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Electron-phonon interaction and transport properties of metallic bulk and monolayer transition metal dichalcogenide TaS\(_2\)

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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 TaS\(_2\). 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 TaS\(_2\) as promising material candidate for transparent conductors or as atomically thin interconnects. Throughout the paper, the electronic and transport properties of TaS\(_2\) are compared to those of its isoelectronic counterpart TaSe\(_2\) and additional informations to the latter are given. We furthermore comment on the conventional superconductivity in TaS\(_2\), where no phonon-mediated enhancement of \(T_c\) in the monolayer compared to the bulk state was found.

A. Introduction

Succeeding the many advances in fundamental science and applications of graphene, other two-dimensional materials, especially the transition-metal dichalcogenides (TMDs), have attracted remarkable interest for their appealing electrical, optical and mechanical properties. TMDs form layered compounds having a metal layer sandwiched between two chalcogen layers. These monolayers are weakly bound to each other by the dispersive van der Waals interaction, leading to a quasi-two-dimensional behavior in the monolayers\(^1,2\). Among the TMDs, exfoliated semi-conducting materials, e.g. MoS\(_2\) and WS\(_2\), have been extensively in focus for their promising electronic mobilities and thus a high potential in thin-film transistor applications\(^3–5\). Recently metallic TMDs are in focus for their potential as transparent conductive electrodes and gained increasing importance for information-, optoelectronic- and energy-applications\(^6,7\). Crucial requirements for these are high electrical conductivity and high transparency; properties that are often contradicting.

Based on density functional theory and semi-classical Boltzmann transport, we will elaborate and discuss the influence of electron-phonon coupling onto the electronic transport properties and high symmetry point, reaching values of \(\Delta = 2.92\)Å in the unit cell. X-ray diffraction of 2H-TaS\(_2\) in powder form yield similar lattice parameters at \(a_{\text{lat}} = 3.31\)Å and \(c_{\text{lat}} = 12.02\)Å, containing two S-Ta-S monolayers separated by an interlayer distances of \(\Delta = 2.92\)Å in the unit cell. X-ray diffraction of 2H-TaS\(_2\) in powder form yield similar lattice parameters at \(a_{\text{lat}} = 3.32\)Å and \(c_{\text{lat}} = 12.10\)Å\(^2\). For the 1H-monolayer a vacuum of 15Å was taken into account to prevent surface-surface hybridization.

While being moderately large in comparison to conventional spin-orbit driven materials, e.g. Bi\(_2\)Te\(_3\), spin-orbit coupling (SOC) leads often to visible band splittings in two-dimensional TMDs. The H-phase own a lattice without inversion symmetry and thus the spin degeneracy is mostly removed, except for the \(\Gamma\)-point. TaS\(_2\) is metallic due to a half-filled 5\(d_{z^2}\) orbital and these splittings are most pronounced near the Fermi energy in the vicinity of the \(K\) high symmetry point, reaching values of around 350 meV. As shown in Fig. 1 the variation in the electronic band structures of 1H and 2H-TaS\(_2\) are rather small in the vicinity of \(E_F\). Lifted degeneracies in the bulk phase at the \(\Gamma\), \(M\) and \(K\) high symmetry points, as well as a closing of the electronic gap below the Fermi energy in the bulk case are the most distinct effects of...
C. Electron-phonon coupling

The phonon band structures of bulk 2H-TaS$_2$ and its monolayer counterpart are shown in Fig. 2(a). Peculiar for 2D materials are the soft out-of-plane acoustic modes (ZA), or flexural acoustic phonons, presenting a quadratic dispersion at small momenta $q$. In 2H-TaS$_2$, as well as 1H-TaS$_2$, high-frequency optical modes are separated from the low-frequency modes by a gap of around 40 cm$^{-1}$, comparable to other TMDs$^{26}$. As expected, bearing in mind the weak interlayer coupling, the phonon branches are almost doubly degenerate for the 2H polytype at frequencies larger than 50 cm$^{-1}$ and no significant variation between the two material systems could be found. The most noticeable deviation can be found for small frequencies, where the bulk branches split into acoustic branches that approach zero frequency for vanishing $q$ (corresponding to in-phase oscillation of equivalent atoms of adjacent monolayers) and optical modes that approach a finite value (corresponding to a phase-difference of $\pi$ in the oscillation of adjacent monolayers)$^{27}$. The total phonon bandwidth (390 cm$^{-1}$) for TaS$_2$ is noticeably smaller than for other prototype 2D materials, e.g. graphene (1700 cm$^{-1}$) or MoS$_2$ (500 cm$^{-1}$), which in turn could lead to strongly reduced lattice thermal conductivity.

With the knowledge of the electron-phonon matrix elements, the effectiveness of phonons with energy $\hbar \omega$ to scatter electrons can be expressed by means of the, a priori state-dependent, Eliashberg spectral function $\alpha^2 \tilde{\mathcal{F}}(E, \omega)^{18,28}$

$$\alpha^2 \tilde{\mathcal{F}}^{\pm}_{\Psi}(E, \omega) = \frac{1}{N_q} \sum_{q\nu} \delta(\omega - \omega_{q\nu}) |g_{q\nu}^{\Psi q}|^2$$

$$\times \delta \left( E - \epsilon_{q+q}^{0} \pm \hbar \omega_{q\nu} \right) \times \left( 1 - \frac{v_{q+q} \cdot v_{k}}{|v_{q+q}| |v_{k}|} \right)$$

The last term in eq. 2 accounts for back-scattering effects via the change of velocity during the scattering process, which then directly leads to the transport Eliashberg spectral function. In an iterative solution of the full Boltzmann equation, this term would largely correspond to the the scattering-in term. In Fig. 2(d) the difference between $\alpha^2 \mathcal{F}(E, \omega)$ and its transport counterpart are shown. We find both to be functional almost identical, with $\alpha^2 \mathcal{F}(E, \omega)$ being suppressed to around 64% (96%) of the value for $\alpha^2 \mathcal{F}(E, \omega)$ for the acoustic (optical) modes. While similarity of $\alpha^2 \mathcal{F}(E, \omega)$ and $\alpha^2 \mathcal{F}(E, \omega)$ is not a general trend, it has been observed before, experimentally and theoretically, for bulk metals$^{23,29}$. This trend indicates that forward scattering (small angles between $v_{k'}$ and $v_{k}$) is preferred over backward scattering (large angles between $v_{k}$ and $v_{k}$).

Integration of the isotropic (transport) Eliashberg spectral function leads to the unitless (transport) electron-phonon coupling constant $\lambda_{\text{tr}} = \int_{0}^{\infty} d\omega \frac{\alpha^2 \mathcal{F}_{\text{tr}}(\omega)}{\omega}$ which is shown, resolved for

\[\text{FIG. 1. Band structure of bulk 2H-TaS}_2\text{ (black solid lines) and monolayer 1H-TaS}_2\text{ (red dashed lines). The corresponding electrical conductivity anisotropy } \sigma_{zz}/\sigma_{xx}\text{ for bulk 2H-TaS}_2\text{ is shown in the right panel.\]
1. Dynes in the supplemental material.

2. The color code (blue to red; λ = 0 to λ = 1.2) depicts the unitless electron phonon coupling parameter λq. An unconventionally strong coupling of phonons with q ≈ ΓM and ω = 120 cm⁻¹ was found. The phonon density of states F(ω), the isotropic (transport) Eliashberg spectral functions α²F(tr)(ω) and the frequency dependent (transport) coupling strength α²F(tr)(ω), respectively. In (d) the cumulative value for the (transport) electron-phonon coupling constant λ(tr) is additionally shown as red solid (yellow dashed) line.

3. For monolayer 1H-TaSe₂ we calculated values of λ = 0.50 and λ(tr) = 0.46. The enhancement of λ is most prominently related to an enhanced phonon bandwidth and we give further details in the supplemental material. Applying the Allen-Dynes formula to estimate the superconducting transition temperature, we obtain T_C = 0.54 K using an Heine-Morel parameter of μ* = 0.11 representing the repulsive retarded Coulomb potential for bulk 2H-TaS₂ and monolayer 1H-TaS₂, respectively. While our calculated T_C for bulk 2H-TaS₂ is in excellent agreement to recent experimental findings, the superconducting transition temperature of ultra-thin TaS₂ was experimentally found to be larger at T_C = 1.79 K. To obtain this temperature a value for the retarded Coulomb parameter of μ* = 0.057 would have to be assumed. This would be in contrast to the fact that 2D electronic systems are rather poor at screening interactions and the Coulomb interaction between electrons should be much stronger in 2D than in 3D, i.e. generally μ²D > μ³D.

The Eliashberg spectral function α²F(E,ω) can be directly linked to the imaginary part Σ⁺ of the electron self energy induced by interaction with a phonon in state.
spectroscopic electronic linewidth as $\Gamma_{\ell}^\text{ph} = \Sigma_{k}^\prime\prime (E, T)$, i.e. $\Sigma_{k}^\prime\prime = \Sigma_{k}^\prime + \Sigma_{k}^\prime\prime + \Sigma_{k}^\prime\prime\prime$, access to the total complex electron-phonon selfenergy, $\Sigma_{k}$, is obtained by solving a linearized Boltzmann equation in relaxation time approximation (RTA) under the explicit influence of electron-phonon scattering as introduced in Refs. 18, 42–44. The components of the electrical conductivity tensor $\sigma_{ij}$ are given by $\sigma_{ij} = \frac{e^2}{V_{uc} N_k} \sum_{k} \tau_{k} v_{k|i}^0 v_{k|j}^0 \left( -\frac{\partial f_{k}}{\partial \epsilon_{k}} \right)$.

Here, $b$ and $f$ are the equilibrium Bose-Einstein and Fermi-Dirac distribution functions, respectively and introduce the explicit temperature dependence. One gains access to the total complex electron-phonon selfenergy, i.e. $\Sigma_{k}^\prime\prime = \Sigma_{k}^\prime + \Sigma_{k}^\prime\prime + \Sigma_{k}^\prime\prime\prime$, via a Kramers-Kronig relation.

Furthermore, we can introduce the spectroscopic electronic linewidth as $\Gamma_{\ell}^\text{ph} = 2 \Sigma_{k}^\prime\prime$ and the phonon-limited transport lifetime $\tau_{k} = -\frac{\hbar}{2\Sigma_{k}^\prime\prime}$.

The energy-surface-averaged transport lifetime is depicted in Fig. 3 together for bulk 2H and monolayer 1H-TaS$_2$ at three distinct temperatures. Again, due to the similarities in the electronic and vibronic structure, no striking difference between the lifetimes of bulk and monolayer TaS$_2$ can be stated. The pronounced peak of the carrier lifetime around the Fermi energy can be related to a freeze-out of phonons and the narrowing of the Fermi surface resulting in a limited phase space for phonon scattering. Technically, this effect is represented in Eq. 3 by weighting the electron-phonon matrix elements with the Bose-Einstein and Fermi-Dirac distribution functions $f(b, f)$. The peak itself can be decomposed into phonon absorption and emission processes relative to energies below and above the Fermi energy, respectively. The lifetimes related to phonon absorption rapidly increase as the electron energy approaches the Fermi energy from below. Pronounced shoulders in the lifetimes at around 25 meV at very low temperatures of 10 K (c.f. inset Fig. 3) indicate the onset of scattering by optical phonons. Due to the smearing of the Fermi-Dirac distribution and the increasing availability of short-wavelength phonons the lifetime peak steadily decreases at higher temperatures.

D. Transport properties

On the basis of the transport relaxation times, the electronic transport properties are subsequently obtained by solving a linearized Boltzmann equation in relaxation time approximation (RTA) under the explicit influence of electron-phonon scattering as introduced in Refs. 18, 42–44. The components of the electrical conductivity tensor can be stated as

$$\sigma_{ij} = \frac{e^2}{V_{uc} N_k} \sum_{k} \tau_{k} v_{k|i}^0 v_{k|j}^0 \left( -\frac{\partial f_{k}}{\partial \epsilon_{k}} \right)$$

where $V_{uc}$ is the unit cell volume and $N_k$ is the number of initial electronic states in the reciprocal space. One obtains the sheet conductance for a film of thickness $d$ directly as $\sigma_{\square} = \frac{d}{2} (\sigma_{xx} + \sigma_{yy})$ and the unipolar phonon limited electron mobility tensor at a given charge carrier concentration $n$ as $\mu_{ij} = \sigma_{ij}/ne$. The convergence of the sums in Eq. 2 and Eq. 4 is ensured by employing adaptive tetrahedron schemes and using, depending on the topology of the iso-energetic surface, a few $10^4$ ($10^3$) $k$-points and $10^5$ ($10^3$) $q$-points for bulk and monolayer, respectively. In Fig. 4(a) we show the electronic mobilities for bulk and monolayer...
we show more details on the electronic, vibronic and ele-
tron-phonon relaxation times. Second to that is the enhanced electron-phonon coupling and hence re-
nalization times introduced via Eq. 3. The remaining band
structure kernel in Eq. 4 varies only a few percent over
the whole temperature range, mainly linked to a change
of the temperature-dependent chemical potential.

We also highlight the out-of plane mobility for bulk
2H-TaS$_2$, noting $\mu = 0.35$ cm$^2$/Vs at 300 K (c.f. red dotted line in Fig. 4(a)). The reduction stems dominantly
from heavily reduced out-of plane carrier velocities in
the vicinity of the Fermi energy. As shown by the con-
ductivity anisotropy in the right panel of Fig. 1, out-
of-plane mobilities very close to in-plane mobility val-
ues can be achieved under heavy electron doping. This
scenario would assume almost filling up the half-filled
5$d_z$-band, which seems to be unfeasible to achieve by
chemical doping. However an inherent pseudo-doping
of about 0.3 electrons was recently observed for epitaxially
grown monolayer 1H-TaS$_2$.

As expected almost the entire temperature-dependence
of $\mu$ arises from the temperature-dependence of the re-
 laxation times introduced via Eq. 3. The remaining band
structure kernel in Eq. 4 varies only a few percent over
the whole temperature range, mainly linked to a change
of the temperature-dependent chemical potential.

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Accessing high out-of-
plane mobilities via electrical field gating turns out to be
very challenging. Assuming a SrTiO$_3$ substrate of 100 nm
thickness and a film thickness of 10 nm 2H-TaS$_2$ would
still require a gating voltage in the order of 100 V to gain
values close to $\mu \parallel \approx \mu \perp$. We note, that $\mu \perp > \mu \parallel$ can be achieved for $-1.5 \text{eV} < E < -0.3 \text{eV}$, which orig-
inas largely from $p$-orbitals forming highly dispersive
bands with high out-of-plane velocities. As mentioned
before and shown in the supplemental material, applying
GW or hybrid functional calculations, these bands will be
shifted downwards in energy by about 0.6 eV, making
them unlikely to accessed by hole doping.

As noted earlier, with regard to potential application as
transparent conductive electrodes, the electrical sheet
conductance should be the material property in focus
and is shown in Fig. 4(b). TaS$_2$ clearly outperforms
the conventional heavily doped semi-conducting 2D ma-
terials MoS$_2$ and Phosphorene and decreases the gap
to graphene by showing values of $\sigma_\parallel = 2 \text{mS}$ at room
temperature. With this still being an order of magna-
tude worse than the sheet conductances of heavily doped
Graphene or a comparable layer of Cu, the advantages of
thin film TaS$_2$ are noteworthy. On the one hand, in
contrast to Graphene, TaS$_2$ is an inherent metal and thus
does not require to be doped or gated to achieve a feasible
sheet conductances. On the other hand it can easily be
exfoliated due to its weak interlayer interactions and
thinned down to atomic thicknesses, bearing a huge ad-
vantage to traditional metals, e.g. Cu or Pb. These
material properties, together with a high optical trans-
mittance in the visible range, enables H-TaS$_2$ to be a
promising system for application as a transparent con-
ducting oxide.

**E. Phonon induced band renormalization**

Having discussed the single-particle electronic and vi-
bron properties, as well as the phonon limited electronic
transport properties of H-TaS$_2$, we focus in the following
on the renormalization of the electronic structure due to
electron-phonon-interaction. Assuming the photoemis-
sion intensity is proportional to the hole spectral function
of the sample, the electron spectral function

$$\mathcal{A}_k(E) = 1 + \left[ \frac{\Sigma_k''(E)}{E - \epsilon_k' - \Sigma_k'(E)} + \frac{\Sigma_k''(E)}{\Sigma_k'(E)} \right]^2.$$

Can be used to describe the electronic structure of a
solid in the presence of many-body effects. We note,
that within our framework the uncorrected self-energy
corresponds to the Fan contribution, which is a
resonable approach for metallic systems. The quasiparticle
Fermi level fulfills Luttingers theorem of Fermi surface volume
conservation.

Comparing to angle-resolved photoemission (ARPES),
the electron spectral function

$$\mathcal{A}_k(E) = 1 + \left[ \frac{\Sigma_k''(E)}{E - \epsilon_k' - \Sigma_k'(E)} + \frac{\Sigma_k''(E)}{\Sigma_k'(E)} \right]^2.$$
FIG. 5. The electronic spectral function $\omega_k(E)$ with respect to the Fermi energy for monolayer 1H-TaS$_2$, (d)-(f) incorporating electron-phonon interaction as calculated via Eq. 5 at different temperatures (color scale: blue-white-red; low to high intensity. Thin black lines show the maximum highlighted in the panel above). (a)-(c) zoom-in of the corresponding extracted maximum of $\omega_k(E)$ around $E_F$ on the $\Gamma - K$ path (red lines). The black lines indicate the bare band structure without interaction$^a$.

$^a$ Note that the spectral functions have been obtained in a structural phase not possessing a charge-density wave.

weak to modest electron-phonon interaction, recovering the Rayleigh-Schrodinger perturbation theory$^{56}$, the maximum of the electronic spectral function can be estimated to lowest order as $E_k \approx \epsilon^0_k + \Sigma' (\epsilon^0_k, T)$ and is shown in Fig. 5(a)-(c) (red lines) in comparison to the bare non-interacting band structure $\epsilon^0_k$ (black lines)$^{53}$. In the low temperature regime (Fig. 5(a)) clear kinks at energies of about $\pm 45$ meV around $E_F$ can be found in the outlying band, roughly corresponding to maximal available phonon energy. For the inner band the scenario slightly differs. While kinks are visible for energies above $E_F$ no states are available for high frequency phonon absorption below $E_F$. As consequence the local band bottom is energetically lifted and the effective mass increases. At $E_F$ $\Sigma' = 0$ and the volume of the interacting and non-interacting Fermi surface is conserved due to Luttingers theorem$^{54}$. Similar findings have been observed in the f-electron material USb$_2$, whereas renormalization due to electron-electron interaction was assumed there$^{57}$. At elevating temperatures ((Fig. 5(b),(e)) the pronounced features in the real part of $\Sigma$ decrease and the bare band structure is almost recovered at room temperature.

F. Conclusion

On basis of density functional perturbation theory and semi-classical Boltzmann transport, we found promising room temperature mobilities and sheet conductances in metallic bulk 2H-TaS$_2$ and its exfoliated monolayer 1H-TaS$_2$. The sheet conductances can compete with other established 2D materials and theoretically predict TaS$_2$ as a promising material system to be used as a transparent conductor or as atomically thin interconnect. We furthermore found no significant change of the electron-phonon coupling due to interlayer coupling when stacking 1H monolayers to the bulk 2H-TaS$_2$ polytype. Hence no phonon-mediated enhancement of the superconducting transition temperature in the thin film compared to the bulk state could be found. To extent and support our findings on the transition metal dichalcogenide TaS$_2$, we discussed our results at key points to comparable findings on the isoelectronic counterpart TaSe$_2$. To keep the manuscript compact and intelligible, more detailed results on H-TaSe$_2$ can be found in the supplemental material.

Note by the authors: Shortly after acceptance of this work, we became aware of a very recent experimental study by Wijayaratne et al.$^{58}$ on moment-dependent electron-phonon interaction in bulk 2H-TaS$_2$. The observed anisotropy and total values of the coupling parameter $\lambda$, as well as phonon-induced band renormalizations compare very well to our theoretical findings for TaS$_2$. 


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The supplemental material includes details on the superconducting transition temperature $T_c$ in bulk and monolayer TaS$_2$, additional details on the electron phonon coupling in monolayer TaS$_2$ and TaSe$_2$, site- and direction resolved phonon band structures for bulk and monolayer TaS$_2$, k-resolved el-ph relaxation times for the case of monolayer TaS$_2$ including the Fermi surface topology, HSE06 electronic band structures for bulk as well as monolayer TaS$_2$ and TaSe$_2$, informations on the electronic band structure and el-ph relaxation times of bulk TaSe$_2$, and k/q-mesh convergence checks on the electronic mobility of all materials.


The energy-surface-averaged transport lifetime is defined as $\langle \tau_k \rangle = \int \tau_k k b^2(k) \frac{d^3k}{\pi^3}$. For purposes of presentation shown results are afterwards smoothed by a spline function.

We note that the overall energy dependency of $\tau(E)$ in this particular case is functionally best described by the available iso-energy area $\int_{E}^{E+dE} \frac{d^3k}{\pi^3}$. However, a qualitative
argumentation using the electronic density of states still holds.


The reader should keep in mind that 2H-TaS$_2$ undergoes an incommensurate phase transition at $T \approx 75$ K, which is not covered within our methodology.


Technically the doping is achieved, by a rigid shift of the chemical potential into the bands of the material. Experimentally electrical gating would most closely correspond to this situation.


