling the junction geometry. This is achieved by designing a molecule with two sets of anchor groups, which bind to gold with significantly different strengths. Hence, it is proposed that the geometry can be controlled by chemical passivisation of one type of anchor group. Using a simple computational model, this experimental hypothesis is verified and the change in conductance upon changing junction geometry is reproduced.

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Computational Discovery of Sustainable Energy Materials

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Organisations: Department of Physics, Theoretical Atomic-scale Physics
Contributors: Thygesen, K. S.
Computational Screening of Energy Materials
The current energy consumption of the world's population relies heavily on fossil fuels. Unfortunately, the consumption of fossil fuels not only results in the emission of greenhouse gases which have deleterious effects on the environment but also the fossil fuel reserve is limited. Therefore, it is the need of the hour to search for environmentally benign renewable energy resources. The biggest source of the renewable energy is our sun and the immense energy it provides can be used to power the whole planet. However, an efficient way to harvest the solar energy to meet all the energy demand has not been realized yet.

A promising way to utilize the solar energy is the photon assisted water splitting. The process involves the absorption of sunlight with a semiconducting material (or a photoabsorber) and the generated electron-hole pair can be used to produce hydrogen by splitting the water. However, a single material cannot accomplish the whole process of the hydrogen evolution. In order to do so, a material should be able to absorb the sunlight and generate the electronhole pairs and evolve hydrogen at the cathode and oxygen at anode using the generated electron and hole respectively.

This thesis using first-principle calculations explores materials for the light absorption with the bandgap, band edge positions and the stability in aqueous conditions as descriptors. This strategy results in a handful of materials which can act as good photoabsorbers for the water splitting reaction. Additionally, strategies to tune the bandgap for different applications is also explored. To carry out the cathode reaction, two-dimensional metal dichalcogenides and oxides are explored with a suggestion of few potential candidates for the hydrogen evolution reaction.

The thermodynamics of all the above process requires an accurate description of the energies with the first-principle calculations. Therefore, along this line the accuracy and predictability of the Meta-Generalized Gradient Approximation functional with Bayesian error estimation is also assessed.

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Controlling Electrical Conductance through a π-Conjugated Cruciform Molecule by Selective Anchoring to Gold Electrodes
Tuning charge transport at the single-molecule level plays a crucial role in the construction of molecular electronic devices. Introduced herein is a promising and operationally simple approach to tune two distinct charge-transport pathways through a cruciform molecule. Upon in situ cleavage of trisopropylsilyl groups, complete conversion from one junction type to another is achieved with a conductance increase by more than one order of magnitude, and it is consistent with predictions from ab initio transport calculations. Although molecules are well known to conduct through different orbitals (either HOMO or LUMO), the present study represents the first experimental realization of switching between HOMO- and LUMO-dominated transport within the same molecule.

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Contributors: Huang, C., Chen, S., Ørnsø, K. B., Reber, D., Baghernejad, M., Fu, Y., Wandlowski, T., Decurtins, S., Hong, W., Thygesen, K. S., Liu, S.
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Controlling Electrical Conductance through a π-Conjugated Cruciform Molecule by Selective Anchoring to Gold Electrodes

Tuning charge transport at the single-molecule level plays a crucial role in the construction of molecular electronic devices. Introduced herein is a promising and operationally simple approach to tune two distinct charge-transport pathways through a cruciform molecule. Upon in situ cleavage of triisopropylsilyl groups, complete conversion from one junction type to another is achieved with a conductance increase by more than one order of magnitude, and it is consistent with predictions from ab initio transport calculations. Although molecules are well known to conduct through different orbitals (either HOMO or LUMO), the present study represents the first experimental realization of switching between HOMO- and LUMO-dominated transport within the same molecule.
Design of two-photon molecular tandem architectures for solar cells by ab initio theory

An extensive database of spectroscopic properties of molecules from ab initio calculations is used to design molecular complexes for use in tandem solar cells that convert two photons into a single electron–hole pair, thereby increasing the output voltage while covering a wider spectral range. Three different architectures are considered: the first two involve a complex consisting of two dye molecules with appropriately matched frontier orbitals, connected by a molecular diode. Optimized combinations of dye molecules are determined by taking advantage of our computational database of the structural and energetic properties of several thousand porphyrin dyes. The third design is a molecular analogy of the intermediate band solar cell, and involves a single dye molecule with strong intersystem crossing to ensure a long lifetime of the intermediate state. Based on the calculated energy levels and molecular orbitals, energy diagrams are presented for the individual steps in the operation of such tandem solar cells. We find that theoretical open circuit voltages of up to 1.8 V can be achieved using these tandem designs. Questions about the practical implementation of prototypical devices, such as the synthesis of the tandem molecules and potential loss mechanisms, are addressed.

General information
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Electron transport in molecular junctions

This thesis addresses the electron transport in molecular junctions, focusing on the energy level alignment and correlation effects. Various levels of theory have been applied to study the structural and electronic effects in different molecular junctions, starting from the single particle density functional theory (DFT) description over the semi-empirical DFT+∑, to the sophisticated fully self-consistent GW approach. We find that in order to obtain a quantitative description of the conductance and the thermopower, it is necessary to go beyond the single particle description.

The effect of side groups on the benzene-diamine (BDA) molecule has furthermore been studied and it is found that the correct energy level alignment for the BDA molecule in Au contacts is only captured by the GW approach. Consequently, the GW approach provides an accurate description for the conductance change resulting from the side groups. The failure of the DFT based description is due to the strong energy level pinning when the BDA molecule is in contact with Au contacts.

The effect of contact geometries on the conductance and the thermopower has also been addressed. It is found that both GW and the DFT+∑ with a certain image charge position are in quantitative agreement with the experiments, while pure DFT is not. This is the consequence of the accurate energy level alignment, where the DFT+∑ method corrects the self-interaction error in the standard DFT functional and uses a static image charge model to include the image charge effect on the energy level renormalization.

Additionally, the gating of the 4,4′-bipyridine (44BP) molecule contacted to either Ni or Au electrodes has been investigated. Here it is found that the gating mechanism is conceptually different between two cases. In the case of Ni contacts where the lowest unoccupied molecular level (LUMO) of the 44BP molecule hybridizes strongly with Ni 3d orbitals, the gating is auxiliary by the so-called spinterface.

Finally, the correlation effect of the image charge beyond the energy level renormalization has been studied. It is shown that the finite response time of the electrodes to form image charge can suppress the conductance by a factor of 2. This correlation effect is only captured in the GW approach.
Excitons in van der Waals heterostructures: The important role of dielectric screening

The existence of strongly bound excitons is one of the hallmarks of the newly discovered atomically thin semiconductors. While it is understood that the large binding energy is mainly due to the weak dielectric screening in two dimensions, a systematic investigation of the role of screening on two-dimensional (2D) excitons is still lacking. Here we provide a critical assessment of a widely used 2D hydrogenic exciton model, which assumes a dielectric function of the form $\varepsilon(q) = 1 + 2\pi\alpha q$, and we develop a quasi-2D model with a much broader applicability. Within the quasi-2D picture, electrons and holes are described as in-plane point charges with a finite extension in the perpendicular direction, and their interaction is screened by a dielectric function with a nonlinear $q$ dependence which is computed ab initio. The screened interaction is used in a generalized Mott-Wannier model to calculate exciton binding energies in both isolated and supported 2D materials. For isolated 2D materials, the quasi-2D treatment yields results almost identical to those of the strict 2D model, and both are in good agreement with ab initio many-body calculations. On the other hand, for more complex structures such as supported layers or layers embedded in a van der Waals heterostructure, the size of the exciton in reciprocal space extends well beyond the linear regime of the dielectric function, and a quasi-2D description has to replace the 2D one. Our methodology has the merit of providing a seamless connection between the strict 2D limit of isolated monolayer materials and the more bulk-like screening characteristics of supported 2D materials or van der Waals heterostructures.
The shift from iodide-based redox mediators in dye-sensitized solar cells toward octahedral cobalt complexes has led to a significant increase in the efficiency. However, due to the nature of this type of complexes the driving force required for the regeneration of the dye is very high, and this limits the achievable efficiency. Here we show that the large driving force is a direct consequence of the large reorganization energy of the dye regeneration reaction. The reorganization energies for charge transfer between a simple zinc porphyrin dye and two popular cobalt-based redox mediators is calculated using ab initio molecular dynamics with explicit solvent. These results are then combined with a Marcus-based extrapolation scheme to obtain the reorganization energies of more than 5000 porphyrin-based dyes. We propose a scheme for scoring the performance of the porphyrin dyes, which is able to identify already known high-performance dyes in addition to a number of even better candidates. Our analysis shows that the large internal reorganization energy of the Co-based redox mediators is an obstacle for achieving higher efficiencies.
Improved description of metal oxide stability: Beyond the random phase approximation with renormalized kernels

The renormalized adiabatic PBE (rAPBE) method has recently been shown to comprise a significant improvement over the random phase approximation (RPA) for total energy calculations of simple solids and molecules. Here we consider the formation energies of 19 group I and II metal oxides and a few transition-metal oxides. The mean absolute error relative to experiments is 0.21 eV and 0.38 eV per oxygen atom for rAPBE and RPA, respectively, and thus the rAPBE method greatly improves the description of metal-oxygen bonds across a wide range of oxides. The failure of the RPA can be partly attributed to the lack of error cancellation between the correlation energy of the oxide on the one hand and the bulk metal and oxygen molecule on the other hand, which are all separately predicted much too negative by the RPA. We ascribe the improved performance of the rAPBE to its significantly better description of absolute correlation energies which reduces the need for error cancellation. The rAPBE is just one out of an entire class of renormalized exchange-correlation kernels which should be further investigated.

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Plasmonic eigenmodes in individual and bow-tie graphene nanotriangles

In classical electrodynamics, nanostructured graphene is commonly modeled by the computationally demanding problem of a three-dimensional conducting film of atomic-scale thickness. Here, we propose an efficient alternative two-dimensional electrostatic approach where all calculation procedures are restricted to the graphene sheet. Furthermore, to explore possible quantum effects, we perform tight-binding calculations, adopting a random-phase approximation. We investigate multiple plasmon modes in 20 nm equilateral triangles of graphene, treating the optical response classically as well as quantum mechanically. Compared to the classical plasmonic spectrum which is "blind" to the edge termination, we find that the quantum plasmon frequencies exhibit blueshifts in the case of armchair edge termination of the underlying atomic lattice, while redshifts are found for zigzag edges. Furthermore, we find spectral features in the zigzag case which are associated with electronic edge states not present for armchair termination. Merging pairs of triangles into dimers, plasmon hybridization leads to energy splitting that appears strongest in classical calculations while splitting is lower for armchair edges and even more reduced for zigzag edges. Our various results illustrate a surprising phenomenon: Even 20 nm large graphene structures clearly exhibit quantum plasmonic features due to atomic-scale details in the edge termination.

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Quantum theory of plasmons in nanostructures
In this thesis, ab initio quantum-mechanical calculations are used to study the properties of plasmons in nanostructures that involve atomic length-scales. The plasmon is an electronic excitation that corresponds to oscillations in the electron charge density in metals, often visualized as water ripples in a pond where the water represents a sea of free electrons. Plasmons on metal surfaces and in nanostructured materials, such as metal nanoparticles and atomically thin two-dimensional materials, have several technological applications due to their ability to confine light on nanoscale.

For a theoretical description of plasmon in such materials, where the electrons are heavily confined in one or more directions, a quantum mechanical description of the electrons in the material is necessary. In this thesis, the ab initio methods Density functional theory (DFT) and linear response time-dependent DFT are applied to calculate the properties of plasmons in nanostructures in different dimensions. In order to identify and visualize localized plasmon modes, a method for calculating plasmon eigenmodes within the ab initio framework has been developed. In the studied materials, quantum mechanical effects such as coupling to single-electronic transitions, electron spill-out from the surface, tunneling, and spatial non-locality, are shown to alter the plasmon excitations.

The studied systems include two-dimensional materials, such as thin metal films, monolayer transition metal dichalcogenides, and graphene. Here, also van der Waals heterostructures (vdWh), which are stacks of different twodimensional materials, are considered. A new multi-scale approach for calculating the dielectric-function of vdWh, which extends ab initio accuracy to the description of hundreds of atomic layers, is presented. Also, one-dimensional plasmons are studied in the case of atomically thin nanowires and edge-states of MoS₂.

Single-Molecule Electrochemical Transistor Utilizing a Nickel-Pyridyl Spinterface
Using a scanning tunnelling microscope break-junction technique, we produce 4,4'-bipyridine (44BP) single-molecule junctions with Ni and Au contacts. Electrochemical control is used to prevent Ni oxidation and to modulate the conductance of the devices via nonredox gating - the first time this has been shown using non-Au contacts. Remarkably the conductance and gain of the resulting Ni-44BP-Ni electrochemical transistors is significantly higher than analogous Au-based devices. Ab-initio calculations reveal that this behavior arises because charge transport is mediated by spin-polarized Ni d-electrons, which hybridize strongly with molecular orbitals to form a "spinterface". Our results highlight the important role of the contact material for single-molecule devices and show that it can be varied to provide control of charge and spin transport.
Strain sensitivity of band gaps of Sn-containing semiconductors

Tuning of band gaps of semiconductors is a way to optimize materials for applications within photovoltaics or as photocatalysts. One way to achieve this is through applying strain to the materials. We investigate the effect of strain on a range of Sn-containing semiconductors using density functional theory and many-body perturbation theory calculations. We find that the band gaps of bulk Sn oxides with SnO$_6$ octahedra are highly sensitive to volumetric strain. By applying a small isotropic strain of 2% (2%), a decrease (increase) of band gaps as large as 0.8 to 1.0 eV are obtained. We attribute the ultrahigh strain sensitivity to the pure Sn s-state character of the conduction-band edges. Other Sn-containing compounds may show both increasing and decreasing gaps under tensile strain and we show that the behavior can be understood by analyzing the role of the Sn s states in both the valence and the conduction bands.
The dielectric genome of van der Waals heterostructures

Vertical stacking of two-dimensional (2D) crystals, such as graphene and hexagonal boron nitride, has recently lead to a new class of materials known as van der Waals heterostructures (vdWHs) with unique and highly tunable electronic properties. Ab initio calculations should in principle provide a powerful tool for modeling and guiding the design of vdWHs, but in their traditional, form such calculations are only feasible for commensurable structures with a few layers. Here we show that the dielectric properties of realistic, incommensurable vdWHs comprising hundreds of layers can be calculated with ab-initio accuracy using a multi-scale approach where the dielectric functions of the individual layers (the dielectric building blocks) are coupled simply via their long-range Coulomb interaction. We use the method to illustrate the 2D-3D dielectric transition in multi-layer MoS$_2$ crystals, the hybridization of quantum plasmons in large graphene/hBN heterostructures, and to demonstrate the intricate effect of substrate screening on the non-Rydberg exciton series in supported WS$_2$. The dielectric building blocks for a variety of 2D crystals are available in an open database together with the software for solving the coupled electrodynamical equations.
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Tuning the Schottky Barrier at the Graphene/MoS₂ Interface by Electron Doping: Density Functional Theory and Many-Body Calculations

Using ab initio calculations we investigate the energy level alignment at the graphene/MoS₂ heterostructure and the use of electron doping as a strategy to lower the Schottky barrier and achieve a low-resistance Ohmic contact. For the neutral heterostructure, density functional theory (DFT) with a generalized gradient approximation predicts a Schottky barrier height of 0.18 eV, whereas the G₀W₀ method increases this value to 0.60 eV. While the DFT band gap of MoS₂ does not change when the heterostructure is formed, the G₀W₀ gap is reduced by 0.30 eV as a result of the enhanced screening by the graphene layer. In contrast to the case of metal substrates, where the band alignment is governed by Pauli repulsion-induced interface dipoles, the graphene/MoS₂ heterostructure shows only a negligible interface dipole. As a consequence, the band alignment at the neutral heterostructure is not changed when the two layers are brought into contact. We systematically follow the band alignment as a function of doping concentration and find that the Fermi level of the graphene crosses the MoS₂ conduction band at a doping concentration of around 10¹² cm⁻². The variation of the energy levels with doping concentration is shown to be mainly governed by the electrostatic potential resulting from the doping charge.

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Two-dimensional metal dichalcogenides and oxides for hydrogen evolution: A computational screening approach

We explore the possibilities of hydrogen evolution by basal planes of 2D metal dichalcogenides and oxides in the 2H and 1T class of structures using the hydrogen binding energy as a computational activity descriptor. For some groups of systems like the Ti, Zr, and Hf dichalcogenides the hydrogen bonding to the 2H structure is stronger than that to the 1T structure, while for the Cr, Mo, and W dichalcogenides the behavior is opposite. This is rationalized by investigating shifts in the chalcogenide p levels comparing the two structures. We find that usually for a given material only at most one of the two phases will be active for the hydrogen evolution reaction; however, in most cases the two phases are very close in formation energy, opening up the possibility for stabilizing the active phase. The study points to many new possible 2D HER materials beyond the few that are already known.

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Accurate Ground-State Energies of Solids and Molecules from Time-Dependent Density-Functional Theory

We demonstrate that ground-state energies approaching chemical accuracy can be obtained by combining the adiabatic-connection fluctuation-dissipation theorem with time-dependent density-functional theory. The key ingredient is a renormalization scheme, which eliminates the divergence of the correlation hole characteristic of any local kernel. This new class of renormalized kernels gives a significantly better description of the short-range correlations in covalent bonds compared to the random phase approximation (RPA) and yields a fourfold improvement of RPA binding energies in both molecules and solids. We also consider examples of barrier heights in chemical reactions, molecular adsorption, and graphene interacting with metal surfaces, which are three examples where the RPA has been successful. In these cases, the renormalized kernel provides results that are of equal quality or even slightly better than the RPA, with a similar computational cost.
Bandgap calculations and trends of organometal halide perovskites

Energy production from the Sun requires a stable efficient light absorber. Promising candidates in this respect are organometal perovskites (ABX₃), which have been intensely investigated during the last years. Here, we have performed electronic structure calculations of 240 perovskites composed of Cs, CH₃NH₃, and HC(NH₂)₂ as A-cation, Sn and Pb as
B-ion, and a combination of Cl, Br, and I as anions. The calculated gaps span over a region from 0.5 to 5.0 eV. In addition, the trends over bandgaps have been investigated: the bandgap increases with an increase of the electronegativities of the constituent species, while it reduces with an increase of the lattice constants of the system.

**Calculated Pourbaix Diagrams of Cubic Perovskites for Water Splitting: Stability Against Corrosion**

We use density functional theory calculations to investigate the stability of cubic perovskites for photo-electrochemical water splitting taking both materials in their bulk crystal structure and dissolved phases into account. The method is validated through a detailed comparison of the calculated and experimental Pourbaix diagrams for TiO₂ and ZnO. For a class of 23 oxides, oxynitrides, and oxyfluorides, which were recently proposed as candidates for one-photon water splitting, our calculations predict most of the materials to be stable at potentials around the water red-ox level. The oxides become less stable at lower potentials, while the oxynitrides become unstable at higher potentials. We discuss the implications of these findings for the problem of photo-corrosion of water splitting electrodes.
Cross-conjugation and quantum interference: a general correlation?

We discuss the relationship between the pi-conjugation pattern, molecular length, and charge transport properties of molecular wires, both from an experimental and a theoretical viewpoint. Specifically, we focus on the role of quantum interference in the conductance properties of cross-conjugated molecules. For this, we compare experiments on two series of dithiolated wires. The first set we synthesized consists of three dithiolated oligo(phenylene ethynylene) (OPE) benchmark compounds with increasing length. The second series synthesized comprises three molecules with different pi-conjugation patterns, but identical lengths, i.e. an anthracene (linear conjugation), an anthraquinone (cross-conjugation), and a dihydroanthracene (broken conjugation) derivative. To benchmark reliable trends, conductance experiments on these series have been performed by various techniques. Here, we compare data obtained by conductive-probe atomic force microscopy (CP-AFM) for self-assembled monolayers (SAMs) with single-molecule break junction and multi-molecule EGaIn data from other groups. For the benchmark OPE-series, we consistently find an exponential decay of the conductance with molecular length characterized by beta = 0.37 +/- 0.03 angstrom(-1) (CP-AFM). Remarkably, for the second series, we do not only find that the linearly conjugated anthracene-containing wire is the most conductive, but also that the cross-conjugated anthraquinone-containing wire is less conductive than the broken-conjugated derivative. We attribute the low conductance values for the cross-conjugated species to quantum interference effects. Moreover, by theoretical modeling, we show that destructive quantum interference is a robust feature for cross-conjugated structures and that the energy at which complete destructive interference occurs can be tuned by the choice of side group. The latter provides an outlook for future devices in this fascinating field connecting chemistry and physics.

General information
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Development and testing of new exchange correlation functionals

Catalysts are used in 90% of the world's chemical processes to produce 60% of its chemical products, and they are thus very important to our modern society. We therefore seek to better understand current catalytic materials, so that we can find alternatives that will improve the energy efficiency, selectivity or similar of current chemical processes, or to make new technologies economical feasible. Kohn-Sham density functional theory (KS-DFT) has proven to be a powerful theory to find trends in current catalytic materials, which can empower a more informed search for better alternatives. KS-DFT relies on accurate and efficient approximations to the exchange correlation functional, yet these functional approximations have lacked a systematic way to estimate the underlying uncertainties. A Bayesian error estimation approach provides a mechanism for calculating approximative uncertainties, and so accurate, computationally feasible exchange-correlation approximations that incorporate it have been called for. This thesis presents significant steps forwards towards providing general applicable exchange-correlation functional approximations with Bayesian error estimation capabilities. A semi-empirical approach was used with a machine learning toolset to improve accuracy and transferability of the functional approximations. The toolset includes Tikhonov regularization of smoothness in a transformed model space, for ensuring sensible model solutions; an explicit model compromise with a geometric mean loss function, for ensuring generally applicable models; a robust MM-estimator loss function, for ensuring resistance to outliers in data; and a hierarchical bootstrap resampling estimating prediction error validation method, for selecting the model complexity that provide best transferability outside the training data. Three new semi-empirical functional approximations have been made: BEEF-vdW, mBEEF, and mBEEFvdW. It is shown that these functionals are able balance the accuracy of predicting energetics of covalent and non-covalent chemistry better than any comparative functional that we have tested, and they could therefore become the functional approximations of choice for understanding chemical processes at the solid-gas and solid-liquid interfaces.
Electrochemical Control of Single-Molecule Conductance by Fermi-Level Tuning and Conjugation Switching

Controlling charge transport through a single molecule connected to metallic electrodes remains one of the most fundamental challenges of nanoelectronics. Here we use electrochemical gating to reversibly tune the conductance of two different organic molecules, both containing anthraquinone (AQ) centers, over >1 order of magnitude. For electrode potentials outside the redox-active region, the effect of the gate is simply to shift the molecular energy levels relative to the metal Fermi level. At the redox potential, the conductance changes abruptly as the AQ unit is oxidized/reduced with an accompanying change in the conjugation pattern between linear and cross conjugation. The most significant change in conductance is observed when the electron pathway connecting the two electrodes is via the AQ unit. This is consistent with the expected occurrence of destructive quantum interference in that case. The experimental results are supported by an excellent agreement with ab initio transport calculations.

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Web of Science (2017): Impact factor 14.357
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Web of Science (2016): Impact factor 13.858
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BFI (2015): BFI-level 2
Scopus rating (2015): CiteScore 12.81 SJR 6.775 SNIP 2.63
Web of Science (2015): Impact factor 13.038
New Light-Harvesting Materials Using Accurate and Efficient Bandgap Calculations

Electronic bandgap calculations are presented for 2400 experimentally known materials from the Materials Project database and the bandgaps, obtained with different types of functionals within density functional theory and (partial) self-consistent GW approximation, are compared for 20 randomly chosen compounds forming an unconventional set of ternary and quaternary materials. It is shown that the computationally cheap GLLB-SC potential gives results in good agreement (around 15%) with the more advanced and demanding eigenvalue-self-consistent GW. This allows for a high-throughput screening of materials for different applications where the bandgaps are used as descriptors for the efficiency of a photoelectrochemical device. Here, new light harvesting materials are proposed to be used in a one-photon photoelectrochemical device for water splitting by combining the estimation of the bandgaps with the stability analysis using Pourbaix diagrams and with the evaluation of the position of the band edges. Using this methodology, 25 candidate materials are obtained and 5 of them appear to have a realistic possibility of being used as photocatalyst in a one-photon water splitting device. © 2014 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim.

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Web of Science (2017): Indexed yes
BFI (2016): BFI-level 1
Scopus rating (2016): CiteScore 12.96 SJR 6.515 SNIP 2.14
Web of Science (2016): Impact factor 16.721
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BFI (2015): BFI-level 1
Scopus rating (2015): CiteScore 14.2 SJR 6.219 SNIP 2.546
Web of Science (2015): Impact factor 15.23
Web of Science (2015): Indexed yes
BFI (2014): BFI-level 1
Scopus rating (2014): CiteScore 15.27 SJR 6.668 SNIP 2.942
Web of Science (2014): Impact factor 16.146
Web of Science (2014): Indexed yes
BFI (2013): BFI-level 1
Scopus rating (2013): CiteScore 13.24 SJR 6.006 SNIP 2.949
Web of Science (2013): Impact factor 14.385
ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
Scopus rating (2012): CiteScore 9.64 SJR 5.575 SNIP 2.181
Web of Science (2012): Impact factor 10.043
Optimizing porphyrins for dye sensitized solar cells using large-scale ab initio calculations

In the search for sustainable energy sources, dye sensitized solar cells (DSSCs) represent an attractive solution due to their low cost, relatively high efficiencies, and flexible design. Porphyrin-based dyes are characterized by strong absorption in the visible part of the spectrum and easy customization allowing their electronic properties to be controlled by structural variations. Here we present a computational screening study of more than 5000 porphyrin-based dyes obtained by modifying the porphyrin backbone (metal center and axial ligands), substituting hydrogen by fluorine, and adding different side and anchoring groups. Based on the calculated frontier orbital energies and optical gaps we quantify the energy level alignment with the TiO2 conduction band and different redox mediators. An analysis of the energy level-structure relationship reveals a significant structural diversity among the dyes with the highest level alignment quality, demonstrating the large degree of flexibility in porphyrin dye design. As a specific example of dye optimization, we show that the level alignment of the high efficiency record dye YD2-o-C8 [Yella et al., Science, 2011, 334, 629-634] can be significantly improved by modest structural variations. All the presented data have been stored in a publicly available database.
Plasmons on the edge of MoS$_2$ nanostructures
Using ab initio calculations we predict the existence of one-dimensional (1D), atomically confined plasmons at the edges of a zigzag MoS$_2$ nanoribbon. The strongest plasmon originates from a metallic edge state localized on the sulfur dimers decorating the Mo edge of the ribbon. A detailed analysis of the dielectric function reveals that the observed deviations from the ideal 1D plasmon behavior result from single-particle transitions between the metallic edge state and the valence and conduction bands of the MoS$_2$ sheet. The Mo and S edges of the ribbon are clearly distinguishable in calculated spatially resolved electron energy loss spectrum owing to the different plasmonic properties of the two edges. The edge plasmons could potentially be utilized for tuning the photocatalytic activity of MoS$_2$ nanoparticles.

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Scopus rating (2015): CiteScore 2.8 SJR 2.377 SNIP 1.13
Web of Science (2015): Impact factor 3.718
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ISI indexed (2013): ISI indexed yes
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Scopus rating (2012): CiteScore 3.57 SJR 3.173 SNIP 1.378
Web of Science (2012): Impact factor 3.767
ISI indexed (2012): ISI indexed yes
Web of Science (2012): Indexed yes
Scopus rating (2011): CiteScore 3.61 SJR 3.326 SNIP 1.423
Web of Science (2011): Impact factor 3.691
ISI indexed (2011): ISI indexed yes
Web of Science (2011): Indexed yes
Scopus rating (2010): SJR 3.318 SNIP 1.447
Web of Science (2010): Impact factor 3.774
Web of Science (2010): Indexed yes
Simultaneous description of conductance and thermopower in single-molecule junctions from many-body ab initio calculations

We investigate the electronic conductance and thermopower of a single-molecule junction consisting of bis-(4-aminophenyl) acetylene (B4APA) connected to gold electrodes. We use nonequilibrium Green's function methods in combination with density-functional theory (DFT) and the many-body GW approximation. To simulate recent break junction experiments, we calculate the transport properties of the junction as it is pulled apart. For all junction configurations, DFT with a standard semilocal functional overestimates the conductance by almost an order of magnitude, while the thermopower is underestimated by up to a factor of 3, except for the most highly stretched junction configurations. In contrast, the GW results for both conductance and thermopower are in excellent agreement with experiments for a wide range of electrode separations. We show that the GW self-energy not only renormalizes the molecular energy levels but also the coupling strength. The latter is a consequence of the finite response time associated with the electronic screening in the metal electrodes.

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Contributors: Jin, C., Markussen, T., Thygesen, K. S.
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Scopus rating (2016): CiteScore 3.16 SJR 2.339 SNIP 1.151
Web of Science (2016): Impact factor 3.836
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Scopus rating (2015): CiteScore 2.8 SJR 2.377 SNIP 1.13
Web of Science (2015): Impact factor 3.718
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Scopus rating (2014): CiteScore 3.3 SJR 2.762 SNIP 1.316
Web of Science (2014): Impact factor 3.736
Web of Science (2014): Indexed yes
Scopus rating (2013): CiteScore 3.55 SJR 2.813 SNIP 1.326
Web of Science (2013): Impact factor 3.664
ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
Scopus rating (2012): CiteScore 3.57 SJR 3.173 SNIP 1.378
Web of Science (2012): Impact factor 3.767
ISI indexed (2012): ISI indexed yes
Web of Science (2012): Indexed yes
Scopus rating (2011): CiteScore 3.61 SJR 3.326 SNIP 1.423
Web of Science (2011): Impact factor 3.691
ISI indexed (2011): ISI indexed yes
Web of Science (2011): Indexed yes
Scopus rating (2010): SJR 3.318 SNIP 1.447
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Web of Science (2010): Indexed yes
Web of Science (2009): Indexed yes
Scopus rating (2008): SJR 2.923 SNIP 1.516
Web of Science (2008): Indexed yes
Scopus rating (2007): SJR 2.892 SNIP 1.588
Web of Science (2007): Indexed yes
Scopus rating (2006): SJR 2.62 SNIP 1.468
Web of Science (2006): Indexed yes
Scopus rating (2005): SJR 2.126 SNIP 1.156
Web of Science (2005): Indexed yes
Scopus rating (2004): SJR 2.012 SNIP 1.103
Web of Science (2004): Indexed yes
Scopus rating (2003): SJR 2.184 SNIP 1.179
Web of Science (2003): Indexed yes
Scopus rating (2002): SJR 2.856 SNIP 1.841
Web of Science (2002): Indexed yes
Scopus rating (2001): SJR 3.132 SNIP 1.727
Web of Science (2001): Indexed yes
Scopus rating (2000): SJR 2.84 SNIP 1.603
Web of Science (2000): Indexed yes
Static correlation beyond the random phase approximation: Dissociating H2 with the Bethe-Salpeter equation and time-dependent GW

We investigate various approximations to the correlation energy of a H2 molecule in the dissociation limit, where the ground state is poorly described by a single Slater determinant. The correlation energies are derived from the density response function and it is shown that response functions derived from Hedin's equations (Random Phase Approximation (RPA), Time-dependent Hartree-Fock (TDHF), Bethe-Salpeter equation (BSE), and Time-Dependent GW) all reproduce the correct dissociation limit. We also show that the BSE improves the correlation energies obtained within RPA and TDHF significantly for intermediate binding distances. A Hubbard model for the dimer allows us to obtain exact analytical results for the various approximations, which is readily compared with the exact diagonalization of the model. Moreover, the model is shown to reproduce all the qualitative results from the ab initio calculations and confirms that BSE greatly improves the RPA and TDHF results despite the fact that the BSE excitation spectrum breaks down in the dissociation limit. In contrast, second order screened exchange gives a poor description of the dissociation limit, which can be attributed to the fact that it cannot be derived from an irreducible response function. (C) 2014 AIP Publishing LLC.
Temperature effects on quantum interference in molecular junctions

A number of experiments have demonstrated that destructive quantum interference (QI) effects in molecular junctions lead to very low conductances even at room temperature. On the other hand, another recent experiment showed increasing conductance with temperature which was attributed to decoherence effects destroying QI at finite temperatures. Here we study the influence of finite temperatures and electron-phonon interactions on QI in molecular junctions. Two different models leading to two inherently different types of QI effects are considered. Each model is exemplified by specific molecules and studied using first-principles calculations. We find that the molecules exhibiting QI show a much stronger temperature dependence of the conductance compared to molecules without QI. However, the large QI-induced suppression of the conductance remains, showing that QI effects are indeed robust against finite temperatures and inelastic scattering.

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Web of Science (2017): Indexed yes
Scopus rating (2016): CiteScore 3.16 SJR 2.339 SNIP 1.151
Web of Science (2016): Impact factor 3.836
Web of Science (2016): Indexed yes
Scopus rating (2015): CiteScore 2.8 SJR 2.377 SNIP 1.13
Web of Science (2015): Impact factor 3.718
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Scopus rating (2014): CiteScore 3.3 SJR 2.762 SNIP 1.316
Web of Science (2014): Impact factor 3.736
Web of Science (2014): Indexed yes
Scopus rating (2013): CiteScore 3.55 SJR 2.813 SNIP 1.326
Web of Science (2013): Impact factor 3.664
ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
Scopus rating (2012): CiteScore 3.57 SJR 3.173 SNIP 1.378
Web of Science (2012): Impact factor 3.767
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Web of Science (2012): Indexed yes
Scopus rating (2011): CiteScore 3.61 SJR 3.326 SNIP 1.423
Web of Science (2011): Impact factor 3.691
ISI indexed (2011): ISI indexed yes
Web of Science (2011): Indexed yes
Scopus rating (2010): SJR 3.318 SNIP 1.447
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Acoustic phonon limited mobility in two-dimensional semiconductors: Deformation potential and piezoelectric scattering in monolayer MoS$_2$ from first principles

We theoretically study the acoustic phonon limited mobility in n-doped two-dimensional MoS$_2$ for temperatures T<100 K and high carrier densities using the Boltzmann equation and first-principles calculations of the acoustic electron-phonon (el-ph) interaction. In combination with a continuum elastic model, analytic expressions and the coupling strengths for the deformation potential and piezoelectric interactions are established. We furthermore show that the deformation potential interaction has contributions from both normal and umklapp processes and that the latter contribution is only weakly affected by carrier screening. Consequently, the calculated mobilities show a transition from a high-temperature $\mu \sim T^{-1}$ behavior to a stronger $\mu \sim T^{-4}$ behavior in the low-temperature Bloch-Grüneisen regime characteristic of unscreened deformation potential scattering. Intrinsic mobilities in excess of 105 cm$^2$ V$^{-1}$ s$^{-1}$ are predicted at T<10 K and high carrier densities (n>10$^{11}$ cm$^{-2}$). At 100 K, the mobility does not exceed $\sim 7 \times 10^3$ cm$^2$ V$^{-1}$ s$^{-1}$. Our findings provide new and important understanding of the acoustic el-ph interaction and its screening by free carriers, and is of high relevance for the understanding of acoustic phonon-limited mobilities in general.

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Organisations: Department of Physics, Theoretical Atomic-scale Physics, Center for Atomic-scale Materials Design, Department of Micro- and Nanotechnology, Nanointegration, Theoretical Nanotechnology, Center for Nanostructured Graphene, Tel Aviv University
Contributors: Kaasbjerg, K., Thygesen, K. S., Jauho, A.
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Avoiding pitfalls in the modeling of electrochemical interfaces

Alignment of metal and molecular electronic energy levels at electrode-electrolyte interfaces is investigated using density functional theory. Three different regimes exhibiting qualitatively different energy level alignments are observed. The regimes are roughly defined by the size of the metal work function relative to the ionization potential and/or electron affinity of the electrolyte. It is demonstrated that proper matching of these quantities is essential for successful ab initio modeling of electrochemical interfaces and it is further discussed how such matching can be obtained by careful tailoring of the interfacial atomic structure.

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Contributors: Björketun, M., Zeng, Z., Ahmed, R., Tripkovic, V., Thygesen, K. S., Rossmeisl, J.
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Scopus rating (2015): CiteScore 1.83 SJR 0.709 SNIP 0.707
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BFI (2014): BFI-level 1
Scopus rating (2014): CiteScore 1.83 SJR 0.755 SNIP 0.765
Web of Science (2014): Impact factor 1.897
BFI (2013): BFI-level 1
Scopus rating (2013): CiteScore 2.07 SJR 0.858 SNIP 0.835
Web of Science (2013): Impact factor 1.991
ISI indexed (2013): ISI indexed yes
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BFI (2012): BFI-level 1
Scopus rating (2012): CiteScore 2.2 SJR 1.104 SNIP 0.915
Bandgap Engineering of Double Perovskites for One- and Two-photon Water Splitting

Computational screening is becoming increasingly useful in the search for new materials. We are interested in the design of new semiconductors to be used for light harvesting in a photoelectrochemical cell. In the present paper, we study the double perovskite structures obtained by combining 46 stable cubic perovskites which was found to have a finite bandgap in a previous screening-study. The four-metal double perovskite space is too large to be investigated completely. For this reason we propose a method for combining different metals to obtain a desired bandgap. We derive some bandgap design rules on how to combine two cubic perovskites to generate a new combination with a larger or smaller bandgap compared with the constituent structures. Those rules are based on the type of orbitals involved in the conduction bands and on the size of the two cubic bandgaps. We also see that a change in the volume has an effect on the size of the bandgap. In addition, we suggest some new candidate materials that can be used as photocatalysts in one- and two-photon water splitting devices.
Beyond the random phase approximation: Improved description of short-range correlation by a renormalized adiabatic local density approximation

We assess the performance of a recently proposed renormalized adiabatic local density approximation (rALDA) for ab initio calculations of electronic correlation energies in solids and molecules. The method is an extension of the random phase approximation (RPA) derived from time-dependent density functional theory and the adiabatic connection fluctuation-dissipation theorem and contains no fitted parameters. The new kernel is shown to preserve the accurate description of dispersive interactions from RPA while significantly improving the description of short-range correlation in molecules, insulators, and metals. For molecular atomization energies, the rALDA is a factor of 7 better than RPA and a factor of 4 better than the Perdew-Burke-Ernzerhof (PBE) functional when compared to experiments, and a factor of 3 (1.5) better than RPA (PBE) for cohesive energies of solids. For transition metals, the inclusion of full shell semicore states is found to be crucial for both RPA and rALDA calculations and can improve the cohesive energies by up to 0.5 shell semicore states.4 eV. Finally, we discuss straightforward generalizations of the method, which might improve results even further.

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Web of Science (2017): Impact factor 3.813
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Scopus rating (2016): CiteScore 3.16 SJR 2.339 SNIP 1.151
Web of Science (2016): Impact factor 3.836
Web of Science (2016): Indexed yes
Scopus rating (2015): CiteScore 2.8 SJR 2.377 SNIP 1.13
Web of Science (2015): Impact factor 3.718
Web of Science (2015): Indexed yes
Scopus rating (2014): CiteScore 3.3 SJR 2.762 SNIP 1.316
Web of Science (2014): Impact factor 3.736
Web of Science (2014): Indexed yes
Scopus rating (2013): CiteScore 3.55 SJR 2.813 SNIP 1.326
Web of Science (2013): Impact factor 3.664
ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
Scopus rating (2012): CiteScore 3.57 SJR 3.173 SNIP 1.378
Web of Science (2012): Impact factor 3.767
ISI indexed (2012): ISI indexed yes
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Scopus rating (2011): CiteScore 3.61 SJR 3.326 SNIP 1.423
Web of Science (2011): Impact factor 3.691
ISI indexed (2011): ISI indexed yes
Web of Science (2011): Indexed yes
Scopus rating (2010): SJR 3.318 SNIP 1.447
Web of Science (2010): Impact factor 3.774
Carbon nanotubes as heat dissipaters in microelectronics
We review our recent modelling work of carbon nanotubes as potential candidates for heat dissipation in microelectronics cooling. In the first part, we analyze the impact of nanotube defects on their thermal transport properties. In the second part, we investigate the loss of thermal properties of nanotubes in presence of an interface with various substances, including air and water. Comparison with previous works is established whenever is possible.

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Computational Approach to Electron Charge Transfer Reactions
The step from ab initio atomic and molecular properties to thermodynamic - or macroscopic - properties requires the combination of several theoretical tools. This dissertation presents constant temperature molecular dynamics with bond length constraints, a hybrid quantum mechanics-molecular mechanics scheme, and tools to analyse statistical data and generate relative free energies and free energy surfaces. The methodology is applied to several charge transfer species and reactions in chemical environments - chemical in the sense that solvent, counter ions and substrate surfaces are taken in to account - which directly influence the reactants and resulting reaction through both physical and chemical interactions. All methods are though general and can be applied to different types of chemistry. First, the basis of the various theoretical tools is presented and applied to several test systems to show general (or expected) properties. Properties such as in the physical and (semi-)chemical interface between classical and quantum systems and the effects of molecular bond length constraints on the temperature during simulations. As a second step the methodology is applied to the symmetric and asymmetric charge transfer reactions between several first-row transition metals in water. The results are compared to experiments and rationalised with classical analytic expressions. Shortcomings of the methods are accounted for with clear steps towards improved accuracy. Later the analysis is extended to more complex systems composed of a larger osmium complex in solution and at the solute-substrate interfaces, where in particular the redox state of the complex is controlled through chemical means. The efficiency of the hybrid-classical and quantum mechanics method is used to generate adequate statistics and a simple post-sampling scheme used to generate free energy surfaces - which compare to full ab initio calculations. In the last part both the molecular dynamics and hybrid classical and quantum mechanics method are used to generate a vast data set for the accurate analysis of dynamical structure modes. This is for a large iridium-iridium dimer complex which shows a dramatic structural (and vibrational) change upon electronic excitation.

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Contributors: Jónsson, E. Ö., Jacobsen, K. W., Thygesen, K. S., Ulstrup, J.
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Computational screening of functionalized zinc porphyrins for dye sensitized solar cells
An efficient dye sensitized solar cell (DSSC) is one possible solution to meet the world's rapidly increasing energy demands and associated climate challenges. This requires inexpensive and stable dyes with well-positioned frontier energy levels for maximal solar absorption, efficient charge separation, and high output voltage. Here we demonstrate an extensive computational screening of zinc porphyrins functionalized with electron donating side groups and electron accepting anchoring groups. The trends in frontier energy levels versus side groups are analyzed and a no-loss DSSC level alignment quality is estimated. Out of the initial 1029 molecules, we find around 50 candidates with level alignment qualities within 5% of the optimal limit. We show that the level alignment of five zinc porphyrin dyes which were recently used in DSSCs with high efficiencies can be further improved by simple side group substitutions. All frontier energy levels, gaps and level alignment quality values are stored in a database publicly available.

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Computational Screening of Materials for Water Splitting Applications

Design new materials for energy production in a photoelectrochemical cell, where water is split into hydrogen and oxygen by solar light, is one possible solution to the problem of increasing energy demand and storage. A screening procedure based on ab-initio density functional theory calculations has been applied to guide the search for new materials. The main descriptors of the properties relevant for the screening are: heat of formation, electronic bandgap, and positions of the band edges with respect to the red-ox levels of water. A recently implemented exchange-correlation functional, called GLLB-SC, has been used for the estimation of the bandgaps. Firstly, a screening procedure has been applied to 19000 cubic perovskite structures. These are obtained by combining 52 metals together with oxygen, nitrogen, sulfur and fluorine as anions. 32 promising materials have been found for visible light harvesting, 20 for the one-photon and 12 for the two-photon water splitting process. In addition, 16 candidates were suggested for the transparent shielding of the photocatalyst. The problem of corrosion has been addressed for the candidates for the one-photon scheme using Pourbaix diagrams.

Later on, the screening has been extended to more complex structures, like double and layered perovskites and new compounds of interest for the light harvesting problem were found. In addition, the trends in the bandgaps have been studied. The bandgaps can be tuned by an opportune combination of the metal atoms in the B-ion position in the double perovskite, and of the B-metal ion with the thickness of the octahedra in the layered perovskite structure. In the first part, the crystal structure has been kept fixed. Later, the calculations were done using the structures provided by the Materials Project database, which is based on the experimental ICSD database, and the bandgaps were calculated with focus on finding materials with potential as light harvesters. 24 materials have been proposed for the one-photon water splitting and 23 for the two-photon mechanism. Another method to obtain energy from Sun is using a photovoltaic cell that converts solar light into electricity. The absorption spectra of 70 experimentally known compounds, that are expected to be useful for light-to-electricity generation, have been calculated. 17 materials have been predicted to be promising for a single-layer solar cell and 11 for a double-layer device.
DFT+U study of polaronic conduction in Li₂O₂ and Li₂CO₃: Implications for Li-air batteries

The main discharge products formed at the cathode of nonaqueous Li-air batteries are known to be Li₂O₂ and residual Li₂CO₃. Recent experiments indicate that the charge transport through these materials is the main limiting factor for the battery performance. It has been also shown that the performance of the battery decreases drastically when the amount of Li₂CO₃ at the cathode increases with respect to Li₂O₂. In this work, we study the formation and transport of hole and electron polarons in Li₂O₂ and Li₂CO₃ using density functional theory (DFT) within the PBE+U approximation. For both materials, we find that the formation of polarons (both hole and electron) is stabilized with respect to the delocalized states for all physically relevant values of U. We find a much higher mobility for hole polarons than for the electron polarons, and we show that the poor charge transport in Li₂CO₃ compared to Li₂O₂ can be understood through a polaronic model for the conduction. Furthermore, the hole polaronic model in Li₂O₂ provides a possible explanation for the experimentally observed preferential growth direction of the films. Our results also suggest that doping is unlikely to be a viable route for improving the transport properties of Li₂O₂ or Li₂CO₃.

General information

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Contributors: García Lastra, J. M., Myrdal, J., Christensen, R., Thøgesen, K., Vegge, T.
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Direct measurement and modulation of single-molecule coordinative bonding forces in a transition metal complex.

Coordination chemistry has been a consistently active branch of chemistry since Werner’s seminal theory of coordination compounds inaugurated in 1893, with the central focus on transition metal complexes. However, control and measurement of metal-ligand interactions at the single-molecule level remain a daunting challenge. Here we demonstrate an interdisciplinary and systematic approach that enables measurement and modulation of the coordinative bonding forces in a transition metal complex. Terpyridine is derived with a thiol linker, facilitating covalent attachment of this ligand on both gold substrate surfaces and gold-coated atomic force microscopy tips. The coordination and bond breaking between terpyridine and osmium are followed in situ by electrochemically controlled atomic force microscopy at the single-molecule level. The redox state of the central metal atom is found to have a significant impact on the metal-ligand interactions. The present approach represents a major advancement in unravelling the nature of metal-ligand interactions and could have broad implications in coordination chemistry.

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Electrochemical CO₂ and CO reduction on metal-functionalized porphyrin-like graphene

Porphyrin-like metal-functionalized graphene structures have been investigated as possible catalysts for CO₂ and CO reduction to methane or methanol. The late transition metals (Cu, Ag, Au, Ni, Pd, Pt, Co, Rh, Ir, Fe, Ru, Os) and some p (B, Al, Ga) and s (Mg) metals comprised the center of the porphyrin ring. A clear difference in catalytic properties compared to extended metal surfaces was observed owing to a different electronic nature of the active site. The preference to bind hydrogen, however, becomes a major obstacle in the reaction path. A possible solution to this problem is to reduce CO instead of CO₂. Volcano plots were constructed on the basis of scaling relations of reaction intermediates, and from these plots the reaction steps with the highest overpotentials were deduced. The Rh-porphyrin-like functionalized graphene was identified as the most active catalyst for producing methanol from CO, featuring an overpotential of 0.22 V. Additionally, we have also examined the hydrogen evolution and oxidation reaction, and in their case, too, Rh-porphyrin turned out to be the best catalyst with an overpotential of 0.15 V. © 2013 American Chemical Society.

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Contributors: Tripkovic, V., Vanin, M., Karamad, M., Björketun, M., Jacobsen, K. W., Thygesen, K. S., Rossmeisl, J.
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Energy level alignment and quantum conductance of functionalized metal-molecule junctions: Density functional theory versus GW calculations

We study the effect of functional groups (CH$_3$$^*$, OCH$_3$, CH$_3$, Cl, CN, F$^*$) on the electronic transport properties of 1,4-benzenediamine molecular junctions using the non-equilibrium Green function method. Exchange and correlation effects are included at various levels of theory, namely density functional theory (DFT), energy level-corrected DFT (DFT+$\Sigma$), Hartree-Fock and the many-body GW approximation. All methods reproduce the expected trends for the energy of the frontier orbitals according to the electron donating or withdrawing character of the substituent group. However, only the GW method predicts the correct ordering of the conductance amongst the molecules. The absolute GW (DFT) conductance is within a factor of two (three) of the experimental values. Correcting the DFT orbital energies by a simple physically motivated scissors operator, $\Sigma$, can bring the DFT conductances close to experiments, but does not improve on the relative ordering. We ascribe this to too strong pinning of the molecular energy levels to the metal Fermi level by DFT which suppresses the variation in orbital energy with functional group.
We present first-principles many-body calculations of the dielectric constant, quasiparticle band structure, and optical absorption spectrum of monolayer MoS$_2$ using a supercell approach. As the separation between the periodically repeated layers is increased, the dielectric function of the layer develops a strong $q$ dependence around $q = 0$. This implies that denser $k$-point grids are required to converge the band gap and exciton binding energies when large supercells are used. In the limit of infinite layer separation, here obtained using a truncated Coulomb interaction, a 45 x 45 $k$-point grid is needed to converge the $G(0)W(0)$ band gap and exciton energy to within 0.1 eV. We provide an extensive comparison with previous studies and explain agreement and variations in the results. It is demonstrated that too coarse $k$-point sampling and the interactions between the repeated layers have opposite effects on the band gap and exciton energy, leading to a fortuitous error cancellation in the previously published results.

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Plasmons in metallic monolayer and bilayer transition metal dichalcogenides

We study the collective electronic excitations in metallic single-layer and bilayer transition metal dichalcogenides (TMDCs) using time dependent density functional theory in the random phase approximation. For very small momentum transfers (below $q \approx 0.02 \, \text{Å}^{-1}$), the plasmon dispersion follows the $\sqrt{q}$ behavior expected for free electrons in two dimensions. For larger momentum transfer, the plasmon energy is significantly redshifted due to screening by interband transitions. At around $q \approx 0.1 \, \text{Å}^{-1}$, the plasmon enters the dissipative electron-hole continuum and the plasmon dispersions flatten out at an energy around 0.6–1.1 eV, depending on the material. Using bilayer NbSe$_2$ as example, we show that the plasmon modes of a bilayer structure take the form of symmetric and antisymmetric hybrids of the single-layer modes. The spatially antisymmetric mode is rather weak with a linear dispersion tending to zero for $q=0$, while the energy of the symmetric mode follows the single-layer mode dispersion with a slight blue shift.
Quantitatively accurate calculations of conductance and thermopower of molecular junctions

Thermopower measurements of molecular junctions have recently gained interest as a characterization technique that supplements the more traditional conductance measurements. Here we investigate the electronic conductance and thermopower of benzenediamine (BDA) and benzenedicarbonitrile (BDCN) connected to gold electrodes using first-principles calculations. We find excellent agreement with experiments for both molecules when exchange–correlation effects are described by the many-body GW approximation. In contrast, results from standard density functional theory (DFT) deviate from experiments by up to two orders of magnitude. The failure of DFT is particularly pronounced for the n-type BDCN junction due to the severe underestimation of the lowest unoccupied molecular orbital (LUMO). The quality of the DFT results can be improved by correcting the molecular energy levels for self-interaction errors and image charge effects. Finally, we show that the conductance and thermopower of the considered junctions are relatively insensitive to the metal–molecule bonding geometry. Our results demonstrate that electronic and thermoelectric properties of molecular junctions can be predicted from first-principles calculations when exchange–correlation effects are taken properly into account.

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Web of Science (2013): Impact factor 1.605
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We present a plane-wave implementation of the GW approximation within the projector augmented wave method code GPAW. The computed band gaps of ten bulk semiconductors and insulators deviate on average by 0.2eV (~5%) from the experimental values, the only exception being ZnO where the calculated band gap is around 1eV too low. Similar relative deviations are found for the ionization potentials of a test set of 32 small molecules. The importance of substrate screening for a correct description of quasiparticle energies and Fermi velocities in supported two-dimensional (2D) materials is illustrated by the case of graphene/h-BN interfaces. Due to the long-range Coulomb interaction between periodically repeated images, the use of a truncated interaction is found to be essential for obtaining converged results for 2D materials. For all systems studied, a plasmon-pole approximation is found to reproduce the full frequency results to within 0.2eV with a significant gain in computational speed. Throughout, we compare the GW results with different exact exchange-based approximations. For completeness, we provide a mathematically rigorous and physically transparent introduction to the notion of quasiparticle states.
Random phase approximation applied to solids, molecules, and graphene-metal interfaces: From van der Waals to covalent bonding

The random phase approximation (RPA) is attracting renewed interest as a universal and accurate method for first-principles total energy calculations. The RPA naturally accounts for long-range dispersive forces without compromising accuracy for short-range interactions making the RPA superior to semilocal and hybrid functionals in systems dominated by weak van der Waals or mixed covalent-dispersive interactions. In this work, we present plane-wave-based RPA calculations for a broad collection of systems with bond types ranging from strong covalent to van der Waals. Our main result is the RPA potential energy surfaces of graphene on the Cu(111), Ni(111), Co(0001), Pd(111), Pt(111), Ag(111), Au(111), and Al(111) metal surfaces, which represent archetypical examples of metal-organic interfaces. Comparison with semilocal density approximations and a nonlocal van der Waals functional show that only the RPA captures both the weak covalent and dispersive forces, which are equally important for these systems. We benchmark our implementation in the GPAW electronic structure code by calculating cohesive energies of graphite and a range of covalently bonded solids and molecules as well as the dissociation curves of H2 and H2+. These results show that the RPA with orbitals from the local density approximation suffers from delocalization errors and systematically underestimates covalent bond energies yielding similar or lower accuracy than the Perdew-Burke-Ernzerhof (PBE) functional for molecules and solids, respectively.

General information

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Web of Science (2016): Impact factor 3.836
Stability and bandgaps of layered perovskites for one- and two-photon water splitting

Direct production of hydrogen from water and sunlight requires stable and abundantly available semiconductors with well positioned band edges relative to the water red-ox potentials. We have used density functional theory (DFT) calculations...
to investigate 300 oxides and oxynitrides in the Ruddlesden–Popper phase of the layered perovskite structure. Based on screening criteria for the stability, bandgaps and band edge positions, we suggest 20 new materials for the light harvesting photo-electrode of a one-photon water splitting device and 5 anode materials for a two-photon device with silicon as photo-cathode. In addition, we explore a simple rule relating the bandgap of the perovskite to the number of octahedra in the layered structure and the B-metal ion. Finally, the quality of the GLLB-SC potential used to obtain the bandgaps, including the derivative discontinuity, is validated against $G_0W_0@LDA$ gaps for 20 previously identified oxides and oxynitrides in the cubic perovskite structure.

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Contributors: Castelli, I. E., García Lastra, J. M., Hüser, F., Thygesen, K. S., Jacobsen, K. W.
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Visualizing hybridized quantum plasmons in coupled nanowires: From classical to tunneling regime

We present full quantum-mechanical calculations of the hybridized plasmon modes of two nanowires at small separation, providing real-space visualization of the modes in the transition from the classical to the quantum tunneling regime. The plasmon modes are obtained as certain eigenfunctions of the dynamical dielectric function, which is computed using time-dependent density functional theory (TDDFT). For freestanding wires, the energy of both surface and bulk plasmon modes deviate from the classical result for low wire radii and high momentum transfer due to effects of electron spill-out, nonlocal response, and coupling to single-particle transitions. For the wire dimer, the shape of the hybridized plasmon modes are continuously altered with decreasing separation, and below 6 Å, the energy dispersion of the modes deviate from classical results due to the onset of weak tunneling. Below 2-3 Å separation, this mode is replaced by a charge-transfer plasmon, which blue shifts with decreasing separation in agreement with experiment and marks the onset of the strong tunneling regime.

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Organisations: Department of Physics, Theoretical Atomic-scale Physics, Department of Photonics Engineering, Structured Electromagnetic Materials, Center for Nanostructured Graphene, Technical University of Denmark
Contributors: Andersen, K., Jensen, K. L., Mortensen, N. A., Thygesen, K. S.
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Ab initio nonequilibrium quantum transport and forces with the real-space projector augmented wave method

We present an efficient implementation of a nonequilibrium Green's function method for self-consistent calculations of electron transport and forces in nanostructured materials. The electronic structure is described at the level of density functional theory using the projector augmented wave method to describe the ionic cores and an atomic orbital basis set for the valence electrons. External bias and gate voltages are treated in a self-consistent manner and the Poisson equation with appropriate boundary conditions is solved in real space. Contour integration of the Green's function and parallelization over k points and real space makes the code highly efficient and applicable to systems containing several hundreds of atoms. The method is applied to a number of different systems, demonstrating the effects of bias and gate voltages, multiterminal setups, nonequilibrium forces, and spin transport.
One of the possible solutions to the world’s rapidly increasing energy demand is the development of new photoelectrochemical cells with improved light absorption. This requires development of semiconductor materials which have appropriate bandgaps to absorb a large part of the solar spectrum at the same time as being stable in aqueous environments. Here we demonstrate an efficient, computational screening of relevant oxide and oxynitride materials based on electronic structure calculations resulting in the reduction of a vast space of 5400 different materials to only 15 promising candidates. The screening is based on an efficient and reliable way of calculating semiconductor band gaps. The outcome of the screening includes all already known successful materials of the types investigated plus some new ones which warrant further experimental investigation.
Conventional and acoustic surface plasmons on noble metal surfaces: a time-dependent density functional theory study

First-principles calculations of the conventional and acoustic surface plasmons (CSPs and ASPs) on the (111) surfaces of Cu, Ag, and Au are presented. The effect of s-d interband transitions on both types of plasmons is investigated by comparing results from the local density approximation and an orbital-dependent exchange-correlation (xc) potential that improves the position and width of the d bands. The plasmon dispersions calculated with the latter xc potential agree well with electron energy loss spectroscopy (EELS) experiments. For both the CSP and ASP, the same trend of Cu < Au < Ag is found for the plasmon energies and is attributed to the reduced screening by interband transitions from Cu, to Au and Ag. This trend for the ASP, however, contradicts a previous model prediction. While the ASP is seen as a weak feature in the EELS, it can be clearly identified in the static and dynamic dielectric band structure.

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Contributors: Yan, J., Jacobsen, K. W., Thygesen, K. S.
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Scopus rating (2012): CiteScore 3.57 SJR 3.173 SNIP 1.378
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The adiabatic connection fluctuation-dissipation theorem with the random phase approximation (RPA) has recently been applied with success to obtain correlation energies of a variety of chemical and solid state systems. The main merit of this approach is the improved description of dispersive forces while chemical bond strengths and absolute correlation energies are systematically underestimated. In this work we extend the RPA by including a parameter-free renormalized version of the adiabatic local-density (ALDA) exchange-correlation kernel. The renormalization consists of a (local) truncation of the ALDA kernel for wave vectors $q > 2k_F$, which is found to yield excellent results for the homogeneous electron gas. In addition, the kernel significantly improves both the absolute correlation energies and atomization energies of small molecules over RPA and ALDA. The renormalization can be straightforwardly applied to other adiabatic local kernels.
First-principles quantum transport modeling of thermoelectricity in single-molecule nanojunctions with graphene nanoribbon electrodes

We overview the nonequilibrium Green function combined with density functional theory (NEGF-DFT) approach to modeling of independent electronic and phononic quantum transport in nanoscale thermoelectrics with examples focused on a new class of devices where a single organic molecule is attached to two metallic zigzag graphene nanoribbons (ZGNRs) via highly transparent contacts. Such contacts make possible injection of evanescent wavefunctions from the ZGNR electrodes, so that their overlap within the molecular region generates a peak in the electronic transmission around the Fermi energy of the device. Additionally, the spatial symmetry properties of the transverse propagating states in the semi-infinite ZGNR electrodes suppress hole-like contributions to the thermopower. Thus optimized thermopower, together with diminished phonon thermal conductance in a ZGNR|molecule|ZGNR inhomogeneous heterojunctions, yields the thermoelectric figure of merit \( ZT \) per thousand integral 0.4 at room temperature with maximum \( ZT \) per thousand integral 3 reached at very low temperatures \( T \) per thousand integral 10 K (so that the latter feature could be exploited for thermoelectric cooling of, e.g., infrared sensors). The reliance on evanescent mode transport and symmetry of propagating states in the electrodes makes the electronic-transport-determined power factor in this class of devices largely insensitive to the type of sufficiently short organic molecule, which we demonstrate by showing that both 18-annulene and C10 molecule sandwiched by the two ZGNR electrodes yield similar thermopower. Thus, one can search for molecules that will further reduce the phonon thermal conductance (in the denominator of \( ZT \)) while keeping the electronic power factor (in the nominator of \( ZT \)) optimized. We also show how the often employed Brenner empirical interatomic potential for hydrocarbon systems fails to describe phonon transport in our single-molecule nanojunctions when contrasted with first-principles results obtained via NEGF-DFT methodology.

General information

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BFI (2018): BFI-level 1
Web of Science (2018): Indexed yes
BFI (2017): BFI-level 1
Scopus rating (2017): CiteScore 1.35 SJR 0.274 SNIP 0.716
Web of Science (2017): Impact factor 1.431
Web of Science (2017): Indexed yes
BFI (2016): BFI-level 1
Scopus rating (2016): CiteScore 1.55 SJR 0.431 SNIP 0.916
Web of Science (2016): Impact factor 1.526
BFI (2015): BFI-level 1
Scopus rating (2015): CiteScore 1.33 SJR 0.46 SNIP 0.68
Web of Science (2015): Impact factor 1.104
Image-charge-induced localization of molecular orbitals at metal-molecule interfaces: Self-consistent GW calculations

Quasiparticle (QP) wave functions, also known as Dyson orbitals, extend the concept of single-particle states to interacting electron systems. Here we employ many-body perturbation theory in the GW approximation to calculate the QP wave functions for a semiempirical model describing a pi-conjugated molecular wire in contact with a metal surface. We find that image charge effects pull the frontier molecular orbitals toward the metal surface, while orbitals with higher or lower energy are pushed away. This affects both the size of the energetic image charge shifts and the coupling of the individual orbitals to the metal substrate. Full diagonalization of the QP equation and, to some extent, self-consistency in the GW self-energy, is important to describe the effect, which is not captured by standard density functional theory or Hartree-Fock. These results should be important for the understanding and theoretical modeling of electron transport across metal-molecule interfaces.
BFI (2019): BFI-level 1
Web of Science (2019): Indexed yes
BFI (2018): BFI-level 1
Web of Science (2018): Indexed yes
BFI (2017): BFI-level 2
Scopus rating (2017): CiteScore 3.34 SJR 1.604 SNIP 1.04
Web of Science (2017): Impact factor 3.813
Web of Science (2017): Indexed yes
Scopus rating (2016): CiteScore 3.16 SJR 2.339 SNIP 1.151
Web of Science (2016): Impact factor 3.836
Web of Science (2016): Indexed yes
Scopus rating (2015): CiteScore 2.8 SJR 2.377 SNIP 1.13
Web of Science (2015): Impact factor 3.718
Web of Science (2015): Indexed yes
Scopus rating (2014): CiteScore 3.3 SJR 2.762 SNIP 1.316
Web of Science (2014): Impact factor 3.736
Web of Science (2014): Indexed yes
Scopus rating (2013): CiteScore 3.55 SJR 2.813 SNIP 1.326
Web of Science (2013): Impact factor 3.664
ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
Scopus rating (2012): CiteScore 3.57 SJR 3.173 SNIP 1.378
Web of Science (2012): Impact factor 3.767
ISI indexed (2012): ISI indexed yes
Web of Science (2012): Indexed yes
Scopus rating (2011): CiteScore 3.61 SJR 3.326 SNIP 1.423
Web of Science (2011): Impact factor 3.691
ISI indexed (2011): ISI indexed yes
Web of Science (2011): Indexed yes
Scopus rating (2010): SJR 3.318 SNIP 1.447
Web of Science (2010): Impact factor 3.774
Web of Science (2010): Indexed yes
Web of Science (2009): Indexed yes
Scopus rating (2008): SJR 2.923 SNIP 1.516
Web of Science (2008): Indexed yes
Scopus rating (2007): SJR 2.892 SNIP 1.588
Web of Science (2007): Indexed yes
Scopus rating (2006): SJR 2.62 SNIP 1.468
Web of Science (2006): Indexed yes
Scopus rating (2005): SJR 2.126 SNIP 1.156
Web of Science (2005): Indexed yes
Scopus rating (2004): SJR 2.012 SNIP 1.103
Web of Science (2004): Indexed yes
Scopus rating (2003): SJR 2.184 SNIP 1.179
Web of Science (2003): Indexed yes
Scopus rating (2002): SJR 2.856 SNIP 1.841
Web of Science (2002): Indexed yes
Scopus rating (2001): SJR 3.132 SNIP 1.727
Web of Science (2001): Indexed yes
Scopus rating (2000): SJR 2.84 SNIP 1.603
Web of Science (2000): Indexed yes
Scopus rating (1999): SJR 2.789 SNIP 1.541
Magnetoresistance and negative differential resistance in Ni/graphene/Ni vertical heterostructures driven by finite bias voltage: a first-principles study

Using the nonequilibrium Green's function formalism combined with density functional theory, we study finite bias quantum transport in Ni/Gr/Ni vertical heterostructures where n graphene layers are sandwiched between two semi-infinite Ni(111) electrodes. We find that the recently predicted "pessimistic" magnetoresistance of 100% for n≥5 junctions at zero bias voltage Vb→0 persists up to Vb≃0.4 V, which makes such devices promising for spin-torque-based device applications. In addition, for parallel orientations of the Ni magnetizations, the n=5 junction exhibits a pronounced negative differential resistance as the bias voltage is increased from Vb=0 V to Vb=0.5 V. We confirm that both of these nonequilibrium transport effects hold for different types of bonding of Gr on the Ni(111) surface while maintaining Bernal stacking between individual Gr layers.
New cubic perovskites for one- and two-photon water splitting using the computational materials repository

A new efficient photoelectrochemical cell (PEC) is one of the possible solutions to the energy and climate problems of our time. Such a device requires development of new semiconducting materials with tailored properties with respect to stability and light absorption. Here we perform computational screening of around 19 000 oxides, oxynitrides, oxyfluorides, and oxyfluoronitrides in the cubic perovskite structure with PEC applications in mind. We address three main applications: light absorbers for one- and two-photon water splitting and high-stability transparent shields to protect against corrosion. We end up with 20, 12, and 15 different combinations of oxides, oxynitrides and oxyfluorides, respectively, inviting further experimental investigation.

General information
State: Published
Organizations: Department of Physics, Theoretical Atomic-scale Physics, Center for Atomic-scale Materials Design, Experimental Surface and Nanomaterials Physics, Center for Individual Nanoparticle Functionality, Stanford University
Contributors: Castelli, I. E., Landis, D., Thygesen, K. S., Dahl, S., Chorkendorff, I., Jaramillo, T. F., Jacobsen, K. W.
Observation of quantum interference in molecular charge transport

As the dimensions of a conductor approach the nanoscale, quantum effects begin to dominate, and it becomes possible to control the conductance through direct manipulation of the electron wavefunction. Such control has been demonstrated in various mesoscopic devices at cryogenic temperatures(1-4), but it has proved to be difficult to exert control over the wavefunction at higher temperatures. Molecules have typical energy level spacings (similar to eV) that are much larger than the thermal energy at 300 K (similar to 25 meV), and are therefore natural candidates for such experiments. Previously, phenomena such as giant magnetoresistance(5), Kondo effects(6) and conductance switching(7-11) have been observed in single molecules, and theoreticians have predicted that it should also be possible to observe quantum interference in molecular conductors(12-18), but until now all the evidence for such behaviour has been indirect. Here, we report the observation of destructive quantum interference in charge transport through two-terminal molecular junctions at room temperature. We studied five different rigid p-conjugated molecular wires, all of which form self-assembled monolayers on a gold surface, and find that the degree of interference can be controlled by simple chemical modifications of the molecular wire.

General information
State: Published
Organisations: Department of Physics, Theoretical Atomic-scale Physics, Leiden University, University of Groningen
Contributors: Guedon, C. M., Valkenier, H., Markussen, T., Thygesen, K. S., Hummelen, J. C., van der Molen, S. J.
Pages: 304-308
Publication date: 2012
Peer-reviewed: Yes

Publication information
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Ratings:
BFI (2019): BFI-level 2
Web of Science (2019): Indexed yes
BFI (2018): BFI-level 2
Web of Science (2018): Indexed yes
BFI (2017): BFI-level 2
Scopus rating (2017): CiteScore 25.47 SJR 20.612 SNIP 8.171
Web of Science (2017): Impact factor 37.49
Web of Science (2017): Indexed yes
BFI (2016): BFI-level 2
Scopus rating (2016): CiteScore 21.85 SJR 18.916 SNIP 7.649
Web of Science (2016): Impact factor 38.986
Web of Science (2016): Indexed yes
BFI (2015): BFI-level 2
Scopus rating (2015): CiteScore 22.1 SJR 18.842 SNIP 8.019
Web of Science (2015): Impact factor 35.267
Web of Science (2015): Indexed yes
BFI (2014): BFI-level 2
Scopus rating (2014): CiteScore 21.76 SJR 17.177 SNIP 8.047
Web of Science (2014): Impact factor 34.048
Web of Science (2014): Indexed yes
BFI (2013): BFI-level 2
Scopus rating (2013): CiteScore 21.94 SJR 16.688 SNIP 7.784
Web of Science (2013): Impact factor 33.265
ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
BFI (2012): BFI-level 2
Scopus rating (2012): CiteScore 17.55 SJR 15.706 SNIP 7.569
Web of Science (2012): Impact factor 31.17
ISI indexed (2012): ISI indexed yes
Optical properties of bulk semiconductors and graphene/boron nitride: the Bethe-Salpeter equation with derivative discontinuity-corrected density functional energies

We present an efficient implementation of the Bethe-Salpeter equation (BSE) for optical properties of materials in the projector augmented wave method Grid-based projector-augmented wave method (GPAW). Single-particle energies and wave functions are obtained from the Gritsenko, Leeuwen, Lenthe, and Baerends potential [Phys. Rev. A51, 1944 (1995)] with the modifications from Kuisma et al. [Phys. Rev. B82, 115106 (2010)] GLLBSC functional which explicitly includes the derivative discontinuity, is computationally inexpensive, and yields excellent fundamental gaps. Electron-hole interactions are included through the BSE using the statically screened interaction evaluated in the random phase approximation. For a representative set of semiconductors and insulators we find excellent agreement with experiments for the dielectric functions, onset of absorption, and lowest excitonic features. For the two-dimensional systems of graphene and hexagonal boron-nitride (h-BN) we find good agreement with previous many-body calculations. For the graphene/h-BN interface we find that the fundamental and optical gaps of the h-BN layer are reduced by 2.0 and 0.7 eV, respectively, compared to freestanding h-BN. This reduction is due to image charge screening which shows up in the GLLBSC calculation as a reduction (vanishing) of the derivative discontinuity.

General information
State: Published
Organisations: Department of Physics, Theoretical Atomic-scale Physics, Center for Nanostructured Graphene
Contributors: Yan, J., Jacobsen, K. W., Thygesen, K. S.
Pages: 045208
Publication date: 2012
Peer-reviewed: Yes

Publication information
Journal: Physical Review B Condensed Matter
Volume: 86
Issue number: 4
ISSN (Print): 0163-1829
Ratings:
BFI (2019): BFI-level 1
Web of Science (2019): Indexed yes
BFI (2018): BFI-level 1
Web of Science (2018): Indexed yes
BFI (2017): BFI-level 2
Phonon-limited mobility in n-type single-layer MoS2 from first principles

We study the phonon-limited mobility in intrinsic n-type single-layer MoS2 for temperatures $T > 100$ K. The materials properties including the electron-phonon interaction are calculated from first principles and the deformation potentials and Frohlich interaction in single-layer MoS2 are established. The calculated room-temperature mobility of similar to 410 cm$^2$/V$\cdot$s$^{-1}$ is found to be dominated by optical phonon scattering via intra and intervalley deformation potential couplings and the Frohlich interaction. The mobility is weakly dependent on the carrier density and follows a $\mu \sim T^{-\gamma}$ temperature dependence with $\gamma = 1.69$ at room temperature. It is shown that a quenching of the characteristic homopolar mode, which is likely to occur in top-gated samples, increases the mobility with similar to 70 cm$^2$/V$\cdot$s$^{-1}$ and can be observed as a decrease in the exponent $\gamma = 1.52$. In comparison to recent experimental findings for the mobility in single-layer MoS2 (similar to 200 cm$^2$/V$\cdot$s$^{-1}$), our results indicate that mobilities close to the intrinsic phonon-limited mobility can be achieved in two-dimensional materials via dielectric engineering that effectively screens static Coulomb scattering on, e.g., charged impurities.
Spatially resolved quantum plasmon modes in metallic nano-films from first-principles

Electron energy loss spectroscopy (EELS) can be used to probe plasmon excitations in nanostructured materials with atomic-scale spatial resolution. For structures smaller than a few nanometers, quantum effects are expected to be important, limiting the validity of widely used semiclassical response models. Here we present a method to identify and compute spatially resolved plasmon modes from first-principles based on a spectral analysis of the dynamical dielectric function. As an example we calculate the plasmon modes of 0.5 to 4 nm thick Na films and find that they can be classified as (conventional) surface modes, subsurface modes, and a discrete set of bulk modes resembling standing waves across the film. We find clear effects of both quantum confinement and nonlocal response. The quantum plasmon modes provide an intuitive picture of collective excitations of confined electron systems and offer a clear interpretation of spatially resolved EELS spectra.
Unraveling the acoustic electron-phonon interaction in graphene

Using a first-principles approach we calculate the electron-phonon couplings in graphene for the transverse and longitudinal acoustic phonons. Analytic forms of the coupling matrix elements valid in the long-wavelength limit are found to give an almost quantitative description of the first-principles matrix elements even at shorter wavelengths. Using the analytic forms of the coupling matrix elements, we study the acoustic phonon-limited carrier mobility and quasiparticle lifetime observable in photoemission spectroscopy for temperatures 0-200 K and high carrier densities of 10^{12}-10^{13} cm^{-2}. We find that the intrinsic effective acoustic deformation potential of graphene is $\Xi_{\text{eff}}=6.8$ eV and that the temperature dependence of the mobility $\mu \sim T^{-\alpha}$ in the Bloch-Gruenisen regime increases beyond an $\alpha=4$ dependence even in the absence of screening when the true coupling matrix elements are considered. The $\alpha>4$ temperature dependence of the mobility is found to originate in a similar temperature dependence of the relaxation time at the Fermi level. The large disagreement between our calculated deformation potential and those extracted from experimental measurements (18-29 eV) indicates that additional or modified acoustic phonon-scattering mechanisms are at play in experimental situations.

General information

State: Published
Organisations: Department of Physics, Theoretical Atomic-scale Physics
Contributors: Kaasbjerg, K., Thygesen, K. S., Jacobsen, K. W.
Pages: 165440
Publication date: 2012
Peer-reviewed: Yes
Ab Initio Calculations of the Electronic Properties of Polypyridine Transition Metal Complexes and Their Adsorption on Metal Surfaces in the Presence of Solvent and Counterions

Os(II)/(III) and Co(II)/(III) polypyridine complexes in aqueous solution are robust molecular entities both in freely solute state and adsorbed on Au(111)- and Pt(111)-electrode surfaces. This class of robust coordination chemical compounds have recently been characterized by electrochemical scanning tunneling microscopy (in situ STM). The Os-complexes...
were found to display strong tunneling spectroscopic (STS) features at the level of resolution of the single molecule while STS features of the Co complexes, although clear, were much weaker. The data was framed by concise but phenomenological theory of interfacial electrochemical electron transfer extended to the electrochemical in situ STM configuration. With a view on first-principle insight into the in situ STM behavior of robust redox (as opposed to nonredox) molecules, we present in this report a density functional theory (DFT) study of the complexes in both free and adsorbate state, in either state exposed to both stoichiometric counterions and a large assembly of solvent water molecules. The oxidation states of the complexes were controlled, first by introducing chlorine counter atoms followed by spontaneous attraction of electrons from the complexes, also at first in electrostatically neutral form. Second, the solvent is found to provide strong dielectric screening of this charge transfer process and to be crucial for achieving the full chemically meaningful charge separated ionic oxidation states. The molecular charge and structure of the complexes in the presence of the solvent, are conserved upon adsorption, whereas the structural features of the different oxidation states are completely lost upon adsorption under vacuum conditions. Detailed microscopic insight such as offered by the present study will be important in molecular-based approaches to “smart” redox molecules enclosed in in situ STM or other nanoscale and single-molecules scale configurations in condensed matter environments.
The band structure and optical absorption spectrum of lithium peroxide (Li₂O₂) is calculated from first-principles using the G0W0 approximation and the Bethe-Salpeter equation, respectively. A strongly localized (Frenkel type) exciton corresponding to the π*→σ* transition on the O₂⁻² peroxide ion gives rise to a narrow absorption peak around 1.2 eV below the calculated bandgap of 4.8 eV. In the excited state, the internal O₂⁻² bond is significantly weakened due to the population of the σ* orbital. As a consequence, the bond is elongated by almost 0.5 Å leading to an extreme Stokes shift of 2.6 eV. The strong vibronic coupling entails significant broadening of the excitonic absorption peak in good agreement with diffuse reflectance data on Li₂O₂ which shows a rather featureless spectrum with an absorption onset around 3.0 eV. These results should be important for understanding the origin of the high potential losses and low current densities, which are presently limiting the performance of Li-air batteries.

**Communication: Strong excitonic and vibronic effects determine the optical properties of Li₂O₂**

The band structure and optical absorption spectrum of lithium peroxide (Li₂O₂) is calculated from first-principles using the G0W0 approximation and the Bethe-Salpeter equation, respectively. A strongly localized (Frenkel type) exciton corresponding to the π*→σ* transition on the O₂⁻² peroxide ion gives rise to a narrow absorption peak around 1.2 eV below the calculated bandgap of 4.8 eV. In the excited state, the internal O₂⁻² bond is significantly weakened due to the population of the σ* orbital. As a consequence, the bond is elongated by almost 0.5 Å leading to an extreme Stokes shift of 2.6 eV. The strong vibronic coupling entails significant broadening of the excitonic absorption peak in good agreement with diffuse reflectance data on Li₂O₂ which shows a rather featureless spectrum with an absorption onset around 3.0 eV. These results should be important for understanding the origin of the high potential losses and low current densities, which are presently limiting the performance of Li-air batteries.

**General information**

State: Published
Organisations: Theoretical Atomic-scale Physics, Department of Physics, IBM Research
Contributors: García Lastra, J. M., Bass, J. D., Thygesen, K. S.
Pages: -
Publication date: 2011
Dispersive and Covalent Interactions between Graphene and Metal Surfaces from the Random Phase Approximation

We calculate the potential energy surfaces for graphene adsorbed on Cu(111), Ni(111), and Co(0001) using density functional theory and the random phase approximation (RPA). For these adsorption systems covalent and dispersive interactions are equally important and while commonly used approximations for exchange-correlation functionals give inadequate descriptions of either van der Waals or chemical bonds, RPA accounts accurately for both. It is found that the adsorption is a delicate competition between a weak chemisorption minimum close to the surface and a physisorption minimum further from the surface.
Efficient electronic structure methods applied to metal nanoparticles

Nano-scale structures are increasingly applied in the design of catalysts and electronic devices. A theoretical understanding of the basic properties of such systems is enabled through modern electronic structure methods such as density functional theory. This thesis describes the development of efficient approaches to density functional theory and the application of these methods to metal nanoparticles.

We describe the formalism and implementation of localized atom-centered basis sets within the projector augmented wave method. Basis sets allow for a dramatic increase in performance compared to plane-wave or real-space methods, but sacrifice accuracy in doing so. This approach is implemented in the GPAW code where it complements the existing real-space approach. For both the real-space and basis set methods we implement parallel code to adapt GPAW for large-scale calculations on the BlueGene/P architecture. Real-space calculations are performed to investigate the convergence of chemical properties of Au and Pt clusters toward the bulk limit. Specifically we study chemisorption of O and CO on cuboctahedral clusters up to 1415 atoms using up to 65536 CPU cores. Small clusters almost universally bind more strongly than large ones. This can be understood mostly as a geometric effect. Convergence of chemisorption energies within 0.1 eV of bulk values happens at about 200 atoms for Pt and 600 atoms for Au. Particularly for O on Au, large variations due to electronic effects are seen for smaller clusters. The basis set method is used to study the electronic effects for the contiguous range of clusters up to several hundred atoms. The s-electrons hybridize to form electronic shells consistent with the jellium model, leading to electronic magic numbers for clusters with full shells. Large electronic gaps and jumps in Fermi level near magic numbers can lead to alkali-like or halogen-like behaviour when main-group atoms adsorb onto gold clusters. A non-self-consistent Newns-Anderson model is used to more closely study the chemisorption of main-group atoms on magic-number Au clusters. The behaviour at magic numbers can be understood from the location of adsorbate-induced states relative to the Fermi level. The relationship between geometric and electronic effects in Au is studied by rough first-principles simulated annealings with up to 150 atoms. Non-magic clusters are found to deform considerably, reducing the total energy through the creation of gaps. Clusters larger than 100 atoms can elongate systematically by up to 15 %. This demonstrates a complex interdependence between electronic and geometric structure in a size regime which in most cases has been studied semiempirically.

Electrical conductivity in Li2O2 and its role in determining capacity limitations in non-aqueous Li-O2 batteries

Non-aqueous Li-air or Li-O2 cells show considerable promise as a very high energy density battery couple. Such cells, however, show sudden death at capacities far below their theoretical capacity and this, among other problems, limits their practicality. In this paper, we show that this sudden death arises from limited charge transport through the growing Li2O2 film to the Li2O2-electrolyte interface, and this limitation defines a critical film thickness, above which it is not possible to support electrochemistry at the Li2O 2-electrolyte interface. We report both electrochemical experiments using a reversible internal redox couple and a first principles metal-insulator-metal charge transport model to probe the electrical conductivity through Li2O2 films produced during Li-O2 discharge. Both experiment and theory show a sudden death in charge transport when film thickness is ~5 to 10 nm. The theoretical model shows that this occurs when the tunneling current through the film can no longer support the electrochemical current. Thus, engineering charge transport through Li2O2 is a serious challenge if Li-O2 batteries are ever to reach their potential. © 2011 American Institute of Physics.
We use density functional theory (DFT) to investigate the electronic structure and chemical properties of gold nanoparticles. Different structural families of clusters are compared. For up to 60 atoms we optimize structures using DFT-based simulated annealing. Cluster geometries are found to distort considerably, creating large band gaps at the Fermi level. For up to 200 atoms we consider structures generated with a simple EMT potential and clusters based on cuboctahedra and icosahedra. All types of cluster geometry exhibit jelliumlike electronic shell structure. We calculate adsorption energies of several atoms on the cuboctahedral clusters. Adsorption energies are found to vary abruptly at magic numbers. Using a Newns-Anderson model we find that the effect of magic numbers on adsorption energy can be understood from the location of adsorbate-induced states with respect to the cluster Fermi level.

Electronic shell structure and chemisorption on gold nanoparticles

We use density functional theory (DFT) to investigate the electronic structure and chemical properties of gold nanoparticles. Different structural families of clusters are compared. For up to 60 atoms we optimize structures using DFT-based simulated annealing. Cluster geometries are found to distort considerably, creating large band gaps at the Fermi level. For up to 200 atoms we consider structures generated with a simple EMT potential and clusters based on cuboctahedra and icosahedra. All types of cluster geometry exhibit jelliumlike electronic shell structure. We calculate adsorption energies of several atoms on the cuboctahedral clusters. Adsorption energies are found to vary abruptly at magic numbers. Using a Newns-Anderson model we find that the effect of magic numbers on adsorption energy can be understood from the location of adsorbate-induced states with respect to the cluster Fermi level.

General information

State: Published
Organisations: Theoretical Atomic-scale Physics, Department of Physics, SLAC National Accelerator Laboratory
Pages: 245429
Publication date: 2011
Peer-reviewed: Yes

Publication information

Journal: Physical Review B Condensed Matter
Volume: 84
Issue number: 24
ISSN (Print): 0163-1829
Ratings:
BFI (2019): BFI-level 1
Finite Size Effects in Chemical Bonding: From Small Clusters to Solids

We address the fundamental question of which size a metallic nano-particle needs to have before its surface chemical properties can be considered to be those of a solid, rather than those of a large molecule. Calculations of adsorption energies for carbon monoxide and oxygen on a series of gold nanoparticles ranging from 13 to 1,415 atoms, or 0.8–3.7 nm, have been made possible by exploiting massively parallel computing on up to 32,768 cores on the Blue Gene/P computer at Argonne National Laboratory. We show that bulk surface properties are obtained for clusters larger than ca. 560 atoms (2.7 nm). Below that critical size, finite-size effects can be observed, and we show those to be related to variations in the local atomic structure augmented by quantum size effects for the smallest clusters.

General information
State: Published
Organisations: Department of Physics, Theoretical Atomic-scale Physics, Argonne National Laboratory, Technical University of Denmark
Pages: 1067-1071
Publication date: 2011
Peer-reviewed: Yes

Publication information
Journal: Catalysis Letters
Volume: 141
Issue number: 8
ISSN (Print): 1011-372X
Ratings:
BFI (2019): BFI-level 1
Web of Science (2019): Indexed yes
BFI (2018): BFI-level 1
Web of Science (2018): Indexed yes
BFI (2017): BFI-level 1
Scopus rating (2017): CiteScore 2.43 SJR 0.73 SNIP 0.729
Web of Science (2017): Impact factor 2.911
Web of Science (2017): Indexed yes
BFI (2016): BFI-level 1
Scopus rating (2016): CiteScore 2.39 SJR 0.755 SNIP 0.786
Web of Science (2016): Impact factor 2.799
BFI (2015): BFI-level 1
Scopus rating (2015): CiteScore 2.27 SJR 0.748 SNIP 0.749
Web of Science (2015): Impact factor 2.294
Web of Science (2015): Indexed yes
BFI (2014): BFI-level 1
Scopus rating (2014): CiteScore 2.56 SJR 0.89 SNIP 0.928
Web of Science (2014): Impact factor 2.307
Web of Science (2014): Indexed yes
BFI (2013): BFI-level 1
Scopus rating (2013): CiteScore 2.45 SJR 0.888 SNIP 0.931
Finite-size effects in surface chemistry of gold nanoparticles

General information
State: Published
Organisations: Department of Physics, Theoretical Atomic-scale Physics, Argonne National Laboratory, SLAC National Accelerator Laboratory
Pages: -
First-principles study of surface plasmons on Ag(111) and H/Ag(111)

Linear-response time-dependent density functional theory is used to investigate the relation between molecular bonding and surface plasmons for the model system H/Ag(111). We employ an orbital-dependent exchange-correlation functional to obtain a correct description of the Ag 3d band, which is crucial to avoid overscreening the plasmon by the s-d interband transitions. For the clean surface, this approach reproduces the experimental plasmon energies and dispersion to within 0.15 eV. Adsorption of hydrogen shifts and damps the Ag(111) surface plasmon and induces a new peak in the loss function at 0.6 eV below the Ag(111) plasmon peak. This feature originates from interband transitions between states located on the hydrogen atoms and states on the Ag surface atoms.

General information
State: Published
Organisations: Theoretical Atomic-scale Physics, Department of Physics
Contributors: Yan, J., Jacobsen, K. W., Thygesen, K. S.
Pages: 235430
Publication date: 2011
Peer-reviewed: Yes

Publication information
Graphical prediction of quantum interference-induced transmission nodes in functionalized organic molecules

Quantum interference (QI) in molecular transport junctions can lead to dramatic reductions of the electron transmission at certain energies. In a recent work [Markussen et al., Nano Lett., 2010, 10, 4260] we showed how the presence of such transmission nodes near the Fermi energy can be predicted solely from the structure of a conjugated molecule when the energies of the atomic pz orbitals do not vary too much. Here we relax the assumption of equal on-site energies and generalize the graphical scheme to molecules containing different atomic species. We use this diagrammatic scheme together with tight-binding and density functional theory calculations to investigate QI in linear molecular chains and aromatic molecules with different side groups. For the molecular chains we find a linear relation between the position of the transmission nodes and the side group π orbital energy. In contrast, the transmission functions of functionalized aromatic molecules generally display a rather complex nodal structure due to the interplay between molecular topology and the energy of the side group orbital.

General information
State: Published
Organisations: Theoretical Atomic-scale Physics, Department of Physics, University of Vienna
Contributors: Markussen, T., Stadler, R., Thygesen, K. S.
Pages: 14311
Publication date: 2011
Peer-reviewed: Yes

Publication information
Journal: Physical Chemistry Chemical Physics
Volume: 13
Issue number: 32
ISSN (Print): 1463-9076
Ratings:
BFI (2019): BFI-level 2
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BFI (2018): BFI-level 2
Web of Science (2018): Indexed yes
BFI (2017): BFI-level 2
Scopus rating (2017): CiteScore 4.04 SJR 1.686 SNIP 1.089
Web of Science (2017): Impact factor 3.906
Web of Science (2017): Indexed yes
BFI (2016): BFI-level 2
Scopus rating (2016): CiteScore 4.06 SJR 1.685 SNIP 1.113
Web of Science (2016): Impact factor 4.123
Web of Science (2016): Indexed yes
BFI (2015): BFI-level 2
Scopus rating (2015): CiteScore 4.45 SJR 1.725 SNIP 1.205
Web of Science (2015): Indexed yes
Improving transition voltage spectroscopy of molecular junctions

Transition voltage spectroscopy (TVS) is a promising spectroscopic tool for molecular junctions. The principles in TVS is to find the minimum on a Fowler-Nordheim plot where $\ln(I/V^2)$ is plotted against $1/V$ and relate the voltage at the minimum $V_{\text{min}}$ to the closest molecular level. Importantly, $V_{\text{min}}$ is approximately half the voltage required to see a peak in the $dI/dV$ curve. Information about the molecular level position can thus be obtained at relatively low voltages. In this work we show that the molecular level position can be determined at even lower voltages, $V_{\text{min}(\alpha)}$, by finding the minimum of $\ln(I/V^\alpha)$ with $\alpha$.

General information
State: Published
Organisations: Theoretical Atomic-scale Physics, Department of Physics
Contributors: Markussen, T., Chen, J., Thygesen, K. S.
Pages: 155407
Publication date: 2011
Peer-reviewed: Yes

Publication information
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Issue number: 15
ISSN (Print): 0163-1829
Ratings:
BFI (2019): BFI-level 1
Web of Science (2019): Indexed yes
BFI (2018): BFI-level 1
Web of Science (2018): Indexed yes
BFI (2017): BFI-level 2
Scopus rating (2017): CiteScore 3.34 SJR 1.604 SNIP 1.04
Web of Science (2017): Impact factor 3.813
Web of Science (2017): Indexed yes
Scopus rating (2016): CiteScore 3.16 SJR 2.339 SNIP 1.151
Web of Science (2016): Impact factor 3.836
Web of Science (2016): Indexed yes
Scopus rating (2015): CiteScore 2.8 SJR 2.377 SNIP 1.13
Web of Science (2015): Impact factor 3.718
Web of Science (2015): Indexed yes
Scopus rating (2014): CiteScore 3.3 SJR 2.762 SNIP 1.316
Web of Science (2014): Impact factor 3.736
Web of Science (2014): Indexed yes
Scopus rating (2013): CiteScore 3.55 SJR 2.813 SNIP 1.326
Web of Science (2013): Impact factor 3.664
ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
Scopus rating (2012): CiteScore 3.57 SJR 3.173 SNIP 1.378
Web of Science (2012): Impact factor 3.767
ISI indexed (2012): ISI indexed yes
Web of Science (2012): Indexed yes
Scopus rating (2011): CiteScore 3.61 SJR 3.326 SNIP 1.423
Web of Science (2011): Impact factor 3.691
ISI indexed (2011): ISI indexed yes
Web of Science (2011): Indexed yes
Scopus rating (2010): SJR 3.318 SNIP 1.447
Web of Science (2010): Impact factor 3.774
Web of Science (2010): Indexed yes
Linear density response function in the projector augmented wave method: Applications to solids, surfaces, and interfaces

We present an implementation of the linear density response function within the projector-augmented wave method with applications to the linear optical and dielectric properties of both solids, surfaces, and interfaces. The response function is represented in plane waves while the single-particle eigenstates can be expanded on a real space grid or in atomic-orbital basis for increased efficiency. The exchange-correlation kernel is treated at the level of the adiabatic local density approximation (ALDA) and crystal local field effects are included. The calculated static and dynamical dielectric functions of Si, C, SiC, AlP, and GaAs compare well with previous calculations. While optical properties of semiconductors, in particular excitonic effects, are generally not well described by ALDA, we obtain excellent agreement with experiments for the surface loss function of graphene and the Mg(0001) surface with plasmon energies deviating by less than 0.2 eV. Finally, the method is applied to study the influence of substrates on the plasmon excitations in graphene.

General information
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Organisations: Theoretical Atomic-scale Physics, Department of Physics
Contributors: Yan, J., Mortensen, J. J., Jacobsen, K. W., Thygesen, K. S.
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Web of Science (2019): Indexed yes
BFI (2018): BFI-level 1
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BFI (2017): BFI-level 2
Multiterminal single-molecule-graphene-nanoribbon junctions with the thermoelectric figure of merit optimized via evanescent mode transport and gate voltage

We propose thermoelectric devices where a single molecule is connected to two metallic zigzag graphene nanoribbons (ZGNRs) via highly transparent contacts that allow the injection of evanescent wave functions from ZGNRs. Their overlap generates a peak in the electronic transmission that is largely insensitive to the type of the short conjugated molecule, while ZGNRs additionally suppress hole-like contributions to the thermopower. Thus optimized thermopower, together with suppression of phonon transport through a ZGNR-molecule-ZGNR structure, yields the thermoelectric figure of merit $ZT$ similar to 0.5 at room temperature and 0.5 < $ZT$ < 2.5 below liquid nitrogen temperature. Using density functional theory combined with the nonequilibrium Green's function formalism for multiterminal devices, we show how the transmission resonance can be manipulated by the voltage applied to a third ZGNR top-gate electrode covering the molecule to further tune the value of $ZT$.

General information
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Organisations: Theoretical Atomic-scale Physics, Department of Physics
Contributors: Saha, K., Markussen, T., Thygesen, K. S., Nikolic, B.
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Web of Science (2019): Indexed yes
BFI (2018): BFI-level 1
Web of Science (2018): Indexed yes
BFI (2017): BFI-level 2
Scopus rating (2017): CiteScore 3.34 SJR 1.604 SNIP 1.04
Web of Science (2017): Impact factor 3.813
Web of Science (2017): Indexed yes
Scopus rating (2016): CiteScore 3.16 SJR 2.339 SNIP 1.151
Web of Science (2016): Impact factor 3.836
Web of Science (2016): Indexed yes
Scopus rating (2015): CiteScore 2.8 SJR 2.377 SNIP 1.13
Web of Science (2015): Impact factor 3.718
Web of Science (2015): Indexed yes
Scopus rating (2014): CiteScore 3.3 SJR 2.762 SNIP 1.316
Web of Science (2014): Impact factor 3.736
Web of Science (2014): Indexed yes
Scopus rating (2013): CiteScore 3.55 SJR 2.813 SNIP 1.326
Web of Science (2013): Impact factor 3.664
ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
Scopus rating (2012): CiteScore 3.57 SJR 3.173 SNIP 1.378
Web of Science (2012): Impact factor 3.767
ISI indexed (2012): ISI indexed yes
Web of Science (2012): Indexed yes
Scopus rating (2011): CiteScore 3.61 SJR 3.326 SNIP 1.423
Web of Science (2011): Impact factor 3.691
ISI indexed (2011): ISI indexed yes
Web of Science (2011): Indexed yes
Scopus rating (2010): SJR 3.318 SNIP 1.447
Web of Science (2010): Impact factor 3.774
Nonlocal Screening of Plasmons in Graphene by Semiconducting and Metallic Substrates: First-Principles Calculations

We investigate the role of substrates on the collective excitations of graphene by using a first-principles implementation of the density response function within the random-phase approximation. Specifically, we consider graphene adsorbed on SiC(0001) and Al(111) as representative examples of a semiconducting and metallic substrate. On SiC(0001), the long wavelength π plasmons are significantly damped although their energies remain almost unaltered. On Al(111), the long wavelength π plasmons are completely quenched due to the coupling to the metal surface plasmon. The strong damping of the plasmon excitations occurs despite the fact that the single-particle band structure of graphene is completely unaffected by the substrates illustrating the nonlocal nature of the effect.

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Organisations: Theoretical Atomic-scale Physics, Department of Physics
Contributors: Yan, J., Thygesen, K. S., Jacobsen, K. W.
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Web of Science (2018): Indexed yes
BFI (2017): BFI-level 2
Scopus rating (2017): CiteScore 7.58 SJR 3.622 SNIP 2.464
Web of Science (2017): Impact factor 8.839
Web of Science (2017): Indexed yes
BFI (2016): BFI-level 2
Scopus rating (2016): CiteScore 6.33 SJR 4.196 SNIP 2.61
Web of Science (2016): Impact factor 8.462
Web of Science (2016): Indexed yes
BFI (2015): BFI-level 2
Scopus rating (2015): CiteScore 5.76 SJR 4.656 SNIP 2.538
Web of Science (2015): Impact factor 7.645
Web of Science (2015): Indexed yes
BFI (2014): BFI-level 2
Scopus rating (2014): CiteScore 6.62 SJR 5.232 SNIP 2.71
Web of Science (2014): Impact factor 7.512
Web of Science (2014): Indexed yes
BFI (2013): BFI-level 2
Scopus rating (2013): CiteScore 7.46 SJR 5.675 SNIP 2.781
ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
BFI (2012): BFI-level 2
Scopus rating (2012): CiteScore 7.19 SJR 6.292 SNIP 2.867
ISI indexed (2012): ISI indexed yes
Web of Science (2012): Indexed yes
BFI (2011): BFI-level 2
Scopus rating (2011): CiteScore 7.02 SJR 6.314 SNIP 2.905
ISI indexed (2011): ISI indexed yes
Web of Science (2011): Indexed yes
BFI (2010): BFI-level 2
Scopus rating (2010): SJR 6.45 SNIP 2.757
Web of Science (2010): Indexed yes
BFI (2009): BFI-level 2
Scopus rating (2009): SJR 6.325 SNIP 2.947
Web of Science (2009): Indexed yes
BFI (2008): BFI-level 2
Scopus rating (2008): SJR 6.194 SNIP 2.837
Web of Science (2008): Indexed yes
Scopus rating (2007): SJR 5.95 SNIP 2.738
Web of Science (2007): Indexed yes
Scopus rating (2006): SJR 4.781 SNIP 2.443
Web of Science (2006): Indexed yes
Scopus rating (2005): SJR 4.082 SNIP 2.101
Web of Science (2005): Indexed yes
Scopus rating (2004): SJR 3.847 SNIP 2.122
Web of Science (2004): Indexed yes
Scopus rating (2003): SJR 4.661 SNIP 2.651
Web of Science (2003): Indexed yes
Web of Science (2002): Indexed yes
Scopus rating (2001): SJR 5.884 SNIP 3.375
Web of Science (2001): Indexed yes
Scopus rating (2000): SJR 5.618 SNIP 3.135
Web of Science (2000): Indexed yes
Scopus rating (1999): SJR 5.771 SNIP 2.941

Original language: English
Renormalization of Optical Excitations in Molecules near a Metal Surface

The lowest electronic excitations of benzene and a set of donor-acceptor molecular complexes are calculated for the gas phase and on the Al(111) surface using the many-body Bethe-Salpeter equation. The energy of the charge-transfer excitations obtained for the gas phase complexes are found to be around 10% lower than the experimental values. When the molecules are placed outside the surface, the enhanced screening from the metal reduces the exciton binding energies by several eVs and the transition energies by up to 1 eV depending on the size of the transition-generated dipole. As a striking consequence we find that close to the metal surface the optical gap of benzene can exceed its quasiparticle gap. A classical image charge model for the screened Coulomb interaction can account for all these effects which, on the other hand, are completely missed by standard time-dependent density functional theory.
Robust conductance of dumbbell molecular junctions with fullerene anchoring groups

The conductance of a molecular wire connected to metallic electrodes is known to be sensitive to the atomic structure of the molecule-metal contact. This contact is to a large extent determined by the anchoring group linking the molecular wire to the metal. It has been found experimentally that a dumbbell construction with C60 molecules acting as anchors yields more well-defined conductances as compared to the widely used thiol anchoring groups. Here, we use density functional theory to investigate the electronic properties of this dumbbell construction. The conductance is found to be stable against variations in the detailed bonding geometry and in good agreement with the experimental value of $G = 3 \times 10^{-4}G_0$. Electron tunneling across the molecular bridge occurs via the lowest unoccupied orbitals of C60 which are pinned close to the Fermi energy due to partial charge transfer. Our findings support the original motivation to achieve conductance values more stable towards changes in the structure of the molecule-metal contact leading to larger reproducibility in experiments. © 2011 American Institute of Physics.

General information

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Organisations: Theoretical Atomic-scale Physics, Department of Physics, Theoretical Microfluidics Group, Theory Section, Department of Micro- and Nanotechnology
Contributors: Markussen, T., Settnes, M., Thygesen, K. S.
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Peer-reviewed: Yes
Self-consistent GW calculations of electronic transport in thiol- and amine-linked molecular junctions

The electronic conductance of a benzene molecule connected to gold electrodes via thiol, thiolate, or amino anchoring groups is calculated using nonequilibrium Green functions in combination with the fully self-consistent GW approximation for exchange and correlation. The calculated conductance of benzenedithiol and benzenediamine is one-fifth that predicted by standard density functional theory (DFT), in very good agreement with experiments. In contrast, the widely studied benzenedithiolate structure is found to have a significantly higher conductance due to the unsaturated sulfur bonds. These findings suggest that more complex gold-thiolate structures where the thiolate anchors are chemically passivated by Au adatoms are responsible for the measured conductance. Analysis of the energy level alignment obtained with DFT, Hartree-Fock, and GW reveals the importance of self-interaction corrections (exchange) on the molecule and dynamical screening at the metal-molecule interface. The main effect of the GW self-energy is to renormalize the level positions; however, its influence on the shape of molecular resonances also affects the conductance. Non-self-consistent G(0)W(0) calculations, starting from either DFT or Hartree-Fock, yield conductance values within 50% of the self-consistent GW results.
The role of transition metal interfaces on the electronic transport in lithium–air batteries

Low electronic conduction is expected to be a main limiting factor in the performance of reversible lithium–air, Li–O2, batteries. Here, we apply density functional theory and non-equilibrium Green's function calculations to determine the electronic transport through lithium peroxide, Li2O2, formed at the cathode during battery discharge. We find the transport to depend on the orientation and lattice matching of the insulator–metal interface in the presence of Au and Pt catalysts. Bulk lithium vacancies are found to be available and mobile under battery charging conditions, and found to pin the Fermi level at the top of the anti bonding peroxide π*(2px) and π*(2py) levels in the Li2O2 valence band. Under an applied bias, this can result in a reduced transmission, since the anti bonding σ*(2pz) level in the Li2O2 conduction band is found to couple strongly to the metal substrate and create localized interface states with poor coupling to the Li2O2 bulk states. These observations provide a possible explanation for the higher overpotential observed for charging than discharge.
Towards quantitative accuracy in first-principles transport calculations: The GW method applied to alkane/gold junctions

The calculation of the electronic conductance of nanoscale junctions from first principles is a long-standing problem in the field of charge transport. Here we demonstrate excellent agreement with experiments for the transport properties of the gold/alkanediamine benchmark system when electron-electron interactions are described by the many-body GW approximation. The conductance follows an exponential length dependence: \( G_n = G_c \exp(-\beta n) \). The main difference from standard density functional theory (DFT) calculations is a significant reduction of the contact conductance, \( G_c \), due to an improved alignment of the molecular energy levels with the metal Fermi energy. The molecular orbitals involved in the tunneling process comprise states delocalized over the carbon backbone and states localized on the amine end groups. We find that dynamic screening effects renormalize the two types of states in qualitatively different ways when the molecule is inserted in the junction. Consequently, the GW transport results cannot be mimicked by DFT calculations employing a simple scissors operator. © 2011 Strange and Thygesen; licensee Beilstein-Institut.
Trends in Metal Oxide Stability for Nanorods, Nanotubes, and Surfaces

The formation energies of nanostructures play an important role in determining their properties, including their catalytic activity. For the case of 15 different rutile and 8 different perovskite metal oxides, we used density functional theory (DFT) to calculate the formation energies of (2,2) nanorods, (3,3) nanotubes, and the (110) and (100) surfaces. These formation energies can be described semiquantitatively (mean absolute error = 0.12 eV) by the fraction of metal–oxygen bonds broken and the metal d-band and p-band centers in the bulk metal oxide.
Benchmarking GW against exact diagonalization for semiempirical models

We calculate ground-state total energies and single-particle excitation energies of seven pi-conjugated molecules described with the semiempirical Pariser-Parr-Pople model using self-consistent many-body perturbation theory at the GW level and exact diagonalization. For the total energies GW captures around 65% of the ground-state correlation energy. The lowest lying excitations are overscreened by GW leading to an underestimation of electron affinities and ionization potentials by 0.15 eV on average corresponding to ~3%. One-shot G0W0 calculations starting from Hartree-Fock reduce the screening and improve the low-lying excitation energies. The effect of the GW self-energy on the molecular excitation energies is shown to be similar to the inclusion of final-state relaxations in Hartree-Fock theory. We discuss the breakdown of the GW approximation in systems with short-range interactions (Hubbard models) where correlation effects dominate over screening/relaxation effects. Finally we illustrate the important role of the derivative discontinuity of the true exchange-correlation functional by computing the exact Kohn-Sham levels of benzene.
We discuss the electrochemical reactions at the oxygen electrode of an aprotic Li-air battery. Using density functional theory to estimate the free energy of intermediates during the discharge and charge of the battery, we introduce a reaction free energy diagram and identify possible origins of the overpotential for both processes. We also address the question of
electron conductivity through the Li2O2 electrode and show that in the presence of Li vacancies Li2O2 becomes a conductor.

**General information**

**State:** Published

**Organisations:** Theoretical Atomic-scale Physics, Department of Physics, Nano-Microstructures in Materials, Materials Research Division, Risø National Laboratory for Sustainable Energy

**Contributors:** Hummelshøj, J. S., Blomquist, J., Datta, S., Vegge, T., Rossmeisl, J., Thygesen, K. S., Luntz, A. C., Jacobsen, K. W., Nørskov, J. K.

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BFI (2019): BFI-level 2

Web of Science (2019): Indexed yes

BFI (2018): BFI-level 2

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BFI (2017): BFI-level 2

Scopus rating (2017): CiteScore 2.5 SJR 1.252 SNIP 0.926

Web of Science (2017): Impact factor 2.843

Web of Science (2017): Indexed yes

BFI (2016): BFI-level 2

Scopus rating (2016): CiteScore 2.13 SJR 1.486 SNIP 0.964

Web of Science (2016): Impact factor 2.965

Web of Science (2016): Indexed yes

BFI (2015): BFI-level 2

Scopus rating (2015): CiteScore 1.98 SJR 1.255 SNIP 0.964

Web of Science (2015): Impact factor 2.894

Web of Science (2015): Indexed yes

BFI (2014): BFI-level 2

Scopus rating (2014): CiteScore 2.54 SJR 1.446 SNIP 1.02

Web of Science (2014): Impact factor 2.952

Web of Science (2014): Indexed yes

BFI (2013): BFI-level 2

Scopus rating (2013): CiteScore 2.95 SJR 1.559 SNIP 1.174

Web of Science (2013): Impact factor 3.122

ISI indexed (2013): ISI indexed yes

Web of Science (2013): Indexed yes

BFI (2012): BFI-level 2

Scopus rating (2012): CiteScore 2.86 SJR 1.832 SNIP 1.137

Web of Science (2012): Impact factor 3.164

ISI indexed (2012): ISI indexed yes

Web of Science (2012): Indexed yes

BFI (2011): BFI-level 2

Scopus rating (2011): CiteScore 3.07 SJR 1.845 SNIP 1.215

Web of Science (2011): Impact factor 3.333

ISI indexed (2011): ISI indexed yes

Web of Science (2011): Indexed yes

BFI (2010): BFI-level 2

Scopus rating (2010): SJR 1.777 SNIP 1.064

Web of Science (2010): Impact factor 2.921
Designing multifunctional chemical sensors using Ni and Cu doped carbon nanotubes

We demonstrate a "bottom up" approach to the computational design of a multifunctional chemical sensor. General techniques are employed for describing the adsorption coverage and resistance properties of the sensor based on density functional theory and non-equilibrium Green's function methodologies, respectively. Specifically, we show how Ni and Cu doped metallic (6,6) single-walled carbon nanotubes may work as effective multifunctional sensors for both CO and NH3.
Ratings:

BFI (2019): BFI-level 1
Web of Science (2019): Indexed yes
BFI (2018): BFI-level 1
Web of Science (2018): Indexed yes
BFI (2017): BFI-level 1
Scopus rating (2017): CiteScore 1.67 SJR 0.602 SNIP 0.786
Web of Science (2017): Impact factor 1.729
Web of Science (2017): Indexed yes
BFI (2016): BFI-level 1
Scopus rating (2016): CiteScore 1.53 SJR 0.96 SNIP 0.753
Web of Science (2016): Impact factor 1.674
BFI (2015): BFI-level 1
Scopus rating (2015): CiteScore 1.43 SJR 0.665 SNIP 0.721
Web of Science (2015): Impact factor 1.522
BFI (2014): BFI-level 1
Scopus rating (2014): CiteScore 1.53 SJR 0.805 SNIP 0.769
Web of Science (2014): Impact factor 1.469
BFI (2013): BFI-level 1
Scopus rating (2013): CiteScore 1.53 SJR 0.831 SNIP 0.776
Web of Science (2013): Impact factor 1.605
ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
BFI (2012): BFI-level 1
Scopus rating (2012): CiteScore 1.39 SJR 0.897 SNIP 0.719
Web of Science (2012): Impact factor 1.489
ISI indexed (2012): ISI indexed yes
BFI (2011): BFI-level 1
Scopus rating (2011): CiteScore 1.34 SJR 0.931 SNIP 0.723
Web of Science (2011): Impact factor 1.316
ISI indexed (2011): ISI indexed yes
Web of Science (2011): Indexed yes
BFI (2010): BFI-level 1
Scopus rating (2010): SJR 0.87 SNIP 0.714
Web of Science (2010): Impact factor 1.349
Web of Science (2010): Indexed yes
BFI (2009): BFI-level 1
Scopus rating (2009): SJR 0.848 SNIP 0.718
BFI (2008): BFI-level 1
Scopus rating (2008): SJR 0.846 SNIP 0.684
Scopus rating (2007): SJR 0.871 SNIP 0.725
Web of Science (2007): Indexed yes
Scopus rating (2006): SJR 0.741 SNIP 0.648
Web of Science (2006): Indexed yes
Scopus rating (2005): SJR 0.746 SNIP 0.607
Scopus rating (2004): SJR 0.82 SNIP 0.62
Web of Science (2004): Indexed yes
Scopus rating (2003): SJR 0.876 SNIP 0.757
Web of Science (2003): Indexed yes
Scopus rating (2002): SJR 0.8 SNIP 0.694
Web of Science (2002): Indexed yes
Scopus rating (2001): SJR 0.818 SNIP 0.614
Web of Science (2001): Indexed yes
Using first-principles calculations we analyze the electronic transport properties of a recently proposed anthraquinone-based electrochemical switch. Robust conductance on/off ratios of several orders of magnitude are observed due to destructive quantum interference present in the anthraquinone but absent in the hydroquinone molecular bridge. A simple explanation of the interference effect is achieved by transforming the frontier molecular orbitals into localized molecular orbitals thereby obtaining a minimal tight-binding model describing the transport in the relevant energy range in terms of hopping via the localized orbitals. The topology of the tight-binding model, which is dictated by the symmetries of the molecular orbitals, determines the amount of quantum interference.
Electronic structure calculations with GPAW: a real-space implementation of the projector augmented-wave method

Electronic structure calculations have become an indispensable tool in many areas of materials science and quantum chemistry. Even though the Kohn-Sham formulation of the density-functional theory (DFT) simplifies the many-body
problem significantly, one is still confronted with several numerical challenges. In this article we present the projector augmented-wave (PAW) method as implemented in the GPAW program package (https://wiki.fysik.dtu.dk/gpaw) using a uniform real-space grid representation of the electronic wavefunctions. Compared to more traditional plane wave or localized basis set approaches, real-space grids offer several advantages, most notably good computational scalability and systematic convergence properties. However, as a unique feature GPAW also facilitates a localized atomic-orbital basis set in addition to the grid. The efficient atomic basis set is complementary to the more accurate grid, and the possibility to seamlessly switch between the two representations provides great flexibility. While DFT allows one to study ground state properties, time-dependent density-functional theory (TDDFT) provides access to the excited states. We have implemented the two common formulations of TDDFT, namely the linear-response and the time propagation schemes. Electron transport calculations under finite-bias conditions can be performed with GPAW using non-equilibrium Green functions and the localized basis set. In addition to the basic features of the real-space PAW method, we also describe the implementation of selected exchange-correlation functionals, parallelization schemes, Delta SCF-method, x-ray absorption spectra, and maximally localized Wannier orbitals.

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BFI (2018): BFI-level 1
Web of Science (2018): Indexed yes
BFI (2017): BFI-level 1
Scopus rating (2017): CiteScore 2.37 SJR 0.875 SNIP 0.921
Web of Science (2017): Impact factor 2.617
Web of Science (2017): Indexed yes
BFI (2016): BFI-level 1
Scopus rating (2016): CiteScore 1.89 SJR 1.553 SNIP 0.91
Web of Science (2016): Impact factor 2.678
Web of Science (2016): Indexed yes
BFI (2015): BFI-level 1
Scopus rating (2015): CiteScore 1.65 SJR 1.043 SNIP 0.889
Web of Science (2015): Impact factor 2.209
Web of Science (2015): Indexed yes
BFI (2014): BFI-level 1
Scopus rating (2014): CiteScore 1.99 SJR 1.284 SNIP 0.987
Web of Science (2014): Impact factor 2.346
Web of Science (2014): Indexed yes
BFI (2013): BFI-level 1
Scopus rating (2013): CiteScore 2.11 SJR 1.326 SNIP 1.022
Web of Science (2013): Impact factor 2.223
ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
BFI (2012): BFI-level 1
Scopus rating (2012): CiteScore 2.33 SJR 1.688 SNIP 1.168
First-principles calculations of graphene nanoribbons in gaseous environments: Structural and electronic properties

The stability of graphene nanoribbons in the presence of typical atmospheric molecules is systematically investigated by means of density-functional theory. We calculate the edge formation free energy of five different edge configurations passivated by H, H-2, O, O-2, N-2, CO, CO2, and H2O, respectively. In addition to the well known hydrogen passivated armchair and zigzag edges, we find the edges saturated by oxygen atoms to be particularly stable under atmospheric conditions. Saturation of the zigzag edge by oxygen leads to the formation of metallic states strictly localized on the oxygen atoms. Finally, the vibrational spectrum of the hydrogen- and oxygen-passivated ribbons are calculated and compared.

General information
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Organisations: Theoretical Atomic-scale Physics, Department of Physics, Section for Structural Engineering, Department of Civil Engineering
Contributors: Vanin, M., Gath, J., Thygesen, K. S., Jacobsen, K. W.
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First-principles modelling of scanning tunneling microscopy using non-equilibrium Green's functions

The investigation of electron transport processes in nano-scale architectures plays a crucial role in the development of surface chemistry and nano-technology. Experimentally, an important driving force within this research area has been the concurrent refinements of scanning tunneling microscopy (STM) techniques. The theoretical treatment of the STM operation has traditionally been based on the Bardeen and Tersoff-Hamann methods which take as input the single-particle wave functions and eigenvalues obtained from finite cluster or slabs models of the surface-tip interface. Here, we present a novel STM simulation scheme based on non-equilibrium Green's functions (NEGF) and Wannier functions which is both accurate and very efficient. The main novelty of the scheme compared to the Bardeen and Tersoff-Hamann approaches is that the coupling to the infinite (macroscopic) electrodes is taken into account. As an illustrating example we apply the NEGF-STM method to the Si(001)(2x1):H surface with sub-surface P doping and discuss the results in comparison to the Bardeen and Tersoff-Hamann methods.
Fully self-consistent GW calculations for molecules

We calculate single-particle excitation energies for a series of 34 molecules using fully self-consistent GW, one-shot G0W0, Hartree-Fock (HF), and hybrid density-functional theory (DFT). All calculations are performed within the projector-augmented wave method using a basis set of Wannier functions augmented by numerical atomic orbitals. The GW self-energy is calculated on the real frequency axis including its full frequency dependence and off-diagonal matrix elements. The mean absolute error of the ionization potential (IP) with respect to experiment is found to be 4.4, 2.6, 0.8, 0.4, and 0.5 eV for DFT-PBE, DFT-PBE0, HF, G0W0[HF], and self-consistent GW, respectively. This shows that although electronic screening is weak in molecular systems, its inclusion at the GW level reduces the error in the IP by up to 50% relative to unscreened HF. In general GW overscreens the HF energies leading to underestimation of the IPs. The best IPs are obtained from one-shot G0W0 calculations based on HF since this reduces the overscreening. Finally, we find that the inclusion of core-valence exchange is important and can affect the excitation energies by as much as 1 eV.
Graphene on metals: A van der Waals density functional study

We use density functional theory (DFT) with a recently developed van der Waals density functional (vdW-DF) to study the adsorption of graphene on Co, Ni, Pd, Ag, Au, Cu, Pt, and Al(111) surfaces. In contrast to the local-density approximation (LDA) which predicts relatively strong binding for Ni, Co, and Pd, the vdW-DF predicts weak binding for all metals and metal-graphene distances in the range 3.40–3.72 Å. At these distances the graphene band structure as calculated with DFT and the many-body G0W0 method is basically unaffected by the substrate, in particular there is no opening of a band gap at the K point.
Modeling nanoscale gas sensors under realistic conditions: Computational screening of metal-doped carbon nanotubes

We use computational screening to systematically investigate the use of transition-metal-doped carbon nanotubes for chemical-gas sensing. For a set of relevant target molecules (CO, NH$_3$, and H$_2$S) and the main components of air (N$_2$, O$_2$, and H$_2$O), we calculate the binding energy and change in conductance upon adsorption on a metal atom occupying a vacancy of a (6,6) carbon nanotube. Based on these descriptors, we identify the most promising dopant candidates for detection of a given target molecule. From the fractional coverage of the metal sites in thermal equilibrium with air, we estimate the change in the nanotube resistance per doping site as a function of the target molecule concentration assuming charge transport in the diffusive regime. Our analysis points to Ni-doped nanotubes as candidates for CO sensors working under typical atmospheric conditions.

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BFI (2019): BFI-level 1
Web of Science (2019): Indexed yes
BFI (2018): BFI-level 1
Web of Science (2018): Indexed yes
BFI (2017): BFI-level 2
Scopus rating (2017): CiteScore 3.34 SJR 1.604 SNIP 1.04
Web of Science (2017): Impact factor 3.813
Web of Science (2017): Indexed yes
Scopus rating (2016): CiteScore 3.16 SJR 2.339 SNIP 1.151
Web of Science (2016): Impact factor 3.836
Quantifying transition voltage spectroscopy of molecular junctions: Ab initio calculations

Transition voltage spectroscopy (TVS) has recently been introduced as a spectroscopic tool for molecular junctions where it offers the possibility to probe molecular level energies at relatively low bias voltages. In this work we perform extensive ab initio calculations of the nonlinear current-voltage relations for a broad class of single-molecule transport junctions in order to assess the applicability and limitations of TVS. We find, that in order to fully utilize TVS as a quantitative spectroscopic tool, it is important to consider asymmetries in the coupling of the molecule to the two electrodes. When this is taken properly into account, the relation between the transition voltage and the energy of the molecular orbital closest to the Fermi level closely follows the trend expected from a simple, analytical model.

General information
State: Published
Organisations: Theoretical Atomic-scale Physics, Department of Physics
Contributors: Chen, J., Markussen, T., Thygesen, K. S.
Pages: 121412
Publication date: 2010
Peer-reviewed: Yes
The Relation between Structure and Quantum Interference in Single Molecule Junctions

Quantum interference (QI) of electron pathways has recently attracted increased interest as an enabling tool for single-molecule electronic devices. Although various molecular systems have been shown to exhibit QI effects and a number of methods have been proposed for its analysis, simple guidelines linking the molecular structure to QI effects in the phase-coherent transport regime have until now been lacking. In the present work we demonstrate that QI in aromatic molecules is intimately related to the topology of the molecule’s π system and establish a simple graphical scheme to predict the existence of QI-induced transmission antiresonances. The generality of the scheme, which is exact for a certain class of tight-binding models, is proved by a comparison to first-principles transport calculations for 10 different configurations of anthraquinone as well as a set of cross-conjugated molecular wires.

General information
State: Published
Organisations: Theoretical Atomic-scale Physics, Department of Physics, University of Vienna
Contributors: Markussen, T., Stadler, R., Thygesen, K. S.
Pages: 4260-4265
Publication date: 2010
Peer-reviewed: Yes

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Journal: Nano Letters
Volume: 10
Issue number: 10
ISSN (Print): 1530-6984
Ratings:
BFI (2019): BFI-level 2
Web of Science (2019): Indexed yes
BFI (2018): BFI-level 2
Web of Science (2018): Indexed yes
BFI (2017): BFI-level 2
Scopus rating (2017): CiteScore 13.07
Conductance of Conjugated Molecular Wires: Length Dependence, Anchoring Groups, and Band Alignment

The conductance of π-conjugated molecular wires bonded to gold electrodes at zero bias is studied using density functional theory combined with nonequilibrium Green’s function method. For all systems considered, we find that the conductance length dependence follows the simple exponential law characteristic of tunneling through a barrier, \( G = G_c \exp(-\beta L) \). For thiophene, pyrrole, and phenyl wires with thiol end-groups, we calculate decay constants \( \beta \) of 0.211,
0.257, and 0.264 Å⁻¹, respectively, and contact conductances (Gc) of 1.25, 2.90, and 1.22G₀, where G₀ = 2e²/h is the conductance quantum. In comparison, the corresponding values for amine-terminated thiophene are calculated to be β = 0.160 Å⁻¹ and Gc = 0.038G₀. These results show that (1) the contact resistance is mainly determined by the anchoring group and (2) the decay constant, which determines the conductance in the long wire limit, is not solely determined by the intrinsic band gap of the molecular wire but also depends on the anchoring group. This is because the alignment of the metal Fermi level with respect to the molecular levels is controlled by charge transfer and interface dipoles which in turn are determined by the local chemistry at the interface. Analysis of the charge transfer at the interface shows that the thiol-bonded molecules receive electrons from the Au electrodes while the amine-bonded molecules donate electrons to the Au electrodes.

**General information**

State: Published
Organisations: Department of Physics, Theoretical Atomic-scale Physics
Contributors: Peng, G., Strange, M., Thygesen, K. S., Mavrikakis, M.
Pages: 20967-20973
Publication date: 2009
Peer-reviewed: Yes

**Publication information**

Journal: Journal of Physical Chemistry Part C: Nanomaterials and Interfaces
Volume: 113
Issue number: 49
ISSN (Print): 1932-7447
Ratings:
- BFI (2019): BFI-level 1
- Web of Science (2019): Indexed yes
- BFI (2018): BFI-level 1
- Web of Science (2018): Indexed yes
- BFI (2017): BFI-level 1
- Scopus rating (2017): CiteScore 4.58 SJR 2.135 SNIP 1.147
- Web of Science (2017): Impact factor 4.484
- Web of Science (2017): Indexed yes
- BFI (2016): BFI-level 1
- Scopus rating (2016): CiteScore 4.48 SJR 1.964 SNIP 1.195
- Web of Science (2016): Impact factor 4.536
- Web of Science (2016): Indexed yes
- BFI (2015): BFI-level 1
- Scopus rating (2015): CiteScore 4.68 SJR 1.886 SNIP 1.26
- Web of Science (2015): Indexed yes
- BFI (2014): BFI-level 1
- Scopus rating (2014): CiteScore 5.08 SJR 2.032 SNIP 1.447
- Web of Science (2014): Impact factor 4.772
- Web of Science (2014): Indexed yes
- BFI (2013): BFI-level 1
- Scopus rating (2013): CiteScore 5.14 SJR 2.143 SNIP 1.445
- Web of Science (2013): Impact factor 4.835
- ISI indexed (2013): ISI indexed yes
- Web of Science (2013): Indexed yes
- BFI (2012): BFI-level 1
- Scopus rating (2012): CiteScore 4.98 SJR 2.529 SNIP 1.461
- Web of Science (2012): Impact factor 4.814
- ISI indexed (2012): ISI indexed yes
- Web of Science (2012): Indexed yes
- BFI (2011): BFI-level 1
- Scopus rating (2011): CiteScore 4.92 SJR 2.339 SNIP 1.465
- Web of Science (2011): Impact factor 4.805
- ISI indexed (2011): ISI indexed yes
Density functional theory based screening of ternary alkali-transition metal borohydrides: A computational material design project

We present a computational screening study of ternary metal borohydrides for reversible hydrogen storage based on density functional theory. We investigate the stability and decomposition of alloys containing 1 alkali metal atom, Li, Na, or K (M1); and 1 alkali, alkaline earth or 3d/4d transition metal atom (M2) plus two to five (BH4)− groups, i.e., M1M2(BH4)2–5, using a number of model structures with trigonal, tetrahedral, octahedral, and free coordination of the metal borohydride complexes. Of the over 700 investigated structures, about 20 were predicted to form potentially stable alloys with promising decomposition energies. The M1(Al/Mn/Fe)(BH4)4, (Li/Na)Zn(BH4)3, and (Na/K)(Ni/Co)(BH4)3 alloys are found to be the most promising, followed by selected M1(Nb/Rh)(BH4)4 alloys.

General information
State: Published
Organisations: Theoretical Atomic-scale Physics, Department of Physics, Nano-Microstructures in Materials, Materials Research Division, Risø National Laboratory for Sustainable Energy, Electroceramics, Fuel Cells and Solid State Chemistry Division, Department of Micro- and Nanotechnology, Theoretical Nanoelectronics Group, Theory Section, Experimental Surface and Nanomaterials Physics, Center for Individual Nanoparticle Functionality, Center for Nanoteknologi, Karlsruher Institut für Technologie, Seoul National University, University of South Florida, University of Jyväskylä, University College Cork, University of Crete, University of California at Berkeley, French Alternative Energies and Atomic Energy Commission, University of Oslo, Swiss Federal Laboratories for Materials Science and Technology (Empa), University of New Mexico, Technical University of Denmark, University of Milan, Trinity University, Japan Advanced Institute of Science and Technology, Paul Scherrer Institute, S.N. Bose National Centre for Basic Sciences, Croatian Physical Society, Aarhus University, KTH - Royal Institute of Technology, University of Valladolid, University of Iceland, University of Massachusetts, University of Virginia, Lomonosov Moscow State University, University of Illinois, Trinity College Dublin, Northwestern University, University of California, University of Wisconsin-Madison, Universidad Politécnica de Madrid, University of Trieste, Johann Wolfgang Goethe-Universität Frankfurt, Universidade Federal do ABC, Nanyang Technological University, Haldor Topsoe AS, Stanford University, Gheorghe Asachi Technical University of Iasi, University of Cincinnati, Universidad del Pais Vasco, Donostia International Physics Center (DIPC), Polish Academy of Sciences, Universidade de Sao Paulo
Hot-electron-assisted femtochemistry at surfaces: A time-dependent density functional theory approach

Using time-evolution time-dependent density functional theory (TDDFT) within the adiabatic local-density approximation, we study the interactions between single electrons and molecular resonances at surfaces. Our system is a nitrogen molecule adsorbed on a ruthenium surface. The surface is modeled at two levels of approximation, first as a simple external potential and later as a 20-atom cluster. We perform a number of calculations on an electron hitting the adsorbed molecule from inside the surface and establish a picture, where the resonance is being probed by the hot electron. This enables us to extract the position of the resonance energy through a fitting procedure. It is demonstrated that with the model we can extract several properties of the system, such as the presence of resonance peaks, the time electrons stay on the molecule before returning to the surface when hitting a molecular resonance and the lowering of the resonance energy due to an image charge effect. Finally we apply the TDDFT procedure to only consider the decay of molecular excitations and find that it agrees quite well with the width of the projected density of Kohn-Sham states.
Inelastic scattering in metal-H$_2$-metal junctions

We present first-principles calculations of the dI/dV characteristics of an H$_2$ molecule sandwiched between Au and Pt electrodes in the presence of electron-phonon interactions. The conductance is found to decrease by a few percentages at threshold voltages corresponding to the excitation energy of longitudinal vibrations of the H$_2$ molecule. In the case of Pt electrodes, the transverse vibrations can mediate transport through otherwise nontransmitting Pt d channels leading to an increase in the differential conductance even though the hydrogen junction is characterized predominately by a single almost fully open transport channel. In the case of Au, the transverse modes do not affect the dI/dV because the Au d states are too far below the Fermi level. A simple explanation of the first-principles results is given using scattering theory. Finally, we compare and discuss our results in relation to experimental data.
Influence of O-2 and N-2 on the conductivity of carbon nanotube networks

In the article, we have performed experiments on single-wall carbon nanotube (SWNT) networks and compared with density-functional theory (DFT) calculations to identify the microscopic origin of the observed sensitivity of the network conductivity to physisorbed O-2 and N-2. Previous DFT calculations of the transmission function for isolated pristine SWNTs have found physisorbed molecules have little influence on their conductivity. However, by calculating the four-terminal transmission function of crossed SWNT junctions, we show that physisorbed O-2 and N-2 do affect the junction's conductance. This may be understood as an increase in tunneling probability due to hopping via molecular orbitals. We find the effect is...
substantially larger for O-2 than for N-2, and for semiconducting rather than metallic SWNTs junctions, in agreement with experiment.

General information
State: Published
Organisations: Theoretical Atomic-scale Physics, Department of Physics
Contributors: Mowbray, D., Morgan, C., Thygesen, K. S.
Pages: 195431
Publication date: 2009
Peer-reviewed: Yes

Publication information
Journal: Physical Review B Condensed Matter
Volume: 79
Issue number: 19
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Ratings:
BFI (2019): BFI-level 1
Web of Science (2019): Indexed yes
BFI (2018): BFI-level 1
Web of Science (2018): Indexed yes
BFI (2017): BFI-level 2
Scopus rating (2017): CiteScore 3.34 SJR 1.604 SNIP 1.04
Web of Science (2017): Impact factor 3.813
Web of Science (2017): Indexed yes
Scopus rating (2016): CiteScore 3.16 SJR 2.339 SNIP 1.151
Web of Science (2016): Impact factor 3.836
Web of Science (2016): Indexed yes
Scopus rating (2015): CiteScore 2.8 SJR 2.377 SNIP 1.13
Web of Science (2015): Impact factor 3.718
Web of Science (2015): Indexed yes
Scopus rating (2014): CiteScore 3.3 SJR 2.762 SNIP 1.316
Web of Science (2014): Impact factor 3.736
Web of Science (2014): Indexed yes
Scopus rating (2013): CiteScore 3.55 SJR 2.813 SNIP 1.326
Web of Science (2013): Impact factor 3.664
ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
Scopus rating (2012): CiteScore 3.57 SJR 3.173 SNIP 1.378
Web of Science (2012): Impact factor 3.767
ISI indexed (2012): ISI indexed yes
Web of Science (2012): Indexed yes
Scopus rating (2011): CiteScore 3.61 SJR 3.326 SNIP 1.423
Web of Science (2011): Impact factor 3.691
ISI indexed (2011): ISI indexed yes
Web of Science (2011): Indexed yes
Scopus rating (2010): SJR 3.318 SNIP 1.447
Web of Science (2010): Impact factor 3.774
Web of Science (2010): Indexed yes
Web of Science (2009): Indexed yes
Scopus rating (2008): SJR 2.923 SNIP 1.516
Web of Science (2008): Indexed yes
Scopus rating (2007): SJR 2.892 SNIP 1.588
Web of Science (2007): Indexed yes
Scopus rating (2006): SJR 2.62 SNIP 1.468
Localized atomic basis set in the projector augmented wave method

We present an implementation of localized atomic-orbital basis sets in the projector augmented wave (PAW) formalism within the density-functional theory. The implementation in the real-space GPAW code provides a complementary basis set to the accurate but computationally more demanding grid representation. The possibility to switch seamlessly between the two representations implies that simulations employing the local basis can be fine tuned at the end of the calculation by switching to the grid, thereby combining the strength of the two representations for optimal performance. The implementation is tested by calculating atomization energies and equilibrium bulk properties of a variety of molecules and solids, comparing to the grid results. Finally, it is demonstrated how a grid-quality structure optimization can be performed with significantly reduced computational effort by switching between the grid and basis representations.

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Organisations: Theoretical Atomic-scale Physics, Department of Physics
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Journal: Physical Review B Condensed Matter
Volume: 80
Issue number: 19
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Ratings:
  BFI (2019): BFI-level 1
  Web of Science (2019): Indexed yes
  BFI (2018): BFI-level 1
  Web of Science (2018): Indexed yes
  BFI (2017): BFI-level 2
  Scopus rating (2017): CiteScore 3.34 SJR 1.604 SNIP 1.04
  Web of Science (2017): Impact factor 3.813
Polarization-induced renormalization of molecular levels at metallic and semiconducting surfaces

On the basis of first-principles G0W0 calculations we systematically study how the electronic levels of a benzene molecule are renormalized by substrate polarization when physisorbed on different metallic and semiconducting surfaces. The polarization-induced reduction in the energy gap between occupied and unoccupied molecular levels is found to scale with the substrate density of states at the Fermi level (for metals) and substrate band gap (for semiconductors). These conclusions are further supported by self-consistent GW calculations on simple lattice models. By expressing the electron self-energy in terms of the substrate’s joint density of states we relate the level shift to the surface electronic structure, thus providing a microscopic explanation of the trends in the GW and G0W0 calculations. While image charge effects are not captured by semilocal and hybrid exchange-correlation functionals, we find that error cancellations lead to remarkably good agreement between the G0W0 and Kohn-Sham energies for the occupied orbitals of the adsorbed molecule.
Renormalization of Molecular Quasiparticle Levels at Metal-Molecule Interfaces: Trends across Binding Regimes

When an electron or a hole is added into an orbital of an adsorbed molecule the substrate electrons will rearrange in order to screen the added charge. This polarization effect reduces the electron addition and removal energies of the adsorbed molecule relative to those of the free molecule. Using a microscopic model of the metal-molecule interface, we illustrate the basic features of this renormalization mechanism through systematic GW, Hartree-Fock, and Kohn-Sham calculations for the molecular energy levels as function of the model parameters. We identify two different polarization mechanisms: (i) polarization of the metal (image charge formation) and (ii) polarization of the molecule via charge transfer across the interface. The importance of (i) and (ii) is found to increase with the metal density of states at the Fermi level and metal-molecule coupling strength, respectively.

**General information**

State: Published
Organisations: Theoretical Atomic-scale Physics, Department of Physics, Center for Nanoteknologi, Universidad del Pais Vasco
Contributors: Thygesen, K. S., Rubio, A.
Pages: 046802
Publication date: 2009
Peer-reviewed: Yes
Stability and Electronic Properties of TiO2 Nanostructures With and Without B and N Doping

We address one of the main challenges to TiO2 photocatalysis, namely band gap narrowing, by combining nanostructural changes with doping. With this aim we compare TiO2's electronic properties for small 0D clusters, 1D nanorods and nanotubes, 2D layers, and 3D surface and bulk phases using different approximations within density functional theory and GW calculations. In particular, we propose very small (R \( \leq 5 \) Å) but surprisingly stable nanotubes with promising properties. The nanotubes are initially formed from TiO2 layers with the PtO2 structure, with the smallest (2,2) nanotube relaxing to a rutile nanorod structure. We find that quantum confinement effects, as expected, generally lead to a widening of the energy gap. However, substitutional doping with boron or nitrogen is found to give rise to (meta-)stable structures and the introduction of dopant and midgap states which effectively reduce the band gap. Boron is seen to always give rise to n-type doping while depending on the local bonding geometry, nitrogen may give rise to n-type or p-type doping. For undercoordinated TiO2 surface structures found in clusters, nanorods, nanotubes, layers and surfaces nitrogen gives rise to acceptor states while for larger clusters and bulk structures donor states are introduced.

General information
State: Published
Organisations: Theoretical Atomic-scale Physics, Department of Physics, Center for Nanoteknologi
Contributors: Mowbray, D., Martinez, J. I., Garcia Lastra, J. M., Thygesen, K. S., Jacobsen, K. W.
Pages: 12301-12308
Publication date: 2009
Peer-reviewed: Yes
Anomalous conductance oscillations and half-metallicity in atomic Ag-O chains

Using spin density functional theory, we study the electronic and magnetic properties of atomically thin, suspended chains containing silver and oxygen atoms in an alternating sequence. Chains longer than 4 atoms develop a half-metallic ground state implying fully spin-polarized charge carriers. The conductances of the chains exhibit weak even-odd oscillations.
around an anomalously low value of 0.1$G(0)$ ($G(0) = 2e^2/h$) which coincide with the averaged experimental conductance in the long chain limit. The unusual conductance properties are explained in terms of a resonating-chain model, which takes the reflection probability and phase shift of a single bulk-chain interface as the only input. The model also explains the conductance oscillations for other metallic chains.

**General information**
State: Published
Organisations: Theoretical Atomic-scale Physics, Department of Physics, Center for Nanoteknologi
Pages: 096804
Publication date: 2008
Peer-reviewed: Yes

**Publication information**
Journal: Physical Review Letters
Volume: 101
Issue number: 9
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Ratings:
BFI (2019): BFI-level 2
Web of Science (2019): Indexed yes
BFI (2018): BFI-level 2
Web of Science (2018): Indexed yes
BFI (2017): BFI-level 2
Scopus rating (2017): CiteScore 7.58 SJR 3.622 SNIP 2.464
Web of Science (2017): Impact factor 8.839
Web of Science (2017): Indexed yes
BFI (2016): BFI-level 2
Scopus rating (2016): CiteScore 6.33 SJR 4.196 SNIP 2.61
Web of Science (2016): Impact factor 8.462
Web of Science (2016): Indexed yes
BFI (2015): BFI-level 2
Scopus rating (2015): CiteScore 5.76 SJR 4.656 SNIP 2.538
Web of Science (2015): Impact factor 7.645
Web of Science (2015): Indexed yes
BFI (2014): BFI-level 2
Scopus rating (2014): CiteScore 6.62 SJR 5.232 SNIP 2.71
Web of Science (2014): Impact factor 7.512
Web of Science (2014): Indexed yes
BFI (2013): BFI-level 2
Scopus rating (2013): CiteScore 7.46 SJR 5.675 SNIP 2.781
ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
BFI (2012): BFI-level 2
Scopus rating (2012): CiteScore 7.19 SJR 6.292 SNIP 2.867
ISI indexed (2012): ISI indexed yes
Web of Science (2012): Indexed yes
BFI (2011): BFI-level 2
Scopus rating (2011): CiteScore 7.02 SJR 6.314 SNIP 2.905
ISI indexed (2011): ISI indexed yes
Web of Science (2011): Indexed yes
BFI (2010): BFI-level 2
Scopus rating (2010): SJR 6.45 SNIP 2.757
Web of Science (2010): Indexed yes
BFI (2009): BFI-level 2
Scopus rating (2009): SJR 6.325 SNIP 2.947
Web of Science (2009): Indexed yes
BFI (2008): BFI-level 2
Scopus rating (2008): SJR 6.194 SNIP 2.837
Web of Science (2008): Indexed yes
Scopus rating (2007): SJR 5.95 SNIP 2.738
Web of Science (2007): Indexed yes
Scopus rating (2006): SJR 4.781 SNIP 2.443
Web of Science (2006): Indexed yes
Scopus rating (2005): SJR 4.082 SNIP 2.101
Web of Science (2005): Indexed yes
Scopus rating (2004): SJR 3.847 SNIP 2.122
Web of Science (2004): Indexed yes
Scopus rating (2003): SJR 4.661 SNIP 2.651
Web of Science (2003): Indexed yes
Web of Science (2002): Indexed yes
Scopus rating (2001): SJR 5.884 SNIP 3.375
Web of Science (2001): Indexed yes
Scopus rating (2000): SJR 5.618 SNIP 3.135
Web of Science (2000): Indexed yes
Scopus rating (1999): SJR 5.771 SNIP 2.941
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Research output: Research - peer-review › Journal article – Annual report year: 2008

Benchmark density functional theory calculations for nanoscale conductance
We present a set of benchmark calculations for the Kohn-Sham elastic transmission function of five representative single-molecule junctions. The transmission functions are calculated using two different density functional theory methods, namely an ultrasoft pseudopotential plane-wave code in combination with maximally localized Wannier functions and the norm-conserving pseudopotential code SIESTA which applies an atomic orbital basis set. All calculations have been converged with respect to the supercell size and the number of k(parallel to) points in the surface plane. For all systems we find that the SIESTA transmission functions converge toward the plane-wave result as the SIESTA basis is enlarged. Overall, we find that an atomic basis with double zeta and polarization is sufficient (and in some cases, even necessary) to ensure quantitative agreement with the plane-wave calculation. We observe a systematic downshift of the SIESTA transmission functions relative to the plane-wave results. The effect diminishes as the atomic orbital basis is enlarged; however, the convergence can be rather slow.

General information
State: Published
Organisations: Theoretical Atomic-scale Physics, Department of Physics, Center for Nanoteknologi
Contributors: Strange, M., Bækgaard, I. S. B., Thygesen, K. S., Jacobsen, K. W.
Pages: 114714
Publication date: 2008
Peer-reviewed: Yes

Publication information
Journal: Journal of Chemical Physics
Volume: 128
Issue number: 11
ISSN (Print): 0021-9606
Ratings:
BFI (2019): BFI-level 2
Web of Science (2019): Indexed yes
BFI (2018): BFI-level 2
Web of Science (2018): Indexed yes
BFI (2017): BFI-level 2
Scopus rating (2017): CiteScore 2.5 SJR 1.252 SNIP 0.926
Web of Science (2017): Impact factor 2.843
Web of Science (2017): Indexed yes
BFI (2016): BFI-level 2
Scopus rating (2016): CiteScore 2.13 SJR 1.486 SNIP 0.964
Web of Science (2016): Impact factor 2.965
Web of Science (2016): Indexed yes
BFI (2015): BFI-level 2
Scopus rating (2015): CiteScore 1.98 SJR 1.255 SNIP 0.964
Web of Science (2015): Impact factor 2.894
Web of Science (2015): Indexed yes
BFI (2014): BFI-level 2
Scopus rating (2014): CiteScore 2.54 SJR 1.446 SNIP 1.02
Web of Science (2014): Impact factor 2.952
Web of Science (2014): Indexed yes
BFI (2013): BFI-level 2
Scopus rating (2013): CiteScore 2.95 SJR 1.559 SNIP 1.174
Web of Science (2013): Impact factor 3.122
ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
BFI (2012): BFI-level 2
Scopus rating (2012): CiteScore 2.86 SJR 1.832 SNIP 1.137
Web of Science (2012): Impact factor 3.164
ISI indexed (2012): ISI indexed yes
Web of Science (2012): Indexed yes
BFI (2011): BFI-level 2
Scopus rating (2011): CiteScore 3.07 SJR 1.845 SNIP 1.215
Web of Science (2011): Impact factor 3.333
ISI indexed (2011): ISI indexed yes
Web of Science (2011): Indexed yes
BFI (2010): BFI-level 2
Scopus rating (2010): SJR 1.777 SNIP 1.064
Web of Science (2010): Impact factor 2.921
Web of Science (2010): Indexed yes
BFI (2009): BFI-level 2
Scopus rating (2009): SJR 2.04 SNIP 1.119
Web of Science (2009): Indexed yes
BFI (2008): BFI-level 2
Scopus rating (2008): SJR 2.27 SNIP 1.144
Web of Science (2008): Indexed yes
Scopus rating (2007): SJR 2.245 SNIP 1.13
Web of Science (2007): Indexed yes
Scopus rating (2006): SJR 2.254 SNIP 1.287
Web of Science (2006): Indexed yes
Scopus rating (2005): SJR 2.323 SNIP 1.349
Web of Science (2005): Indexed yes
Scopus rating (2004): SJR 2.272 SNIP 1.39
Comparative study of anchoring groups for molecular electronics: structure and conductance of Au-S-Au and Au-NH2-Au junctions

The electrical properties of single-molecule junctions, consisting of an organic molecule coupled to metal electrodes, are sensitive to the detailed atomic structure of the molecule-metal contact. This, in turn, is determined by the anchoring group linking the molecule to the metal. With the aim of identifying and comparing the intrinsic properties of two commonly used anchoring groups, namely thiol and amine groups, we have calculated the atomic structure and conductance traces of different Au-S-Au and Au-NH2-Au nanojunctions using density functional theory (DFT). Whereas NH2 shows a strong structural selectivity towards atop-gold configurations, S shows large variability in its bonding geometries. As a result, the conductance of the Au-NH2-Au junction is less sensitive to the structure of the gold contacts than the Au-S-Au junction. These findings support recent experiments which show that amine-bonded molecules exhibit more well-defined conductance properties than do thiol-bonded molecules.
Web of Science (2017): Indexed yes
BFI (2016): BFI-level 1
Scopus rating (2016): CiteScore 1.89 SJR 1.553 SNIP 0.91
Web of Science (2016): Impact factor 2.678
Web of Science (2016): Indexed yes
BFI (2015): BFI-level 1
Scopus rating (2015): CiteScore 1.65 SJR 1.043 SNIP 0.889
Web of Science (2015): Impact factor 2.209
Web of Science (2015): Indexed yes
BFI (2014): BFI-level 1
Scopus rating (2014): CiteScore 1.99 SJR 1.284 SNIP 0.987
Web of Science (2014): Impact factor 2.346
Web of Science (2014): Indexed yes
BFI (2013): BFI-level 1
Scopus rating (2013): CiteScore 2.11 SJR 1.326 SNIP 1.022
Web of Science (2013): Impact factor 2.223
ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
BFI (2012): BFI-level 1
Scopus rating (2012): CiteScore 2.33 SJR 1.688 SNIP 1.168
Web of Science (2012): Impact factor 2.355
ISI indexed (2012): ISI indexed yes
Web of Science (2012): Indexed yes
BFI (2011): BFI-level 1
Scopus rating (2011): CiteScore 2.31 SJR 1.66 SNIP 1.161
Web of Science (2011): Impact factor 2.546
ISI indexed (2011): ISI indexed yes
BFI (2010): BFI-level 1
Scopus rating (2010): SJR 1.663 SNIP 1.054
Web of Science (2010): Impact factor 2.332
Web of Science (2010): Indexed yes
BFI (2009): BFI-level 1
Scopus rating (2009): SJR 1.525 SNIP 1.015
Web of Science (2009): Indexed yes
BFI (2008): BFI-level 1
Scopus rating (2008): SJR 1.467 SNIP 1.071
Web of Science (2008): Indexed yes
Scopus rating (2007): SJR 1.561 SNIP 1.143
Web of Science (2007): Indexed yes
Scopus rating (2006): SJR 1.689 SNIP 1.229
Web of Science (2006): Indexed yes
Scopus rating (2005): SJR 1.725 SNIP 1.174
Web of Science (2005): Indexed yes
Scopus rating (2004): SJR 1.587 SNIP 1.19
Web of Science (2004): Indexed yes
Scopus rating (2003): SJR 1.464 SNIP 1.06
Web of Science (2003): Indexed yes
Scopus rating (2002): SJR 1.364 SNIP 1.223
Web of Science (2002): Indexed yes
Scopus rating (2001): SJR 1.376 SNIP 1.045
Web of Science (2001): Indexed yes
Scopus rating (2000): SJR 1.411 SNIP 1.093
Web of Science (2000): Indexed yes
Conductance of Sidewall-Functionalized Carbon Nanotubes: Universal Dependence on Adsorption Sites

We use density functional theory to study the effect of molecular adsorbates on the conductance of metallic carbon nanotubes (CNT). The five molecules considered (NO2, NH2, H, COOH, OH) lead to very similar scattering of the electrons. The adsorption of a single molecule suppresses one of the two available transport channels at the Fermi level while the other is left undisturbed. If more molecules are adsorbed on the same sublattice, the remaining open channel may or may not be blocked, depending on the relative position of the adsorbates. If the relative positions satisfy a simple geometric condition, this channel remains fully open independently of the number of adsorbed molecules.
Conserving GW scheme for nonequilibrium quantum transport in molecular contacts

We give a detailed presentation of our recent scheme to include correlation effects in molecular transport calculations using the nonequilibrium Keldysh formalism. The scheme is general and can be used with any quasiparticle self-energy, but for practical reasons, we mainly specialize to the so-called GW self-energy, widely used to describe the quasiparticle band structures and spectroscopic properties of extended and low-dimensional systems. We restrict the GW self-energy to a finite, central region containing the molecule, and we describe the leads by density functional theory (DFT). A minimal basis of maximally localized Wannier functions is applied both in the central GW region and the leads. The importance of using a conserving, i.e., fully self-consistent, GW self-energy is demonstrated both analytically and numerically. We introduce an effective spin-dependent interaction which automatically reduces self-interaction errors to all orders in the interaction. The scheme is applied to the Anderson model in and out of equilibrium. In equilibrium at zero temperature, we find that GW describes the Kondo resonance fairly well for intermediate interaction strengths. Out of equilibrium, we demonstrate that the one-shot G(0)W(0) approximation can produce severe errors, in particular, at high bias. Finally, we consider a benzene molecule between featureless leads. It is found that the molecule's highest occupied molecular orbital-
lowest unoccupied molecular orbital gap as calculated in GW is significantly reduced as the coupling to the leads is increased, reflecting the more efficient screening in the strongly coupled junction. For the I-V characteristics of the junction, we find that Hartree-Fock (HF) and G(0)W(0) [G(HF)] yield results closer to GW than does DFT and G(0)W(0)[G(DFT)]. This is explained in terms of self-interaction effects and lifetime reduction due to electron-electron interactions.
Impact of exchange-correlation effects on the IV characteristics of a molecular junction

The role of exchange-correlation effects in nonequilibrium quantum transport through molecular junctions is assessed by analyzing the IV curve of a generic two-level model using self-consistent many-body perturbation theory (second Born and GW approximations) on the Keldysh contour. It is demonstrated how the variation of the molecule's energy levels with the bias voltage can produce anomalous peaks in the dI/dV curve. This effect is suppressed by electronic self-interactions and is therefore underestimated in standard transport calculations based on density functional theory. Inclusion of dynamic correlations introduces quasiparticle (QP) scattering which in turn broadens the molecular resonances. The broadening increases strongly with bias and can have a large impact on the calculated IV characteristic.

General information
State: Published
Organisations: Theoretical Atomic-scale Physics, Department of Physics, Center for Nanoteknologi
Contributors: Thygesen, K. S.
Pages: 166804
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Peer-reviewed: Yes

Publication information
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ISSN (Print): 0031-9007
Ratings:
BFI (2019): BFI-level 2
Web of Science (2019): Indexed yes
BFI (2018): BFI-level 2
Web of Science (2018): Indexed yes
Influence of functional groups on charge transport in molecular junctions

Using density functional theory (DFT), we analyze the influence of five classes of functional groups, as exemplified by NO2, OCH3, CH3, CCl3, and I, on the transport properties of a 1,4-benzenedithiolate (BDT) and 1,4-benzenediamine (BDA) molecular junction with gold electrodes. Our analysis demonstrates how ideas from functional group chemistry may be used to engineer a molecule's transport properties, as was shown experimentally and using a semiempirical model for BDA [Nano Lett. 7, 502 (2007)]. In particular, we show that the qualitative change in conductance due to a given functional group can be predicted from its known electronic effect (whether it is sigma/pi donating/withdrawing). However, the influence of functional groups on a molecule's conductance is very weak, as was also found in the BDA experiments. The calculated DFT conductances for the BDA species are five times larger than the experimental values, but good agreement is obtained after correcting for self-interaction and image charge effects. (c) 2008 American Institute of Physics.
Quantum conductance of 4,4-bipyridine molecular junctions: Role of electrode work function and local d band

We present density-functional theory calculations for the geometry and conductance of 4,4-bipyridine (BPD) nanojunctions with Au and Pt electrodes. The fact that transport takes place via bipyridine's lowest unoccupied molecular orbital (LUMO) suggests that the Au-BPD junction should have larger conductance than the Pt-BPD junction due to the smaller work function of Au as compared to Pt. On the other hand, coupling to the local d band is stronger in the case of Pt and this broadens the LUMO resonance. We find that these effects largely outbalance each other leading to conductances of 0.01G(0) and 0.02G(0) for the Au and Pt contacts, respectively (G(0)=2e(2)/h is the conductance quantum). The effect of coupling to the electrodes is investigated by means of the group orbital which makes precise the concept of the local band. The construction allows us to explain and rationalize the first-principles results within a simple single-level model.
Nonequilibrium GW approach to quantum transport in nano-scale contacts
General information
State: Published
Organisations: University of the Basque Country, Free University of Berlin
Contributors: Thygesen, K. S., Rubio, A.
Pages: 091101
Publication date: 2007
Peer-reviewed: Yes

Publication information
Journal: Journal of Chemical Physics
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ISSN (Print): 0021-9606
Ratings:
BFI (2019): BFI-level 2
Web of Science (2019): Indexed yes
BFI (2018): BFI-level 2
Web of Science (2018): Indexed yes
BFI (2017): BFI-level 2
Scopus rating (2017): CiteScore 2.5 SJR 1.252 SNIP 0.926
Web of Science (2017): Impact factor 2.843
Web of Science (2017): Indexed yes
BFI (2016): BFI-level 2
Scopus rating (2016): CiteScore 2.13 SJR 1.486 SNIP 0.964
Web of Science (2016): Impact factor 2.965
Web of Science (2016): Indexed yes
BFI (2015): BFI-level 2
Scopus rating (2015): CiteScore 1.98 SJR 1.255 SNIP 0.964
Web of Science (2015): Impact factor 2.894
Web of Science (2015): Indexed yes
BFI (2014): BFI-level 2
Scopus rating (2014): CiteScore 2.54 SJR 1.446 SNIP 1.02
Web of Science (2014): Impact factor 2.952
Web of Science (2014): Indexed yes
BFI (2013): BFI-level 2
Scopus rating (2013): CiteScore 2.95 SJR 1.559 SNIP 1.174
Web of Science (2013): Impact factor 3.122
ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
BFI (2012): BFI-level 2
Scopus rating (2012): CiteScore 2.86 SJR 1.832 SNIP 1.137
Web of Science (2012): Impact factor 3.164
ISI indexed (2012): ISI indexed yes
Web of Science (2012): Indexed yes
BFI (2011): BFI-level 2
Scopus rating (2011): CiteScore 3.07 SJR 1.845 SNIP 1.215
Web of Science (2011): Impact factor 3.333
ISI indexed (2011): ISI indexed yes
Web of Science (2011): Indexed yes
BFI (2010): BFI-level 2
Scopus rating (2010): SJR 1.777 SNIP 1.064
Web of Science (2010): Impact factor 2.921
Web of Science (2010): Indexed yes
BFI (2009): BFI-level 2
Scopus rating (2009): SJR 2.04 SNIP 1.119
Web of Science (2009): Indexed yes
Electron transport in a Pt-CO-Pt nanocontact: Density functional theory calculations

We have performed first-principles calculations for the mechanic and electric properties of pure Pt nanocontacts and a Pt contact with a single CO molecule adsorbed. For the pure Pt contacts we see a clear difference between point contacts and short chains in good agreement with experiments. We identify a tilted bridge configuration for the Pt-CO-Pt contact, which is stable and has a conductance close to 0.5G(0) (G(0)=2 e(2)/h), and we propose that this structure is responsible for an observed peak at 0.5G(0) in the conductance histogram for Pt exposed to a CO gas. We explain the main features of the transmission function for the Pt-CO-Pt contact, and show that the conductance is largely determined by the local d band at the Pt apex atoms.

General information
State: Published
Organisations: Department of Physics, Theoretical Atomic-scale Physics, Center for Nanoteknologii
Contributors: Strange, M., Thygesen, K. S., Jacobsen, K. W.
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Scopus rating (2017): CiteScore 3.34 SJR 1.604 SNIP 1.04
Web of Science (2017): Impact factor 3.813
Web of Science (2017): Indexed yes
Scopus rating (2016): CiteScore 3.16 SJR 2.339 SNIP 1.151
Web of Science (2016): Impact factor 3.836
Web of Science (2016): Indexed yes
Scopus rating (2015): CiteScore 2.8 SJR 2.377 SNIP 1.13
Web of Science (2015): Impact factor 3.718
Web of Science (2015): Indexed yes
Scopus rating (2014): CiteScore 3.3 SJR 2.762 SNIP 1.316
Web of Science (2014): Impact factor 3.736
Web of Science (2014): Indexed yes
Scopus rating (2013): CiteScore 3.55 SJR 2.813 SNIP 1.326
Web of Science (2013): Impact factor 3.664
ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
Scopus rating (2012): CiteScore 3.57 SJR 3.173 SNIP 1.378
Web of Science (2012): Impact factor 3.767
ISI indexed (2012): ISI indexed yes
Web of Science (2012): Indexed yes
Scopus rating (2011): CiteScore 3.61 SJR 3.326 SNIP 1.423
Web of Science (2011): Impact factor 3.691
ISI indexed (2011): ISI indexed yes
Web of Science (2011): Indexed yes
Scopus rating (2010): SJR 3.318 SNIP 1.447
Web of Science (2010): Impact factor 3.774
Web of Science (2010): Indexed yes
Web of Science (2009): Indexed yes
Scopus rating (2008): SJR 2.923 SNIP 1.516
Web of Science (2008): Indexed yes
Scopus rating (2007): SJR 2.892 SNIP 1.588
Web of Science (2007): Indexed yes
Scopus rating (2006): SJR 2.62 SNIP 1.468
Web of Science (2006): Indexed yes
Scopus rating (2005): SJR 2.126 SNIP 1.156
Web of Science (2005): Indexed yes
Scopus rating (2004): SJR 2.012 SNIP 1.103
Web of Science (2004): Indexed yes
Scopus rating (2003): SJR 2.184 SNIP 1.179
Web of Science (2003): Indexed yes
Scopus rating (2002): SJR 2.856 SNIP 1.841
Web of Science (2002): Indexed yes
Scopus rating (2001): SJR 3.132 SNIP 1.727
Web of Science (2001): Indexed yes
Scopus rating (2000): SJR 2.84 SNIP 1.603
Web of Science (2000): Indexed yes
Scopus rating (1999): SJR 2.789 SNIP 1.541
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Bibliographical note
Copyright 2006 American Physical Society
An ab initio study of electron transport through nitrobenzene: The influence of leads and contacts

General information
State: Published
Organisations: Department of Physics
Contributors: Stadler, R., Thygesen, K. S., Jacobsen, K. W.
Pages: S155-S160
Publication date: 2005
Peer-reviewed: Yes

Publication information
Journal: Nanotechnology
Volume: 16
ISSN (Print): 0957-4484
Ratings:
BFI (2019): BFI-level 1
Web of Science (2019): Indexed yes
BFI (2018): BFI-level 1
Web of Science (2018): Indexed yes
BFI (2017): BFI-level 2
Scopus rating (2017): CiteScore 3.01 SJR 1.079 SNIP 0.788
Web of Science (2017): Impact factor 3.404
Web of Science (2017): Indexed yes
BFI (2016): BFI-level 2
Scopus rating (2016): CiteScore 2.87 SJR 1.339 SNIP 0.945
Web of Science (2016): Impact factor 3.44
Web of Science (2016): Indexed yes
BFI (2015): BFI-level 2
Scopus rating (2015): CiteScore 3.07 SJR 1.257 SNIP 1.035
Web of Science (2015): Impact factor 3.573
Web of Science (2015): Indexed yes
BFI (2014): BFI-level 2
Scopus rating (2014): CiteScore 3.09 SJR 1.497 SNIP 1.269
Web of Science (2014): Impact factor 3.821
Web of Science (2014): Indexed yes
BFI (2013): BFI-level 2
Scopus rating (2013): CiteScore 2.74 SJR 1.602 SNIP 1.231
Web of Science (2013): Impact factor 3.672
ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
BFI (2012): BFI-level 2
Scopus rating (2012): CiteScore 3.34 SJR 1.861 SNIP 1.307
Web of Science (2012): Impact factor 3.842
ISI indexed (2012): ISI indexed yes
Web of Science (2012): Indexed yes
BFI (2011): BFI-level 1
Scopus rating (2011): CiteScore 3.86 SJR 1.899 SNIP 1.451
Web of Science (2011): Impact factor 3.979
ISI indexed (2011): ISI indexed yes
Web of Science (2011): Indexed yes
BFI (2010): BFI-level 1
Scopus rating (2010): SJR 1.844 SNIP 1.252
Conduction Mechanism in a Molecular Hydrogen Contact

We present first principles calculations for the conductance of a hydrogen molecule bridging a pair of Pt electrodes. The transmission function has a wide plateau with $T \approx 1$ which extends across the Fermi level and indicates the existence of a single, robust conductance channel with nearly perfect transmission. Through a detailed Wannier function analysis we show that the H-2 bonding state is not involved in the transport and that the plateau forms due to strong hybridization between the H-2 antibonding state and states on the adjacent Pt atoms. The Wannier functions furthermore allow us to derive a resonant-level model for the system with all parameters determined from the fully self-consistent Kohn-Sham Hamiltonian.

General information

State: Published
Organisations: Theoretical Atomic-scale Physics, Department of Physics
Contributors: Thygesen, K. S., Jacobsen, K. W.
Pages: 036807
Publication date: 2005
Peer-reviewed: Yes

Publication information

Journal: Physical Review Letters
Volume: 94
Issue number: 3
ISSN (Print): 0031-9007
Ratings:
BFI (2019): BFI-level 2
Web of Science (2019): Indexed yes
BFI (2018): BFI-level 2
Web of Science (2018): Indexed yes
BFI (2017): BFI-level 2
Scopus rating (2017): CiteScore 7.58 SJR 3.622 SNIP 2.464
Web of Science (2017): Impact factor 8.839
Web of Science (2017): Indexed yes
BFI (2016): BFI-level 2
Scopus rating (2016): CiteScore 6.33 SJR 4.196 SNIP 2.61
Web of Science (2016): Impact factor 8.462
Web of Science (2016): Indexed yes
BFI (2015): BFI-level 2
Scopus rating (2015): CiteScore 5.76 SJR 4.656 SNIP 2.538
Web of Science (2015): Impact factor 7.645
Web of Science (2015): Indexed yes
BFI (2014): BFI-level 2
Scopus rating (2014): CiteScore 6.62 SJR 5.232 SNIP 2.71
Web of Science (2014): Impact factor 7.512
Web of Science (2014): Indexed yes
BFI (2013): BFI-level 2
Scopus rating (2013): CiteScore 7.46 SJR 5.675 SNIP 2.781
ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
BFI (2012): BFI-level 2
Scopus rating (2012): CiteScore 7.19 SJR 6.292 SNIP 2.867
ISI indexed (2012): ISI indexed yes
Web of Science (2012): Indexed yes
BFI (2011): BFI-level 2
Scopus rating (2011): CiteScore 7.02 SJR 6.314 SNIP 2.905
ISI indexed (2011): ISI indexed yes
Web of Science (2011): Indexed yes
BFI (2010): BFI-level 2
Scopus rating (2010): SJR 6.45 SNIP 2.757
Web of Science (2010): Indexed yes
BFI (2009): BFI-level 2
Scopus rating (2009): SJR 6.325 SNIP 2.947
Web of Science (2009): Indexed yes
BFI (2008): BFI-level 2
Scopus rating (2008): SJR 6.194 SNIP 2.837
Web of Science (2008): Indexed yes
Scopus rating (2007): SJR 5.95 SNIP 2.738
Web of Science (2007): Indexed yes
Scopus rating (2006): SJR 4.781 SNIP 2.443
Web of Science (2006): Indexed yes
Scopus rating (2005): SJR 4.082 SNIP 2.101
Web of Science (2005): Indexed yes
Scopus rating (2004): SJR 3.847 SNIP 2.122
Web of Science (2004): Indexed yes
Scopus rating (2003): SJR 4.661 SNIP 2.651
Web of Science (2003): Indexed yes
Web of Science (2002): Indexed yes
Scopus rating (2001): SJR 5.884 SNIP 3.375
Web of Science (2001): Indexed yes
Scopus rating (2000): SJR 5.618 SNIP 3.135
Web of Science (2000): Indexed yes
Scopus rating (1999): SJR 5.771 SNIP 2.941
Original language: English
Forces and conductances in a single-molecule bipyridine junction

Inspired by recent measurements of forces and conductances of bipyridine nanojunctions, we have performed density functional theory calculations of structure and electron transport in a bipyridine molecule attached between gold electrodes for seven different contact geometries. The calculations show that both the bonding force and the conductance are sensitive to the surface structure, and that both properties are in good agreement with experiment for contact geometries characterized by intermediate coordination of the metal atoms corresponding to a stepped surface. The conductance is mediated by the lowest unoccupied molecular orbital, which can be illustrated by a quantitative comparison with a one-level model. Implications for the interpretation of the experimentally determined force and conductance distributions are discussed.
Interference and k-point sampling in the supercell approach to phase-coherent transport - art. no. 0333401

We present a systematic study of interference and k-point sampling effects in the supercell approach to phase-coherent electron transport. We use a representative tight-binding model to show that interference between the repeated images is a small effect compared to the error introduced by using only the Gamma-point for a supercell containing (3,3) sites in the transverse plane. An insufficient k-point sampling can introduce strong but unphysical features in the transmission function which can be traced to the presence of van Hove singularities in the lead. We present a first-principles calculation of the transmission through a Pt contact which shows that the k-point sampling is also important for realistic systems.

General information
State: Published
Organisations: Theoretical Atomic-scale Physics, Department of Physics
Contributors: Thygesen, K. S., Jacobsen, K. W.
Molecular transport calculations with Wannier Functions

We present a scheme for calculating coherent electron transport in atomic-scale contacts. The method combines a formally exact Green's function formalism with a mean-field description of the electronic structure based on the Kohn-Sham scheme of density functional theory. We use an accurate plane-wave electronic structure method to calculate the eigenstates which are subsequently transformed into a set of localized Wannier functions (WFs). The WFs provide a highly efficient basis set which at the same time is well suited for analysis due to the chemical information contained in the WFs. The method is applied to a hydrogen molecule in an infinite Pt wire and a benzene-dithiol (BDT) molecule between Au(111) surfaces. We show that the transmission function of BDT in a wide energy window around the Fermi level can be completely accounted for by only two molecular orbitals. (c) 2005 Elsevier B.V. All rights reserved.
Scopus rating (2015): CiteScore 1.73 SJR 0.661 SNIP 0.804
Web of Science (2015): Impact factor 1.758
Web of Science (2015): Indexed yes
BFI (2014): BFI-level 1
Scopus rating (2014): CiteScore 1.59 SJR 0.666 SNIP 0.716
Web of Science (2014): Impact factor 1.652
Web of Science (2014): Indexed yes
BFI (2013): BFI-level 1
Scopus rating (2013): CiteScore 1.75 SJR 0.662 SNIP 0.717
Web of Science (2013): Impact factor 2.028
ISI indexed (2013): ISI indexed yes
BFI (2012): BFI-level 1
Scopus rating (2012): CiteScore 1.81 SJR 0.862 SNIP 0.896
Web of Science (2012): Impact factor 1.957
ISI indexed (2012): ISI indexed yes
Web of Science (2012): Indexed yes
BFI (2011): BFI-level 1
Scopus rating (2011): CiteScore 1.89 SJR 0.889 SNIP 0.867
Web of Science (2011): Impact factor 1.896
ISI indexed (2011): ISI indexed yes
BFI (2010): BFI-level 1
Scopus rating (2010): SJR 0.98 SNIP 0.923
Web of Science (2010): Impact factor 2.017
Web of Science (2010): Indexed yes
BFI (2009): BFI-level 1
Scopus rating (2009): SJR 1.127 SNIP 0.928
Web of Science (2009): Indexed yes
BFI (2008): BFI-level 1
Scopus rating (2008): SJR 1.2 SNIP 0.885
Web of Science (2008): Indexed yes
Scopus rating (2007): SJR 1.092 SNIP 0.853
Scopus rating (2006): SJR 1.096 SNIP 0.973
Web of Science (2006): Indexed yes
Scopus rating (2005): SJR 1.287 SNIP 0.995
Web of Science (2005): Indexed yes
Scopus rating (2004): SJR 1.293 SNIP 0.999
Web of Science (2004): Indexed yes
Scopus rating (2003): SJR 1.458 SNIP 1.027
Web of Science (2003): Indexed yes
Scopus rating (2002): SJR 1.33 SNIP 1.022
Web of Science (2002): Indexed yes
Scopus rating (2001): SJR 1.411 SNIP 0.978
Web of Science (2001): Indexed yes
Scopus rating (2000): SJR 1.071 SNIP 0.953
Web of Science (2000): Indexed yes
Scopus rating (1999): SJR 0.998 SNIP 0.903
Original language: English
DOI: 10.1016/j.chemphys.2005.05.032
Source: orbit
Source-ID: 185470
Research output: Research - peer-review › Journal article – Annual report year: 2005
Partly occupied Wannier functions
We introduce a scheme for constructing partly occupied, maximally localized Wannier functions (WFs) for both molecular and periodic systems. Compared to the traditional occupied WFs the partly occupied WFs possess improved symmetry and localization properties achieved through a bonding-antibonding closing procedure. We demonstrate the equivalence between bonding-antibonding closure and the minimization of the average spread of the WFs in the case of a benzene molecule and a linear chain of Pt atoms. The general applicability of the method is demonstrated through the calculation of WFs for a metallic system with an impurity: a Pt wire with a hydrogen molecular bridge.
Partly occupied Wannier functions: Construction and applications
We have developed a practical scheme to construct partly occupied, maximally localized Wannier functions (WFs) for a wide range of systems. We explain and demonstrate how the inclusion of selected unoccupied states in the definition of the WFs can improve both their localization and symmetry properties. A systematic selection of the relevant unoccupied states is achieved by minimizing the spread of the resulting WFs. The method is applied to a silicon cluster, a copper crystal, and a Cu(100) surface with nitrogen adsorbed. In all cases we demonstrate the existence of a set of WFs with particularly good localization and symmetry properties, and we show that this set of WFs is characterized by a maximal average localization.
Stretching dependence of the vibration modes of a single-molecule Pt-H-2-Pt bridge

A conducting bridge of a single hydrogen molecule between Pt electrodes is formed in a break junction experiment. It has a conductance near the quantum unit, $G(0)=2e^2/h$, carried by a single channel. Using point-contact spectroscopy three vibration modes are observed and their variation upon isotope substitution is obtained. The stretching dependence for each of the modes allows uniquely classifying them as longitudinal or transversal modes. The interpretation of the experiment in terms of a Pt-H-2-Pt bridge is verified by density-functional theory calculations for the stability, vibrational modes, and conductance of the structure.
Structure and transport in nano-scale contacts

General information
State: Published
Organisations: Department of Physics
Contributors: Thygesen, K. S., Jacobsen, K. W.
Publication date: 2005

Publication Information
Original language: English
Source: orbit
Source-ID: 181147
Conductance calculations with a wavelet basis set

We present a method based on density functional theory (DFT) for calculating the conductance of a phase-coherent system. The metallic contacts and the central region where the electron scattering occurs, are treated on the same footing taking their full atomic and electronic structure into account. The linear-response conductance is calculated from the Green’s function which is represented in terms of a system-independent basis set containing wavelets with compact support. This allows us to rigorously separate the central region from the contacts and to test for convergence in a systematic way. The method supports the use of both norm-conserving and ultrasoft pseudopotentials. We use the method to study the effect of adsorbates on the conductance of an infinitely long, atomically thin Al wire, and find that hydrogen and oxygen effectively reduce the conductance of the wire by one and two conductance quanta, respectively.

General information
State: Published
Organisations: Theoretical Atomic-scale Physics, Department of Physics
Contributors: Thygesen, K. S., Bollinger, M., Jacobsen, K. W.
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Ratings:
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Web of Science (2019): Indexed yes
BFI (2018): BFI-level 1
Web of Science (2018): Indexed yes
BFI (2017): BFI-level 2
Scopus rating (2017): CiteScore 3.34 SJR 1.604 SNIP 1.04
Web of Science (2017): Impact factor 3.813
Web of Science (2017): Indexed yes
Scopus rating (2016): CiteScore 3.16 SJR 2.339 SNIP 1.151
Web of Science (2016): Impact factor 3.836
Web of Science (2016): Indexed yes
Scopus rating (2015): CiteScore 2.8 SJR 2.377 SNIP 1.13
Web of Science (2015): Impact factor 3.718
Web of Science (2015): Indexed yes
Scopus rating (2014): CiteScore 3.3 SJR 2.762 SNIP 1.316
Web of Science (2014): Impact factor 3.736
Web of Science (2014): Indexed yes
Scopus rating (2013): CiteScore 3.55 SJR 2.813 SNIP 1.326
Web of Science (2013): Impact factor 3.664
ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
Scopus rating (2012): CiteScore 3.57 SJR 3.173 SNIP 1.378
Web of Science (2012): Impact factor 3.767
ISI indexed (2012): ISI indexed yes
Web of Science (2012): Indexed yes
Scopus rating (2011): CiteScore 3.61 SJR 3.326 SNIP 1.423
Web of Science (2011): Impact factor 3.691
ISI indexed (2011): ISI indexed yes
Web of Science (2011): Indexed yes
Scopus rating (2010): SJR 3.318 SNIP 1.447
Web of Science (2010): Impact factor 3.774
Four-atom period in the conductance of monatomic Al wires

We present first-principles calculations based on density functional theory for the conductance of monatomic Al wires between Al(111) electrodes. In contrast to the even-odd oscillations observed in other metallic wires, the conductance of the Al wires is found to oscillate with a period of four atoms as the length of the wire is varied. Although local charge neutrality can account for the observed period, it leads to an incorrect phase. We explain the conductance behavior using a resonant transport model based on the electronic structure of the infinite wire.

General information
State: Published
Organisations: Theoretical Atomic-scale Physics, Department of Physics
Contributors: Thygesen, K. S., Jacobsen, K. W.
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BFI (2018): BFI-level 2
Web of Science (2018): Indexed yes
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Scopus rating (2017): CiteScore 7.58 SJR 3.622 SNIP 2.464
Web of Science (2017): Impact factor 8.839
Web of Science (2017): Indexed yes
BFI (2016): BFI-level 2
Scopus rating (2016): CiteScore 6.33 SJR 4.196 SNIP 2.61
Web of Science (2016): Impact factor 8.462
Web of Science (2016): Indexed yes
BFI (2015): BFI-level 2
Scopus rating (2015): CiteScore 5.76 SJR 4.656 SNIP 2.538
Web of Science (2015): Impact factor 7.645
Web of Science (2015): Indexed yes
BFI (2014): BFI-level 2
Scopus rating (2014): CiteScore 6.62 SJR 5.232 SNIP 2.71
Web of Science (2014): Impact factor 7.512
Web of Science (2014): Indexed yes
BFI (2013): BFI-level 2
Scopus rating (2013): CiteScore 7.46 SJR 5.675 SNIP 2.781
ISI indexed (2013): ISI indexed yes
Web of Science (2013): Indexed yes
BFI (2012): BFI-level 2
Scopus rating (2012): CiteScore 7.19 SJR 6.292 SNIP 2.867
ISI indexed (2012): ISI indexed yes
Web of Science (2012): Indexed yes
BFI (2011): BFI-level 2
Scopus rating (2011): CiteScore 7.02 SJR 6.314 SNIP 2.905
ISI indexed (2011): ISI indexed yes
Web of Science (2011): Indexed yes
BFI (2010): BFI-level 2
Scopus rating (2010): SJR 6.45 SNIP 2.757
Web of Science (2010): Indexed yes
BFI (2009): BFI-level 2
Scopus rating (2009): SJR 6.325 SNIP 2.947
Web of Science (2009): Indexed yes
BFI (2008): BFI-level 2
Scopus rating (2008): SJR 6.194 SNIP 2.837
Web of Science (2008): Indexed yes
Scopus rating (2007): SJR 5.95 SNIP 2.738
Web of Science (2007): Indexed yes
Scopus rating (2006): SJR 4.781 SNIP 2.443
Web of Science (2006): Indexed yes
Scopus rating (2005): SJR 4.082 SNIP 2.101
Web of Science (2005): Indexed yes
Scopus rating (2004): SJR 3.847 SNIP 2.122
Web of Science (2004): Indexed yes
Scopus rating (2003): SJR 4.661 SNIP 2.651
Web of Science (2003): Indexed yes
Web of Science (2002): Indexed yes
Theoretical studies of materials for water splitting
Garrijo del Río, E., PhD Student, Department of Physics
Jacobsen, K. W., Main Supervisor, Department of Physics
Thygesen, K. S., Supervisor, Department of Physics
Samfinansieret - Andet
01/08/2017 → 31/07/2020
Award relations: Theoretical studies of materials for water splitting
Project: PhD

Computational studies of two-dimension materials and heterosstructures
Riis-Jensen, A. C., PhD Student, Department of Physics
Thygesen, K. S., Main Supervisor, Department of Physics
Jacobsen, K. W., Supervisor, Department of Physics
Institut stipendie (DTU)
15/07/2017 → 14/07/2020
Award relations: Computational studies of two-dimension materials and heterosstructures
Project: PhD

Spin-valley physics and quantum transport in 2D materials
Handberg Juul Martiny, J., PhD Student, Department of Physics
Jauho, A., Main Supervisor, Center for Nanostructured Graphene
Kaasbjerg, K., Supervisor, Department of Physics
Thygesen, K. S., Supervisor, Department of Physics
Samfinansieret - Andet
01/02/2017 → 31/01/2020
Award relations: Spin-valley physics and quantum transport in 2D materials
Project: PhD

Nye funktionelle nanorør
Strange, M., PhD Student, Department of Physics
Jacobsen, K. W., Main Supervisor, Department of Physics
Thygesen, K. S., Supervisor, Department of Physics
Schiøtz, J., Examiner, Department of Physics
Puska, M. J., Examiner
Stokbro, K., Examiner, Department of Micro- and Nanotechnology
Programbevilling
15/06/2005 → 03/09/2008
Award relations: Nye funktionelle nanorør
Project: PhD

Molekylære kontakers struktur, dynamik og transport
Bækgaard, I. S. B., PhD Student, Department of Applied Mathematics and Computer Science
Jacobsen, K. W., Main Supervisor, Department of Physics
Thygesen, K. S., Supervisor, Department of Physics
Schiøtz, J., Examiner, Department of Physics
Todorov, T. N., Examiner
DTU-lønnet stipendie
01/03/2005 → 16/12/2009
Award relations: Molekylære kontakers struktur, dynamik og transport
Project: PhD

Ultrafast electronic and coupled electronic-nuclear dynamics of solvated metal complexes
Zederkof, D. B., PhD Student, Department of Physics
Haldrup, K., Main Supervisor, Department of Physics
Nielsen, M. M., Supervisor, Department of Physics
Thygesen, K. S., Supervisor, Department of Physics
Institut stipendie (DTU)
01/09/2016 → 31/08/2019
Award relations: Ultrafast electronic and coupled electronic-nuclear dynamics of solvated metal complexes
Project: PhD
Atomic-scale modelling of interfaces in electronic devices
Jelver, L., PhD Student, Theoretical Atomic-scale Physics
Jacobsen, K. W., Main Supervisor, Department of Physics
Stokbro, K., Supervisor, Department of Micro- and Nanotechnology
Stradi, D., Supervisor, Department of Micro- and Nanotechnology
Thygesen, K. S., Supervisor, Department of Physics
Industrial PhD
01/06/2016 → 03/02/2021
Award relations: Atomic-scale modelling of interfaces in electronic devices
Project: PhD

Atomic-scale modelling of carrier dynamics in photo-excited semi-conductors
Haastrup, S., PhD Student, Department of Physics
Thygesen, K. S., Main Supervisor, Department of Physics
Jacobsen, K. W., Supervisor, Department of Physics
Schietz, J., Examiner, Department of Physics
Heine, T., Examiner
Hennig, R. G., Examiner
Institut stipendie (DTU)
01/10/2015 → 30/09/2018
Award relations: Atomic-scale modelling of carrier dynamics in photo-excited semi-conductors
Project: PhD

Ballistic Transport in van der Waals Heterostructure Devices
Zultak, J., PhD Student, Department of Micro- and Nanotechnology
Bøggild, P., Main Supervisor, Center for Nanostructured Graphene
Booth, T., Supervisor, Center for Nanostructured Graphene
Thygesen, K. S., Examiner, Department of Physics
Chernikov, A., Examiner
Withers, F., Examiner
Gorbachev, R., Supervisor
Samfinansieret - Andet
15/08/2015 → 30/09/2018
Award relations: Ballistic Transport in van der Waals Heterostructure Devices
Project: PhD

Theory of quantum plasmonics
Wedel, K. O., PhD Student, Department of Photonics Engineering
Wubs, M., Main Supervisor, Department of Photonics Engineering
Mortensen, N. A., Supervisor, Department of Physics
Thygesen, K. S., Supervisor, Department of Physics
Laurynenka, A., Examiner, Department of Photonics Engineering
Kohler, S., Examiner
Pedersen, T. G., Examiner
Samfinansieret - Andet
15/04/2015 → 19/11/2018
Award relations: Theory of quantum plasmonics
Project: PhD

Computational screening of new inorganic materials for high-efficiency solar cells
Kuhar, K., PhD Student, Department of Physics
Jacobsen, K. W., Main Supervisor, Department of Physics
Thygesen, K. S., Supervisor, Department of Physics
Schietz, J., Examiner, Department of Physics
Jonsson, H., Examiner
Hautier, G., Examiner
Institut stipendie (DTU)
01/11/2014 → 17/01/2018
Award relations: Computational screening of new inorganic materials for high-efficiency solar cells
Project: PhD
**Advanced methods for total energy calculations of complex materials**
Schmidt, P. S., PhD Student, Department of Physics
Thygesen, K. S., Main Supervisor, Department of Physics
Schütz, J., Examiner, Department of Physics
Rohlfing, M., Examiner
Grüneis, A., Examiner

Institut stipendie (DTU)
01/09/2014 → 15/11/2017
Award relations: Advanced methods for total energy calculations of complex materials
Project: PhD

**Characterization of Nanomaterials with Experimental Measurements and Atomistic Simulations**
Larsen, P. M., PhD Student, Department of Physics
Schütz, J., Main Supervisor, Department of Physics
Schmidt, S., Supervisor, Department of Physics
Thygesen, K. S., Examiner, Department of Physics
Goedecker, S., Examiner
Ferrando, R., Examiner

Institut stipendie (DTU)
01/09/2014 → 15/11/2017
Award relations: Characterization of Nanomaterials with Experimental Measurements and Atomistic Simulations
Project: PhD

**Theoretical study of Photo-Catalytic oxygen evolution**
Man, I. C., PhD Student, Department of Physics
Rossmeisl, J., Main Supervisor, Department of Physics
Thygesen, K. S., Examiner, Department of Physics
Hellman, A., Examiner
Krtíl, P., Examiner

Marie Curie (EU-stipendium)
01/03/2008 → 01/06/2011
Award relations: Theoretical study of Photo-Catalytic oxygen evolution
Project: PhD

**Electronic Properties of Transition Metal Dichalcogenide Heterostructures**
Latini, S., PhD Student, Department of Physics
Thygesen, K. S., Main Supervisor, Department of Physics
Jacobsen, K. W., Examiner, Department of Physics
Gatti, M., Examiner
Wehling, T. O., Examiner
Gatti, M., Examiner
Wehling, T. O., Examiner
Grundforskningsfonden
15/09/2013 → 12/12/2016
Award relations: Electronic Properties of Transition Metal Dichalcogenide Heterostructures
Project: PhD

**Ab-initio modelling of electronic excitations in nano-structured graphene**
Rasmussen, F. A., PhD Student, Department of Physics
Thygesen, K. S., Main Supervisor, Department of Physics
Brandbyge, M., Examiner, Department of Physics
Draxl, C., Examiner
Hofmann, P., Examiner
Grundforskningsfonden
15/10/2012 → 31/03/2016
Award relations: Ab-initio modelling of electronic excitations in nano-structured graphene
Project: PhD

**DFT-Simulations of low temperature fuel cells catalysis**
Hansen, M. H., PhD Student, Department of Physics
Thygesen, K. S., Main Supervisor, Department of Physics
Rossmeisl, J., Supervisor, Department of Physics
Jacobsen, K. W., Examiner, Department of Physics
Gross, A., Examiner
Koper, M. T. M., Examiner
Groß, A., Examiner
Koper, M. T. M., Examiner
Forskningsrådsfinansiering
15/05/2013 → 25/08/2016
Award relations: DFT-Simulations of low temperature fuel cells catalysis
Project: PhD

Modeling of level alignment and charge separation at donor/acceptor interfaces
Ørnsø, K. B., PhD Student, Department of Physics
Thygesen, K. S., Main Supervisor, Department of Physics
Jacobsen, K. W., Supervisor, Department of Physics
Vegge, T., Examiner, Department of Energy Conversion and Storage
Boschloo, G. K., Examiner
Troisi, A., Examiner
Boschloo, G. K., Examiner
Troisi, A., Examiner
Forskningsrådsfinansiering
01/08/2012 → 23/10/2015
Award relations: Modeling of level alignment and charge separation at donor/acceptor interfaces
Project: PhD

X-ray Tracking of Atomic Motion During Chemical Reactions Using Free Electron Lasers
Biasin, E., PhD Student, Department of Physics
Nielsen, M. M., Main Supervisor, Department of Physics
Christensen, M., Supervisor, Department of Physics
Haldrup, K., Supervisor, Department of Physics
Thygesen, K. S., Examiner, Department of Physics
Milne, C. J., Examiner
Nilsson, A., Examiner
Institut, samfinansiering
01/09/2013 → 12/12/2016
Award relations: X-ray Tracking of Atomic Motion During Chemical Reactions Using Free Electron Lasers
Project: PhD

Theoretical study of ammonia synthesis under ambient conditions
Petzold, V. G., PhD Student, Department of Physics
Jacobsen, K. W., Main Supervisor, Department of Physics
Bligaard, T., Supervisor, Department of Physics
Narskov, J. K., Supervisor, Department of Physics
Thygesen, K. S., Examiner, Department of Physics
Hyldgaard, P., Examiner
Reuter, K., Examiner
DTU-lønnet stipendie
01/04/2007 → 02/03/2011
Award relations: Theoretical study of ammonia synthesis under ambient conditions
Project: PhD

Direct NO decomposition over non-transition metal surfaces
Jiang, T., PhD Student, Department of Physics
Bligaard, T., Main Supervisor, Department of Physics
Thygesen, K. S., Examiner, Department of Physics
Lopez, N., Examiner, Department of Physics
Jonsson, H., Examiner
Jonsson, H., Examiner
Forskningsrådsfinansiering
01/10/2007 → 20/04/2011
Award relations: Direct NO decomposition over non-transition metal surfaces
Project: PhD
**Elektrontransport i Nanostrukturer**

Rostgaard, C., PhD Student, Department of Physics
Jacobson, K. W., Main Supervisor, Department of Physics
Thygesen, K. S., Supervisor, Department of Physics
Bligaard, T., Examiner, Department of Physics
Gao, S., Examiner
van Leeuwen, R., Examiner

DTU-lønnet stipendie
15/08/2006 → 10/02/2010

Award relations: Elektrontransport i Nanostrukturer
Project: PhD

**Nanostrukturers opbygning, dynamik og transportegenskaber**

Thygesen, K. S., PhD Student, Department of Physics
Jacobson, K. W., Main Supervisor, Department of Physics
Nørskov, J. K., Examiner, Department of Physics
Gonze, X., Examiner
Stokbro, K., Examiner, Department of Micro- and Nanotechnology

DTU-lønnet stipendie
01/02/2002 → 06/06/2005

Award relations: Nanostrukturers opbygning, dynamik og transportegenskaber
Project: PhD

**Katalytiske og elektroniske egenskaber af metalnanopartikler**

Larsen, A. H., PhD Student, Department of Physics
Jacobson, K. W., Main Supervisor, Department of Physics
Thygesen, K. S., Supervisor, Department of Physics
Rossmeisl, J., Examiner, Department of Physics
Grönbeck, H., Examiner
Manninen, M. J., Examiner
Institut stipendie (DTU) Samf.
01/09/2008 → 20/01/2012

Award relations: Katalytiske og elektroniske egenskaber af metalnanopartikler
Project: PhD

**Computationel tools and studies of Graphene Nanostructures**

Papior, N. R., PhD Student, Theoretical Nanoelectronics
Brandbyge, M., Main Supervisor, Department of Physics
Thygesen, K. S., Examiner, Department of Physics
Pecchia, A., Examiner
Stokbro, K., Examiner, Department of Micro- and Nanotechnology
Grundforskningsfonden
01/08/2012 → 20/04/2016

Award relations: Computationel tools and studies of Graphene Nanostructures
Project: PhD

**Computational Screening of Energy Materials**

Pandey, M., PhD Student, Department of Physics
Jacobson, K. W., Main Supervisor, Department of Physics
Thygesen, K. S., Supervisor, Department of Physics
Schiotz, J., Examiner, Department of Physics
Jonsson, H., Examiner
Wahnström, G., Examiner
1/3 FUU, 1/3 inst 1/3 Andet
01/08/2012 → 30/09/2015

Award relations: Computational Screening of Energy Materials
Project: PhD

**Computational search for new light harvesting materials for solar-to-fuel energy conversion**

Castelli, I. E., PhD Student, Department of Physics
Jacobsen, K. W., Main Supervisor, Department of Physics
Thygesen, K. S., Supervisor, Department of Physics
Schiøtz, J., Examiner, Department of Physics
Marzari, N., Examiner
Pedersen, T. G., Examiner
Forskningsrådsfinansiering
01/06/2010 → 26/09/2013
Award relations: Computational search for new light harvesting materials for solar-to-fuel energy conversion
Project: PhD

Quantum and field effects of oxide heterostructures
Trier, F., PhD Student, Department of Energy Conversion and Storage
Pryds, N., Main Supervisor, Department of Energy Conversion and Storage
Chen, Y., Supervisor, Department of Energy Conversion and Storage
Jespersen, T. S., Supervisor
Thygesen, K. S., Examiner, Department of Physics
Gabay, M., Examiner
Granazio, F. M., Examiner
Gabay, M., Examiner
Institut/centerfinansieret
01/03/2013 → 30/09/2016
Award relations: Quantum and field effects of oxide heterostructures
Project: PhD

Electronic-structure simulations of photo-absorptin properties in extended systems
Hüser, F. J., PhD Student, Department of Physics
Thygesen, K. S., Main Supervisor, Department of Physics
Schiøtz, J., Supervisor, Department of Physics
Rossmeisl, J., Examiner, Department of Physics
Aryasetiawan, F., Examiner
Rinke, P., Examiner
Rinke, E., Examiner
Forskningsrådsfinansiering
01/11/2010 → 24/01/2014
Award relations: Electronic-structure simulations of photo-absorptin properties in extended systems
Project: PhD

X-ray studies og dynamical effects in liquids and at interfaces
Brandt van Driel, T., PhD Student, Risø National Laboratory for Sustainable Energy
Nielsen, M. M., Main Supervisor, Risø National Laboratory for Sustainable Energy
Thygesen, K. S., Examiner, Department of Physics
Anders, M., Examiner
Collet, E., Examiner
Anders, M., Examiner
Grundforskningsfonden
01/01/2011 → 26/08/2014
Award relations: X-ray studies og dynamical effects in liquids and at interfaces
Project: PhD

Nanomodulated graphene devices
Jessen, B. S., PhD Student, Department of Micro- and Nanotechnology
Bægild, P., Main Supervisor, Department of Micro- and Nanotechnology
Brandbyge, M., Supervisor, Department of Physics
Thygesen, K. S., Examiner, Department of Physics
Eroms, J., Examiner
Gorbachev, R., Examiner
Eroms, J., Examiner
Gorbachev, R., Examiner
Grundforskningsfonden
15/11/2012 → 07/12/2016
Award relations: Nanomodulated graphene devices
Project: PhD
Interfacial Electron Transfer of Large Transition Metal Complexes in Condensed Matter Environments
Jóonsson, E. Ó., PhD Student, Department of Energy Conversion and Storage
Jacobsen, K. W., Main Supervisor, Department of Physics
Thygesen, K. S., Supervisor, Department of Physics
Ulstrup, J., Supervisor, Department of Chemistry
Rossmeisl, J., Examiner, Department of Physics
Jonsson, H., Examiner
Sprik, M., Examiner
Institut stipendie (DTU)
01/10/2010 → 25/04/2014
Award relations: Interfacial Electron Transfer of Large Transition Metal Complexes in Condensed Matter Environments
Project: PhD

In-situ TEM observation of growth and properties of group-IV doped and SiGe heterostructure nanowires
Pennington, R. S., PhD Student, Center for Electron Nanoscopy
Boothroyd, C., Supervisor, Center for Electron Nanoscopy
Dunin-Borkowski, R. E., Supervisor, Center for Electron Nanoscopy
Wagner, J. B., Supervisor, Center for Electron Nanoscopy
Thygesen, K. S., Examiner, Department of Physics
Rosenauer, A., Examiner
Rouviere, J., Examiner
Institut/centerfinansieret
01/01/2009 → 30/04/2012
Award relations: In-situ TEM observation of growth and properties of group-IV doped and SiGe heterostructure nanowires
Project: PhD

Computational modelling of electron transport at metal-organic interfaces
Jin, C., PhD Student, Department of Physics
Thygesen, K. S., Main Supervisor, Department of Physics
Jacobsen, K. W., Examiner, Department of Physics
Pauly, F., Examiner
Ratner, M. A., Examiner
Forskningsrådsfinansiering
15/02/2012 → 21/09/2015
Award relations: Computational modelling of electron transport at metal-organic interfaces
Project: PhD

First-principles theory of Light-matter interaction in Low-dimensional Materials
Gjerding, M. N., PhD Student, Department of Physics
Thygesen, K. S., Main Supervisor, Department of Physics
Andersen, U. L., Examiner, Department of Physics
Peres, N. M. R., Examiner
Wirtz, L., Examiner
Wirtz, L., Examiner
Institut stipendie (DTU)
01/09/2013 → 15/03/2017
Award relations: First-principles theory of Light-matter interaction in Low-dimensional Materials
Project: PhD

Theoretical Investigation of Plasmonic Materials using Electronic Structure Methods
Winther, K. T., PhD Student, Department of Physics
Thygesen, K. S., Main Supervisor, Department of Physics
Jacobsen, K. W., Supervisor, Department of Physics
Schiatz, J., Examiner, Department of Physics
García de Abajo, F. J., Examiner
Puska, M. J., Examiner
Institut stipendie (DTU)
**Development and testing of new exchange-correlation functionals**
Lundgård, K. T., PhD Student, Department of Physics
Jacobsen, K. W., Main Supervisor, Department of Physics
Thygesen, K. S., Supervisor, Department of Physics
Schiatz, J., Examiner, Department of Physics
Moses, P. G., Examiner, Department of Physics
Tkatchenko, A., Examiner
Tkatchenko, A., Examiner
Institut stipendie (DTU)
01/02/2011 → 20/03/2014
Award relations: Development and testing of new exchange-correlation functionals
Project: PhD

**Molecular Dynamics of Nano-Conductors in the Presence of Electronic Current**
Gunst, T., PhD Student, Department of Micro- and Nanotechnology
Brandbyge, M., Main Supervisor, Department of Physics
Thygesen, K. S., Examiner, Department of Physics
Nikolic, B. K., Examiner
Stokbro, K., Examiner, Department of Micro- and Nanotechnology
Institut stipendie (DTU)
01/10/2010 → 11/12/2013
Award relations: Molecular Dynamics of Nano-Conductors in the Presence of Electronic Current
Project: PhD

**Quantum theory of plasmonic excitations in metallic nanostructures**
Toscano, G., PhD Student, Department of Photonics Engineering
Mortensen, N. A., Main Supervisor, Department of Photonics Engineering
Jauho, A., Supervisor, Department of Micro- and Nanotechnology
Wubs, M., Supervisor, Department of Photonics Engineering
Thygesen, K. S., Examiner, Department of Physics
García de Abajo, F. J., Examiner
Nordlander, P. J. A., Examiner
Institut stipendie (DTU)
01/09/2009 → 04/04/2013
Award relations: Quantum theory of plasmonic excitations in metallic nanostructures
Project: PhD

**Electron Transport and Chemistry of Graphene**
Vanin, M., PhD Student, Department of Physics
Thygesen, K. S., Main Supervisor, Department of Physics
Jacobsen, K. W., Supervisor, Department of Physics
Brandbyge, M., Examiner, Department of Physics
Brocks, G. H. L. A., Examiner
Hornekær, L., Examiner
Institut stipendie (DTU)
01/09/2008 → 30/09/2011
Award relations: Electron Transport and Chemistry of Graphene
Project: PhD