Light-matter interaction in low-dimensional materials. A theoretical study - DTU Orbit (10/04/2019)

Light-matter interaction in low-dimensional materials. A theoretical study
In this thesis we have investigated the optical properties of layered and two-dimensional materials for application in the field of plasmonics and metamaterials using Density Functional Theory (DFT). Both of these fields, if successful in their goals, promise new technologies for small scale photonics beyond the diffraction limit. A technological breakthrough of such a caliber would have far reaching consequences such as enabling a practical interface to nano scale integrated electronic circuits or enable the construction of novel devices like a superlens with resolution well beyond diffraction limit. However, the progress of these fields is currently inhibited by large losses that can only be resolved through the discovery of new materials.

Using linear response time-dependent DFT we calculate the optical properties of several experimentally known layered transition metal dichalcogenides (TMDs) with the chemical formula of MX₂ where M is a transition metal and X is a chalcogen atom (S, Se, Te). The TMDs constitute an interesting class of materials due to their diverse range of properties including both metals and semi-conductors. We find that the TMDs with group 5 transition metal atoms in the H monolayer exhibit a special bandstructure in which metallic bands are separated from other valence and conduction bands by finite energy gaps which has the potential to minimize the optical losses by reducing the density of states for scattering. The size of the energy gaps are, however, not sufficiently large to completely eliminate optical losses. We therefore propose a new class of layered materials with the chemical formula 2H-MXY where M is a group 4 transition metal atom, X is a chalcogen atom, and Y is a halogen atom (Cl, Br, I) which increases the size of the energy gaps and significantly reduces optical losses. This entails improved plasmonic normalized propagation lengths and superior lifetimes compared to the best plasmonic material, namely, silver.

We show that all of the TMDs are natural hyperbolic materials, which means that they exhibit a strongly anisotropic dielectric response reflected by a sign-difference in their dielectric tensor resulting in hyperbolic isofrequency contours. Hyperbolic metamaterials obtain their anisotropic response from an artificial sub-wavelength structuring and are limited in their performance by the period of the structuring - the smaller, the better. In contrast, natural hyperbolic materials suffer no such limitation due to their lack of artificial structuring, and indeed, we find a much greater performance for all applications of hyperbolic materials.

The possibility of stacking individual two-dimensional materials into so-called van der Waals heterostructures is perhaps one of the most interesting technological developments in the field of two-dimensional materials. We show that effective medium theory for the dielectric properties of graphene and hexagonal boron nitride heterostructures, which treat the heterostructure as a continuous medium, break down for atomically thin layers due to quantum mechanical effects but also for thick components due to multiple reflection effects. We propose an extended version of effective medium theory to account for the interface layers and show that the effective medium description is improved.

The determination of the quality of materials for application within plasmonics and metamaterials requires an accurate calculation of the optical properties of materials which can be computationally demanding. To reduce the computational costs, the linear tetrahedron method and the employment of symmetries have been implemented which in some cases can reduce the computational costs by a factor of 200.

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