

Calculations of electron mobility in II-VI semiconductors

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Electron mobility in the conduction band of II-VI semiconductors ZnSe, CdTe, ZnTe, and CdSe was studied. Temperature dependence of mobility was calculated using the methodology based on density functional theory calculations of the electronic states, phonon modes, and electron-phonon coupling constants, along with Fourier-Wannier procedure for interpolation to a dense grid in momentum space. The mobilities obtained from calculations within generalized gradient approximation of density functional theory overestimate the experimental mobility several times. The calculation that used improved electronic band structure and high-frequency dielectric constants obtained using a hybrid functional lead to a very good agreement with experimental mobilities for most of the materials studied. It was also found that the Fröhlich model provides a reasonably good estimate of mobilities around room temperature where longitudinal optical phonons provide the dominant scattering mechanism, as expected for these direct gap materials where all relevant electronic states are in the vicinity of the Γ point. The results indicate as well that the long-ranged part of electron-phonon interaction fully determines the electron mobility in the materials studied. For this reason, the approach where only this part of electron-phonon interaction is calculated using the relevant analytical formulas allows for accurate calculation of mobility without the use of the interpolation procedure for electron-phonon coupling constants.

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I. INTRODUCTION

Mobility of charge carriers (electrons or holes) in a semiconductor material is a physical quantity that strongly affects the performance of electronic and optoelectronic devices based on that material. While it is relatively straightforward to perform the measurement of its value, it is still challenging to theoretically predict it starting from the crystal structure of the material.

The mobility of charge carriers in ultrapure samples is limited by the interaction of charge carriers with phonons. In realistic samples at room temperature it is typically also the carrier-phonon interaction that limits the mobility, whereas at low temperatures and high impurity concentration the scattering of carriers on impurities may play the dominant role. For these reasons, it is necessary to study the electronic structure, the phonon dispersions and the interaction between electrons and phonons to obtain the mobility of charge carriers in the material.

Density functional theory (DFT) [1–3] and density functional perturbation theory (DFPT) [4,5] give a theoretical framework for calculation of electronic states, phonon modes, and electron-phonon coupling constants. However, several challenges had to be overcome to come to a practical scheme that can be used to evaluate phonon-limited electron mobility of a semiconducting material. The electron-phonon coupling constants can vary rapidly as a function of phonon or electron momentum and for this reason a dense momentum grid in the first Brillouin zone is required to accurately represent them

and to reliably calculate the physical quantities that depend on them. On the other hand, reliable electronic structure and phonon dispersions can be obtained from the calculations on a relatively coarse grid (typically on the order of $8 \times 8 \times 8$) of electron and phonon momenta.

The development of the method for interpolation of electron-phonon coupling constants obtained on a coarse grid to a dense grid [6] was a crucial step toward enabling calculations of phonon-limited mobility. The method is based on the transformation between the reciprocal space representation and the local representation. While the relevant quantities in the reciprocal space representation are the coupling parameters between the Bloch electronic states with quasimomentum \mathbf{k} and phonon modes with quasimomentum \mathbf{q} , in the local representation one uses the coupling parameters between localized electronic orbitals (where typically Wannier functions are used as localized orbitals) due to displacements of individual atoms. The electron-phonon coupling constants on a coarse grid in reciprocal space representation are therefore first transformed to a local representation. Using these parameters in the local representation one can then calculate the electron-phonon coupling constants in the reciprocal space representation for arbitrary wave vectors of electrons \mathbf{K} and phonons \mathbf{Q} , not only those from the original coarse grid. First studies of carrier mobility in semiconductors using this approach [7–9] or somewhat different approach [10] were focused on silicon, the most well-known semiconducting material.

The method in this form, however, implicitly contains the assumption that the electron-lattice coupling parameters in the local representation are short ranged, i.e., that the potential change caused by the displacement of an atom in a

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certain direction is short ranged. This is not the case in polar semiconductors where each atom carries an effective charge (so-called Born effective charge), which creates a long-ranged potential when the atom is displaced from its equilibrium position. This issue was overcome using the approach developed in Refs. [11] and [12]. The main idea of the approach is to remove the long-ranged part of electron-lattice coupling parameters caused by the potential of Born effective charges, perform the interpolation starting from short-ranged parameters, and return the long-ranged part at the end. This approach was used in the calculations of mobility for the most representative polar semiconductor GaAs [8,13,14]. Other materials that have been studied afterwards include GaN [15,16], diamond [17], halide perovskites [18], SnSe [19], SiC [20] (for a review of these works see Ref. [21]). There have been also other works where *ab initio* calculations were used to some extent to study the mobility in semiconductors [22–26].

Studies of mobilities in other semiconducting materials are necessary to further verify or improve the whole approach. In this work, we focus on II-VI semiconductors with the formula AB, where A is Zn or Cd, while B is Se or Te. These are well-established semiconductor materials with a wide range of applications including solar cells, x-ray, and γ -ray detectors, infrared optical materials, etc. [27]. We perform calculations of temperature dependence of the mobility of electrons in these materials. We include in the procedure the corrections to known shortcomings of local approximations to density functional theory, such as inaccurate band dispersion and overscreening. It turns out that these corrections are necessary to obtain the mobilities that are in good agreement with experimental results from the literature. Finally, we analyze different contributions to the mobility and identify simpler models that can be used to obtain reasonably accurate values of the mobility at a smaller computational cost.

The paper is organized as follows. In Sec. II we give an overview of the methodology that we used to calculate the electron mobility in the material. The results of our calculations are given in Sec. III with electronic structure presented in Sec. III A and the results for phonons, electron-phonon coupling constants, Born-effective charges, and dielectric constants given in Sec. III B. The comparison of calculated mobilities with experimental results from the literature is performed in Sec. III C and the results for mobility using different models are given in Sec. III D. Final remarks are presented in Sec. IV.

II. METHODOLOGY

The methodology that we used to obtain the electron mobility in the material consists of the calculation of electronic structure and construction of Wannier functions, interpolation of electron-lattice coupling constants from a coarse to a dense momentum grid, calculation of phonons and finally the calculation of electron mobility.

Density functional theory is used to obtain the wave functions of electronic states and corresponding energies. The wave functions obtained are then used to construct the localized Wannier functions [28], i.e., the unitary matrices that describe the transformation from the delocalized Bloch wave functions to localized Wannier functions. With these matrices

at hand one obtains the single-particle Hamiltonian in the Wannier basis, which can then be diagonalized to obtain the wave functions and energies at arbitrary point in the Brillouin zone, not only the ones from the original grid of electronic wave vectors. The steps just described constitute the procedure for Wannier interpolation of electronic band structure. Mathematical details of this procedure are given in Sec. I A of the Supplemental Material [29].

Fourier-Wannier interpolation procedure [6] is used next to perform the interpolation of electron-lattice coupling constants. These constants are obtained first from a density functional perturbation theory calculation on a coarse grid in reciprocal space. The long-ranged part of these constants is then removed following the ideas of Refs. [11] and [12]. In these works, the most important dipole contribution is considered and we note in passing that most recent studies [30,31] have addressed the possibility of including additional quadrupole terms. The transformation of electron-lattice coupling constants to localized Wannier representation is performed next using the same unitary matrices obtained during Wannier interpolation of electronic band structure. In the following step, these constants are transformed back to reciprocal space representation, where these can be obtained now for arbitrary wave vectors, not only for those from original coarse grid. In the final step, long-ranged part of electron-lattice coupling constants is put back. In this step, we allow also for the possibility to correct the long-ranged part of these constants by using the corrected values of high-frequency dielectric constants or Born effective charges. Mathematical details of this procedure that include also comments on some differences of our implementation in comparison to previous works are given in Sec. I B of Supplemental Material [29].

The dynamical matrices, the phonon energies and the displacements of phonon modes on the coarse phonon wave vector grid are obtained from density functional perturbation theory. Interpolation of dynamical matrices to arbitrary phonon wave vectors is performed in a standard manner (see, for example, Refs. [4,5]) by (i) removing the long-ranged part of dynamical matrices; (ii) transforming short-ranged part of dynamical matrices to force constant representation; (iii) evaluating the short-ranged part of dynamical matrices on the dense grid; (iv) putting back the long-ranged part of dynamical matrices; (v) obtaining the phonon energies and displacements from the eigenvalue problem of dynamical matrices. As in the case of electron-phonon coupling constants, we allow here for the possibility to introduce the corrected dielectric constants and Born effective charges in step (iv) of the procedure.

In this work we consider the electrons in the conduction band and assume that the concentration of electrons is relatively low. The mobility obtained this way can be considered as representative mobility of electrons in the conduction band of the material. Within the momentum relaxation time approximation, the components of the charge carrier mobility tensor are given by the expression [32]

$$\mu_{ij} = -e_0 \frac{\sum_{\mathbf{k}} v_{\mathbf{k}}^{(i)} v_{\mathbf{k}}^{(j)} \tau_{\mathbf{k}} \frac{\partial f_{\mathbf{k}}}{\partial \epsilon_{\mathbf{k}}}}{\sum_{\mathbf{k}} f_{\mathbf{k}}}. \quad (1)$$

In this equation $f_{\mathbf{K}}$ is the population of the state at wave vector \mathbf{K} given by the Maxwell-Boltzmann distribution $f_{\mathbf{K}} \propto e^{-\frac{1}{k_B T} \tilde{\epsilon}_{\mathbf{K}}}$, $\tilde{\epsilon}_{\mathbf{K}}$ is its energy, $v_{\mathbf{K}}^{(i)}$ are the components of group velocity $v_{\mathbf{K}}^i = \frac{1}{\hbar} \frac{\partial \tilde{\epsilon}_{\mathbf{K}}}{\partial K_i}$, e_0 is the elementary charge, T is the temperature, while $\tau_{\mathbf{K}}$ is the momentum relaxation time. In the limit of low carrier concentration considered here, this time is given as

$$\frac{1}{\tau_{\mathbf{K}}} = \frac{2\pi}{\hbar N_{\mathbf{Q}}} \sum_{\mathbf{Q}} \sum_{\lambda, \pm} |\gamma^{\lambda}(\mathbf{K}, \mathbf{Q})|^2 \left(n_{\lambda \mathbf{Q}} + \frac{1}{2} \mp \frac{1}{2} \right) \times \delta(\tilde{\epsilon}_{\mathbf{K}} \pm \hbar \omega_{\lambda \mathbf{Q}} - \tilde{\epsilon}_{\mathbf{K} \pm \mathbf{Q}}) (1 - \cos \theta_{\mathbf{K}, \mathbf{K} \pm \mathbf{Q}}). \quad (2)$$

In the previous equation $\gamma^{\lambda}(\mathbf{K}, \mathbf{Q})$ is the electron-phonon coupling element for scattering of an electron in state \mathbf{K} to the state $\mathbf{K} \pm \mathbf{Q}$ due to interaction with phonon in mode λ with wave vector \mathbf{Q} , $\hbar \omega_{\lambda \mathbf{Q}}$ is the energy of that phonon, $n_{\lambda \mathbf{Q}}$ is the occupation of that phonon state given by the Bose-Einstein distribution, while $N_{\mathbf{Q}}$ is the number of points on the dense reciprocal space grid. The term $\cos \theta_{\mathbf{K}, \mathbf{K} \pm \mathbf{Q}}$ is given as

$$\cos \theta_{\mathbf{K}, \mathbf{K} \pm \mathbf{Q}} = \frac{\mathbf{v}_{\mathbf{K}} \cdot \mathbf{v}_{\mathbf{K} \pm \mathbf{Q}}}{|\mathbf{v}_{\mathbf{K}}| \cdot |\mathbf{v}_{\mathbf{K} \pm \mathbf{Q}}|}. \quad (3)$$

Band indices were omitted in previous equation because only the states at the bottom of the conduction band are relevant in this work.

III. RESULTS

A. Electronic structure

Density functional theory calculations of the electronic structure were performed either by approximating the exchange-correlation functional using the Perdew-Burke-Ernzerhof (PBE) generalized gradient approximation (GGA) [33] or by using the hybrid Heyd-Scuseria-Ernzerhof (HSE06) functional [34,35]. The effect of spin-orbit interaction was not included since we focus on the electronic states in the conduction band that mainly originate from s atomic orbitals. Calculations were performed using the ABINIT code [36,37]. The effect of core electrons was modeled using optimized norm-conserving scalar relativistic Vanderbilt pseudopotentials [38,39]. Kinetic energy cutoff for plane-wave representation of the wave functions was 35 Ha for ZnSe and ZnTe, and 40 Ha for CdSe and CdTe, while the $8 \times 8 \times 8$ Monkhorst-Pack grid of points in reciprocal space was used. Experimental lattice constants at room temperature taken from Ref. [40] were used in all calculations. Their values are summarized in Table I.

Wannier functions were constructed from 18 Bloch bands. The top eight valence bands and the lowest ten conduction bands were used to construct ten localized Wannier functions. We have used the procedure for construction of localized Wannier functions for entangled energy bands [41] whereas the Bloch states that lie within the energy window containing the top three valence bands and the bottom conduction band were left unchanged in the band disentanglement procedure (this energy window is called the frozen energy window in terminology of Ref. [42]). Calculations of the unitary matrices that connect the Bloch functions and the Wannier functions (Eq. (2) in Supplemental Material [29]) were performed using

TABLE I. Lattice constants used in the calculation and effective masses and band gaps obtained from electronic structure calculations. a_0 (expt) denotes the experimental value of the lattice constant taken from Ref. [40]. $m_{\text{eff}}(m_0)$ denotes the conduction band effective mass in units of free electron mass m_0 , while E_g denotes the band gap. The label (expt) denotes experimental values of effective masses and gaps taken from the compilation in Ref. [27], while the labels (PBE) and (HSE06) denote our calculations using the PBE and HSE06 functional.

	ZnSe	ZnTe	CdSe	CdTe
a_0 (Å) (expt)	5.667	6.0882	6.078	6.482
$m_{\text{eff}}(m_0)$ (expt)	0.137	0.117	0.119	0.090
$m_{\text{eff}}(m_0)$ (PBE)	0.096	0.087	0.061	0.064
$m_{\text{eff}}(m_0)$ (HSE06)	0.14	0.13	0.13	0.13
E_g (eV) (expt)	2.72	2.27	1.68	1.51
E_g (eV) (PBE)	1.29	1.28	0.61	0.77
E_g (eV) (HSE06)	2.32	2.19	1.53	1.61

the WANNIER 90 code [42]. In Sec. II of Supplemental Material [29] we compare the band structure obtained using direct DFT calculation with the band structure obtained by Wannier interpolation. The results suggest that the dispersion of the first conduction band, which is the relevant band in this study is excellently reproduced by Wannier interpolation.

Calculated band structure of ZnSe along several high-symmetry directions is presented in Fig. 1, while the plot for three other materials that we considered is given in Supplemental material [29] (Sec. III therein). Since we focus on electronic transport in the conduction band, which is strongly influenced by the conduction band effective mass, it is instructive to assess the accuracy of effective masses obtained. For this reason, we compare in Table I the conduction band effective mass obtained from our calculations to the experimental effective mass. The results indicate that the PBE functional significantly underestimates the effective masses. On the other hand, the HSE06 functional gives rather accurate effective masses for ZnSe, ZnTe, and CdSe (interestingly the effective

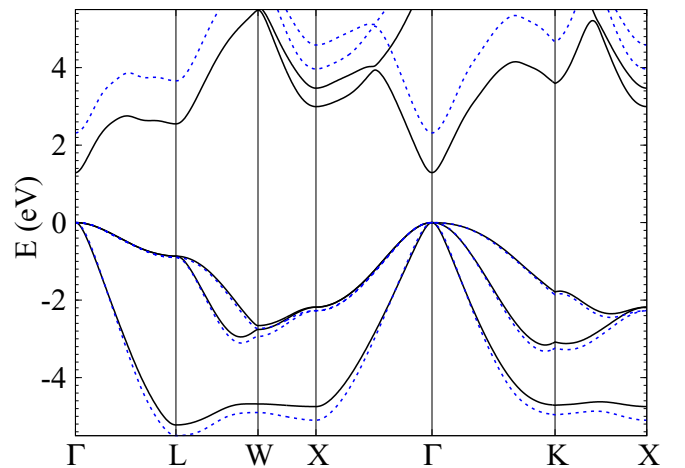


FIG. 1. Electronic band structure of ZnSe calculated using the PBE functional (full line) and the hybrid HSE06 functional (dashed line).

masses of the three materials are rather similar; experimental ones differ by no more than $0.02 m_0$, while the HSE06 values differ by no more than $0.01 m_0$), while it overestimates the effective mass of CdTe. These facts should be born in mind in the analysis of electron mobility results, which is the main focus of this work.

Next, we discuss the need to use the density functional, which yields reasonably accurate values of the band gap. At first thought, this does not seem to be important because the relevant states for electron mobility are the states at the bottom of the conduction band and the value of the material band gap is not relevant for transport properties in the conduction band. The calculated values of the band gap are nevertheless presented in Table I. We see that the HSE06 gaps agree much better with experiment than the PBE gaps, which are too small due to the well-known band gap problem [43] of local and semilocal approximations for exchange-correlation functional in DFT. On the other hand, it has been previously established that HSE06 functional yields quite accurate values of the band gap [44]. We therefore see that both effective masses and band gaps are underestimated in PBE calculations. Such a result could have been expected based on an approximate perturbative formula that connects the conduction band effective masses and the band gap as (see, for example, Ref. [45], p. 12)

$$\frac{1}{m_{\text{eff}}} = \frac{1}{m_0} + \frac{2P^2}{\hbar^2 E_g}, \quad (4)$$

with the matrix element P given as $P = \frac{\hbar}{m_0} \langle S | p_x | X_v \rangle$, where $|S\rangle$ is the conduction band Bloch state at Γ , p_x is the operator of the x component of the momentum, while $|X_v\rangle$ is the top valence band state at Γ that transforms as x under the operation of the symmetry group of the crystal. We see from Eq. (4) that the underestimation of the gap is likely to lead to the underestimation of the conduction band effective mass. As a consequence, to obtain reasonably good conduction band effective masses, one needs to obtain reasonably good band gaps as well.

B. Phonons, electron-phonon coupling constants, Born effective charges, and high-frequency dielectric constants

The calculation of phonons and electron-lattice coupling constants on the coarse $8 \times 8 \times 8$ Monkhorst-Pack grid was also performed using the ABINIT code [36,37]. The PBE exchange-correlation functional was used in the calculation. The phonon band structure of ZnSe obtained from the calculation and subsequent interpolation is presented in Fig. 2, while the phonon band structure of the other materials considered is given in Supplemental Material [29] (Sec. IV therein). We note that HSE06 dielectric constants were used to correct the long-ranged part of the dynamical matrix, as mentioned in Sec. II. The comparison of phonon energies at characteristic points in the Brillouin zone in the cases when this correction was included and excluded with experimental results is given in Tables I–IV in Sec. IV of Supplemental Material [29]. These results suggest that differences between the results obtained in the two cases are rather small and that the results of the calculation agree well with experiment. Namely, the largest difference between the calculation with correction and

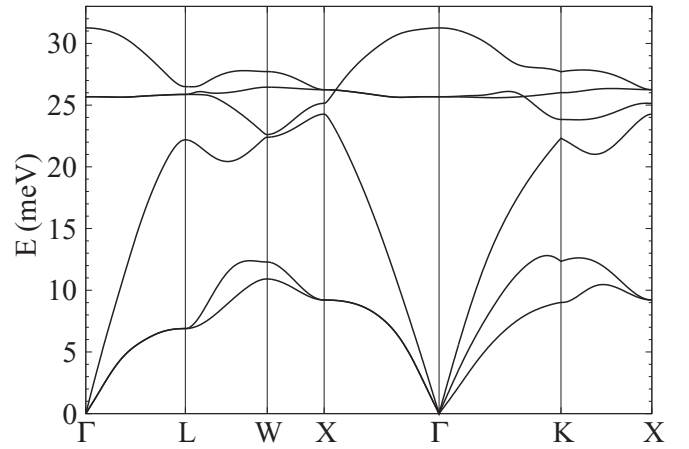


FIG. 2. Phonon band structure of ZnSe.

experiment is 1.3 meV, the largest difference between the calculation without correction and experiment is 1.7 meV, while the largest difference between calculations with and without correction is also 1.3 meV.

The electron-phonon coupling constants [defined after Eq. (2)] obtained from the interpolation procedure are presented in Fig. 3 in the case of ZnSe, and in Supplemental Material [29] (Sec. V therein) for other materials. The interpolation procedure was performed using our in-house code that uses as input the output of DFT and DFPT calculations performed using ABINIT. As a check of accuracy of our implementation of interpolation procedure, we show in Sec. VI of Supplemental Material [29] the comparison of electron-lattice coupling elements obtained directly from DFPT (circles in these figures) and those obtained after interpolation (full line in these figures).

The most relevant electronic states for electronic transport are those in the vicinity of the Γ point. For this reason we present the electron-phonon coupling constants for scattering

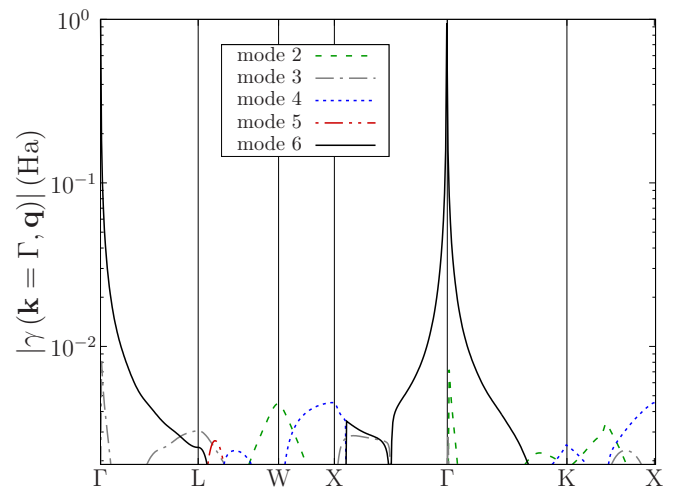


FIG. 3. Electron-phonon coupling constants (obtained from the interpolation procedure) for scattering of an electron at wave vector $\mathbf{k} = \Gamma$ with a phonon of a given mode at wave vector \mathbf{q} in ZnSe. The modes are labeled in ascending order of their energies at \mathbf{q} .

TABLE II. High-frequency dielectric constants and Born effective charges obtained from the calculations using the PBE and HSE06 functional and experimental data for dielectric constants. Experimental data were taken from Ref. [27] where an average is taken from the values obtain from different sources.

	ZnSe	ZnTe	CdSe	CdTe
ϵ_r^∞ (expt)	5.9	6.9	6.2	7.1
ϵ_r^∞ (PBE)	7.26	8.86	7.72	8.62
ϵ_r^∞ (HSE06)	6.15	7.45	6.13	7.01
Z (PBE)	2.05	2.00	2.23	2.18
Z (HSE06)	2.09	2.05	2.24	2.18

of an electron at Γ with phonon of different wave vectors \mathbf{q} . The most relevant among these are the coupling constants at low $|\mathbf{q}|$ because only these phonons can satisfy the energy conservation law for scattering between two electronic states in the vicinity of Γ . As can be seen from Fig. 3, in this region of electron and phonon wave vectors, by far the largest electron-phonon coupling constants, are those with highest energy phonon mode, which is the longitudinal optical (LO) phonon mode. One might even be tempted to conclude from Fig. 3 that the interaction of an electron with phonons in other modes could be completely neglected. However, one should bear in mind that at low temperatures, the number of phonons in high-energy modes decreases exponentially, which leads to a strong reduction of electron-phonon scattering rate. We note that our work does not consider the effect of screening of electron-phonon interaction that becomes important for higher concentration of carriers [46,47].

We next discuss the values of Born effective charges and high-frequency dielectric constants. Since these appear directly in the expression for long-ranged part of the electron-phonon interaction, one should pay particular attention to accuracy of their values. Within PBE approximation to DFT we have calculated the Born effective charges and high-frequency dielectric constants using two different methods: (i) using density functional perturbation theory [5]; (ii) by performing the calculation of polarization and atomic forces in finite electric field [48,49] and extracting the slope of the dependence of these quantities on field at low fields. In calculations that used the HSE06 functional, we have performed only the calculation using the method (ii) due to higher computational cost of hybrid functional calculations and a lack of code that implements DFPT calculation with hybrid functionals. All calculations were performed using the ABINIT code, while additional technical details of the calculations are given in Supplemental Material [29] (Sec. VII therein). We emphasize that larger grids in reciprocal space than those used to obtain the band structure are needed to obtain converged values of dielectric constants. The results of the calculations are summarized in Table II. The results indicate significant differences between dielectric constants obtained using PBE and HSE06 and show that HSE06 results are in much closer agreement with experiment. On the other hand, Born effective charges calculated using the two functionals are very similar. Consequently, we set ϵ_{r1}^∞ in Eq. (9) in Supplemental Material [29] to the value obtained using PBE and we set ϵ_{r2}^∞ in Eq.

(17) in Supplemental Material [29] to the value obtained using HSE06, while we set both Z_1 and Z_2 in these equations to the value obtained using PBE. The difference between dielectric constants obtained using the two functionals originates from the fact that dielectric constants describe the screening of the electric field, which is sensitive to the functional used. On the other hand, the Born effective charges defined as the restoring force on an atom in a unit electric field are largely a local property and appear to be rather insensitive to the functional used.

C. Mobility: Comparison with experiment

Next, we present the calculated temperature dependence of the mobility for the materials under consideration. The mobility was calculated using Eq. (1). The summation was performed over \mathbf{K} points from a $120 \times 120 \times 120$ Monkhorst-Pack grid, where only the points that satisfy the condition $\tilde{\epsilon}_{\mathbf{K}} - \epsilon_{\min} < \epsilon_c$ were taken, where ϵ_{\min} denotes the energy of the conduction band minimum. The value of $\epsilon_c = 250$ meV is sufficient to get converged results at all temperatures considered. Detailed convergence checks with respect to the size of the \mathbf{K} points grid and with respect to the value of ϵ_c are presented in Supplemental Material [29] (Secs. VIII and IX therein). The sums in Eq. (2) containing the δ function were transformed to integrals that were calculated using the tetrahedron method [50,51]. All calculations were performed using our in-house code. Our calculations are based on the momentum relaxation time approximation [Eqs. (2) and (1)]. For this reason, we have estimated the accuracy of this approximation by comparing the results with the full solution of Boltzmann equations in the case of the model with parabolic bands and Fröhlich electron-phonon coupling with a single dispersionless phonon mode. The results of the comparison are presented in Sec. X in Supplemental Material [29]. The results suggest that the error introduced due to momentum relaxation time approximation is smaller than 5% at room temperature and smaller than 30% at lowest temperatures considered. The conclusion in this regard is similar as the conclusion for GaAs in Ref. [8] where the error of momentum relaxation time approximation was also small at higher temperatures and increased as the temperature decreased.

Calculated temperature dependence of the mobility in ZnSe is presented in Fig. 4 along with experimental data from Refs. [52–58]. It can be seen from Fig. 4 that the results obtained when PBE functional is used throughout the calculation significantly overestimate the experimental results. Two main reasons of this overestimation can be identified. We have shown in Sec. III A that the PBE functional calculations underestimate the effective masses. This leads to overestimate of the mobility since lower effective masses are beneficial for efficient charge transport and therefore lead to higher mobilities. In Sec. III B, we have demonstrated that ϵ_r^∞ obtained from PBE is larger than the experimental one. Since the long-ranged part of electron-phonon interaction is inversely proportional (see Eq. (17) in Supplemental Material [29]) to ϵ_r^∞ , it follows that overestimate of ϵ_r^∞ leads to underestimate of electron-phonon scattering matrix elements. Smaller scattering is also beneficial for charge transport and therefore larger mobility is obtained in the calculation.

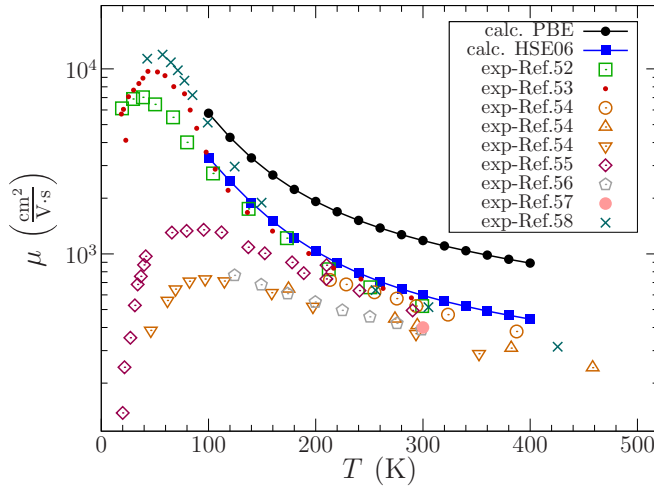


FIG. 4. Temperature dependence of electron mobility in ZnSe. The label “calc. PBE” denotes the calculation where PBE exchange correlation functional is used throughout the calculation, while “calc. HSE06” denotes the calculation where energies and dielectric constants obtained using PBE are replaced with those obtained using HSE06. Experimental data from Refs. [52–58] are given for comparison.

To overcome the limitations of the PBE functional, we performed the calculations where more accurate conduction band structure and ϵ_r^∞ are used in the calculation. More precisely, to improve the conduction band structure, we change in the diagonal matrix $h_d(\mathbf{k})$ in Eq. (3) in Supplemental Material [29] the eigenenergies obtained by PBE with those obtained in HSE06 calculation. To obtain a better description of the screening of the long-ranged part of the electron-phonon interaction, we set ϵ_r^∞ in Eq. (17) in Supplemental Material [29] to the value obtained using HSE06 instead of the value ϵ_r^∞ obtained using PBE that is used in Eq. (9) in Supplemental Material [29]. For brevity, we refer to these calculations as HSE06 calculations, although we note that this does not mean that full DFPT calculation with HSE06 functional was performed. The results obtained in such a calculation are also shown in Fig. 4. It can be seen from the figure that the level of agreement of these results with experimental data from the literature becomes much better, as will be discussed in what follows.

The agreement between HSE06 calculation and experiment is rather good for experimental results presented in Ref. [52] (the high mobility sample in Fig. 4 therein) that concern high-quality samples of ZnSe grown by molecular beam epitaxy. The same is the case regarding the results of Ref. [53] (Fig. 3 therein). The calculation results are also only slightly larger than the measurement results on the highest mobility sample in Ref. [54] (Fig. 5 therein), while two other samples in Ref. [54] have lower mobilities (these are also presented in our Fig. 4) suggesting that nonintrinsic scattering mechanisms could be present. Our calculation results are somewhat larger than the experimental data in Ref. [55] [Fig. 2(b) therein] in the temperature range between 200 K and 300 K. At lower temperatures experimental data of Ref. [55] show a decrease of mobility with decreasing temperature, which is a signature of the effects of impurity scattering and for this reason comparison of our calculation with experiment would not

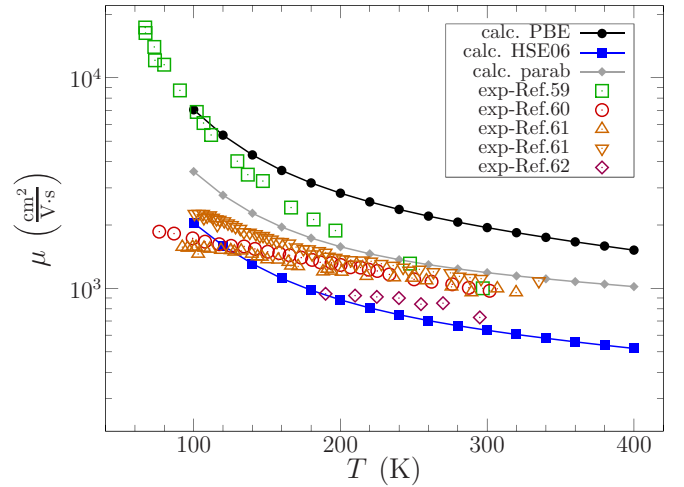


FIG. 5. Temperature dependence of electron mobility in CdTe. The label “calc. PBE” denotes the calculation where PBE exchange correlation functional is used throughout the calculation, “calc. HSE06” denotes the calculation where energies and dielectric constants obtained using PBE are replaced with those obtained using HSE06, while “calc. parab.” denotes the calculation where parabolic band structure with experimental effective mass is used along the HSE06 dielectric constants. Experimental data from Refs. [59–62] are given for comparison.

be meaningful in this temperature range. The experimental results of Refs. [56] (Fig. 1 therein) and [57] (Table I therein) are noticeably smaller than the results of our calculation, even around room temperature, suggesting that these samples could be of somewhat lower quality than the samples previously considered. Our calculation results are also in reasonably good agreement with the results of Ref. [58] except at the lowest temperatures (around 100 K) where, surprisingly, larger values of mobility are obtained in experiment. We note that this is the region where the concentration of carriers (and therefore the current in the experiment) is rather small and that the origin of the difference could potentially be the issues with accurate measurements of small currents. Overall, Fig. 4 suggests that HSE06 calculation results are at the upper limit of the results obtained in experiments and are in agreement with the experimental results obtained on highest purity and highest mobility samples.

Next, we turn our attention to the CdTe material and compare in Fig. 5 the results of our calculations with experimental data from Refs. [59] (highest mobility sample in Fig. 1 therein), [60] (highest mobility sample in Fig. 3 therein), [61] (Figs. 2 and 6), [62] (Fig. 3). As in the case of ZnSe, PBE calculation gives higher mobility than the HSE06 calculation due to differences in effective masses and dielectric constants predicted by two different functionals. The HSE06 results in the region of temperatures near 300 K are lower than experimental results. To understand why this is the case, we recall from Table I that HSE06 effective mass of CdTe is larger than experimental effective mass. For this reason, the calculation that uses the HSE06 band structure underestimates the material mobility. On the other hand, the PBE calculation where effective mass is underestimated overestimates the mobility. Such results point to the necessity of accurately describing

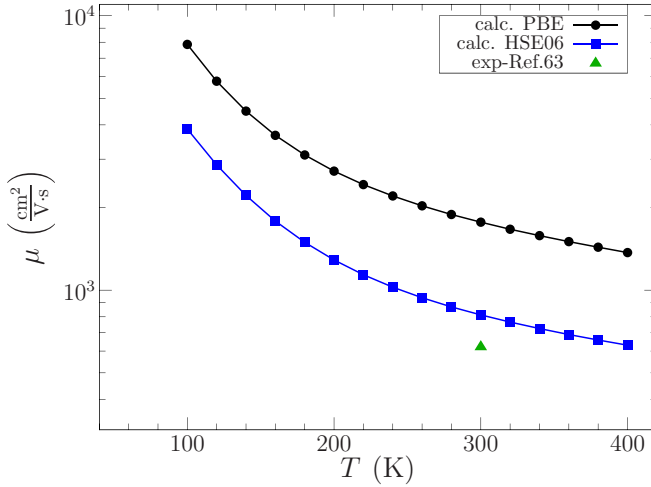


FIG. 6. Temperature dependence of electron mobility in ZnTe. The label “calc. PBE” denotes the calculation where PBE exchange correlation functional is used throughout the calculation, while “calc. HSE06” denotes the calculation where energies and dielectric constants obtained using PBE are replaced with those obtained using HSE06. Experimental data from Ref. [63] are given for comparison.

the band curvature near the bottom of conduction band (i.e., the conduction band effective mass) for accurate prediction of electron mobility. To confirm that this is the case, we performed an additional calculation where a parabolic band structure with experimental effective mass was assumed. As expected, this calculation gives a rather good agreement with experiment at temperatures around the room temperature, see Fig. 5. Nevertheless, one should note that the calculation can no longer be considered as an *ab initio* calculation if the band structure obtained from experimental effective mass is used in the calculation.

We consider next the ZnTe material. Experimental data for mobility in *n*-type ZnTe are rather scarce probably because *n*-type doping of ZnTe is rather difficult [40]. We compare in Fig. 6 our HSE06 result with the results of Ref. [63] (the point at lowest concentration in Fig. 2 therein) and obtain reasonably good agreement. As in the case of other materials, our PBE results are significantly larger than HSE06 results.

Finally, we discuss the results obtained for CdSe. The comparison of our calculation results with experimental data from Refs. [64] (Fig. 3) and [65] (Fig. 2) is presented in Fig. 7. Our HSE06 calculation results closely follow the results of Ref. [64] and are somewhat smaller than the results of Ref. [65]. It should be noted that our calculations concern the zincblende form of CdSe, while it has been reported that experimental data correspond to wurtzite form of CdSe. However, the arrangement of atoms and the distances between them in two forms of CdSe are rather similar and the values of most relevant material parameters (such as the conduction band effective mass, the high-frequency dielectric constant, the band gap, the optical phonon energies, and the material density) are also rather similar (see, for example, Ref. [27] for the values of these parameters in wurtzite and zincblende CdSe). For this reason, the comparison of our results with experimental data could be meaningful.

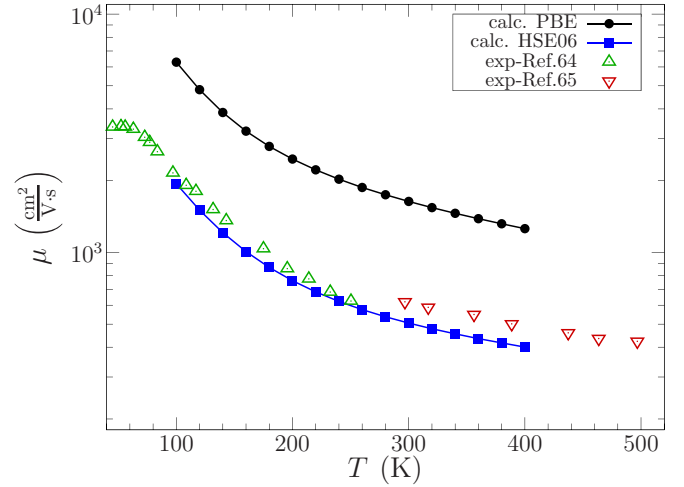


FIG. 7. Temperature dependence of electron mobility in CdSe. The label “calc. PBE” denotes the calculation where PBE exchange correlation functional is used throughout the calculation, while “calc. HSE06” denotes the calculation where energies and dielectric constants obtained using PBE are replaced with those obtained using HSE06. Experimental data from Refs. [64,65] are given for comparison.

It is evident from previous results that better results for mobility were obtained by introducing two corrections: the correction to the dielectric constant and the correction to the band structure. It is instructive to assess the importance of each of the two corrections. For this reason, we have also performed the calculations where only one of these corrections was included. The results presented in Sec. XI of Supplemental Material [29] indicate that correction to the band structure is larger and that both contributions are significant. For example, in the case of ZnSe, the correction to the dielectric constant reduces the room-temperature mobility by a factor of approximately 1.25, while the correction to the band structure reduces it by a factor of 1.6, yielding a total reduction by approximately a factor of 2.

We note that all our calculations concern the drift mobility μ_d of electrons in the material, while most of the experiments report the Hall mobility μ_H . The two are related as $\mu_H = r_H \mu_d$, where r_H is the so-called Hall factor. A good estimate of the Hall factor can be obtained from a result derived for a parabolic band that reads (see, for example, Ref. [32], p. 196)

$$r_H = \frac{\langle \varepsilon \rangle \cdot \langle \varepsilon \cdot \tau^2 \rangle}{\langle \varepsilon \cdot \tau \rangle^2}, \quad (5)$$

where ε is the energy of the carrier with respect to the bottom of the conduction band, τ is the momentum relaxation time, and the averages are defined as $\langle x \rangle \equiv \frac{\sum_{\mathbf{k}} f_{\mathbf{k}} x}{\sum_{\mathbf{k}} f_{\mathbf{k}}}$. We expect that this is a good estimate for the materials at hand because the bottom of the conduction band can be well approximated to be parabolic. We have calculated r_H using Eq. (5) for all materials that we considered throughout the whole range of temperatures and we have found that r_H is within the range (1.03, 1.17), while corresponding values at four different temperatures for all material considered are given in Sec. XII of Supplemental Material [29]. Since it only slightly deviates

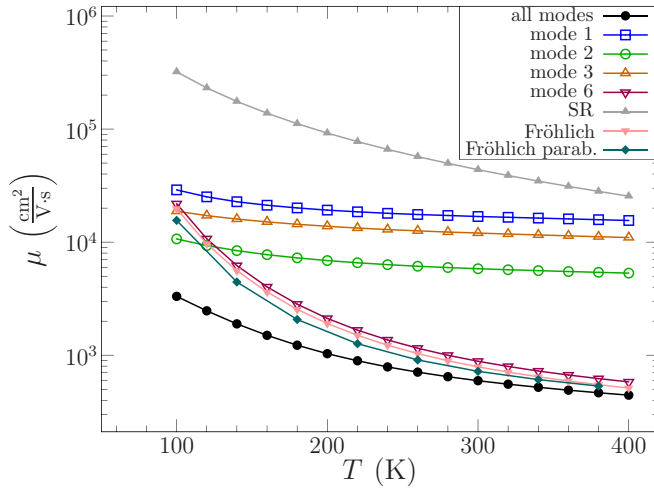


FIG. 8. Temperature dependence of mobility in ZnSe: different contributions and the results from different models. The label “all modes” denotes the results when all phonon modes are included, while the labels “modes 1–6” denote the results when only a particular phonon mode is included. The phonon modes are numbered in ascending order of their energies. The label “SR” concerns the results where only short-ranged part of electron-phonon coupling is included. The label “Fröhlich” denotes the results where only mode 6 is included and where Fröhlich model for electron-phonon coupling is used. The label “Fröhlich parab.” refers to the previous case where the band energies are replaced with those obtained from a parabolic band dispersion model.

from the value of 1 the comparison of calculated drift mobilities with experimental Hall mobilities is fully appropriate.

D. Mobility: Results of different models

Next, we analyze different contributions to the total mobility of the material and use the results obtained to consider the possibility to obtain accurate mobility results without performing the whole procedure of evaluating electron-phonon coupling constants on a dense reciprocal space grid.

In Fig. 8 we plot the results that are obtained when only one phonon mode is included in the calculation. The results indicate that mobility in the presence of scattering at longitudinal optical phonons (mode 6) is very similar to the total mobility at temperatures around the room temperature. This implies that LO-phonon scattering is the dominant scattering mechanism at these temperatures. At low temperatures, the mobility in the presence of scattering at acoustic phonons (modes 1–3) becomes comparable or even smaller than the mobility in the presence of LO-phonon scattering. These modes then provide the dominant scattering mechanism (We note that in realistic samples at low temperatures or at high impurity concentration one should as well consider impurity scattering as an important scattering mechanism). Such a behavior is fully expected and has been identified starting from the early studies [66] of transport in these materials. The origin of such a behavior comes from the fact that at low temperatures there are very few LO phonons and very few electrons at the states from which LO phonon emission could be possible.

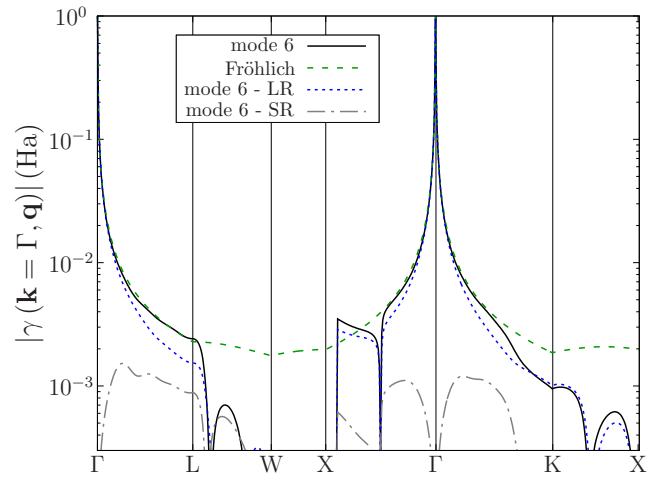


FIG. 9. Electron-phonon coupling constants for scattering of an electron at wave vector $\mathbf{k} = \Gamma$ with a mode 6 phonon at wave vector \mathbf{q} in ZnSe. The label “mode 6” denotes the results of the full calculation, “mode 6 - LR” and “mode 6 - SR” denote its long-ranged and short-ranged part, while the label “Fröhlich” denotes the result for the Fröhlich model.

Given the fact that mobility is mostly determined by LO phonon scattering at higher temperatures, it is interesting to consider if the same results could be obtained using some relevant model for electron-phonon interaction without performing the evaluation of electron-phonon coupling constants on the dense grid. The model that is expected to provide a reasonably good description of the interaction between electrons and LO phonons is the Fröhlich model since we consider direct band gap materials where all relevant electronic states are at the vicinity of the Γ point and no intervalley scattering effects are relevant. Within the Fröhlich model, the electron-phonon coupling constant is given as

$$|\gamma(\mathbf{k}, \mathbf{q})|^2 = \frac{\hbar e_0^2 \omega_{LO}}{2V \epsilon_0} \left(\frac{1}{\epsilon_r^\infty} - \frac{1}{\epsilon_r^{st}} \right) \frac{1}{q^2}, \quad (6)$$

where $\hbar \omega_{LO}$ is the LO phonon energy, V is the volume of the primitive cell of the crystal and ϵ_r^{st} is the static relative dielectric constant. The values of the parameters used in the Fröhlich model are summarized in Supplemental Material [29] (Sec. XIII). The results obtained with electron-phonon coupling from Eq. (6) and by taking into account only the LO phonon mode are also shown in Fig. 8 (labeled as “Fröhlich”). These results are very similar to the results obtained when only mode 6 is considered and consequently they are close to the full mobility results at higher temperature when mode 6 provides the dominant scattering mechanism. To better understand the origin of such a good agreement we compare in Fig. 9 the electron-phonon coupling constants for mode 6 in the full calculation and within the Fröhlich model. The figure shows that the long-ranged part of electron-phonon interaction dominates over the short-ranged part and that it nicely matches the Fröhlich model in the relevant range of \mathbf{q} vectors around the Γ point.

The agreement between electron-phonon coupling constants further leads to the agreement in scattering times. In Fig. 10 we present the dependence of scattering time on carrier

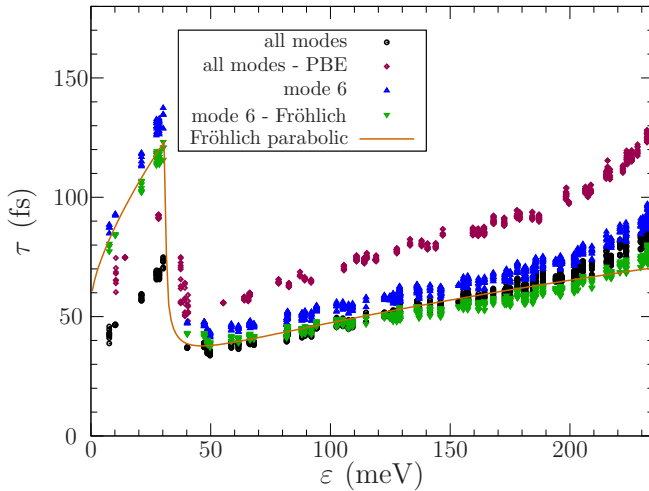


FIG. 10. Dependence of momentum relaxation time on electron energy at a temperature of $T = 300$ K in ZnSe. The label “all modes” denotes the results of the full calculation, the label “all modes - PBE” refers to the results of the full calculation when PBE functional is used, the label “mode 6” denotes the results when only phonon mode 6 is included, the label “mode 6 - Fröhlich” denotes the results for the Fröhlich model for mode 6, while the label “Fröhlich parabolic” denotes the results for the Fröhlich model with parabolic band structure.

energy with respect to the bottom of the conduction band. There is an overall good agreement between the scattering time due to mode 6 and the scattering times in the Fröhlich model. The scattering times caused by all phonon modes differ from these at low energies where LO phonon emission is not possible and where main contribution therefore comes from acoustic phonons. Due to this difference, it is not possible to accurately predict mobility at lower temperatures using a model that includes LO phonons only. All conclusions stemming from Figs. 8, 9, and 10 carry over to other materials, while analogous figures are given in Supplemental Material [29] (Secs. XIV, XV, and XVI therein).

We also note that the results indicate that the momentum relaxation times increase when electron energy increases except in the region around LO phonon energy when phonon emission events become energetically allowed. There are two opposing factors that determine the dependence of momentum relaxation time on electronic energy. On the one hand, for larger energy, the phase space for scattering increases and for this reason one would expect the momentum relaxation time to decrease. On the other hand, for larger energy, the phonon momenta involved in a scattering event become larger on average (when electron energy is sufficiently large that LO phonon emission is possible) and hence the scattering matrix element becomes smaller, an effect that would lead to an increase of momentum relaxation time. Which of these two opposing effects will prevail can be concluded with certainty only from a full calculation. In Sec. XVII of the Supplemental Material [29] we show an analytic expression in the case of the Fröhlich model with parabolic band structure. The results of this model (shown in Fig. 10 and in Figs. 42–44 in Supplemental Material [29]) exhibit essentially the same behavior as the results of the full calculation. Such an agreement is certainly not expected to extend to large energies above the bottom of the conduction

band. For example, it was shown in Ref. [67] that in GaN the scattering rates from the Fröhlich model agree with full calculation up to energies of ~ 150 meV above the bottom of the conduction band, while in the case of GaAs this agreement cannot extend up to more than ~ 250 meV when L valley states start to play a role [13]. We find that in the case of ZnSe this agreement extends up to ~ 600 meV. Such conclusion was drawn from Fig. 45 in Sec. XVIII of Supplemental Material [29]. That section contains also a more detailed discussion on the relation of our results for momentum relaxation times and scattering times and the results of Refs. [67] and [13].

Comparison of the momentum relaxation times for calculations with PBE and HSE06 functional gives as an additional insight why the HSE06 calculation gives lower values of mobility. As already discussed, the HSE06 calculation yields a higher effective mass than PBE calculation. It is not obvious whether a higher effective mass would lead to larger or smaller momentum relaxation times, because two opposing effects stemming from effective mass lead to the change of momentum relaxation time. On the one hand, larger effective mass leads to smaller phase space for scattering, which would lead to increased momentum relaxation time. On the other hand, for larger effective mass, the phonon momenta involved in the scattering event are smaller on average (when electron energy is sufficiently large that LO phonon emission is possible), the scattering matrix element is therefore larger, which would lead to decreased momentum relaxation time. In addition, HSE scattering matrix elements are larger because of smaller dielectric constant, which would lead to decreased momentum relaxation time. The results shown in Fig. 10 and in Figs. 42–44 in Supplemental Material [29] suggest that the effects that tend to decrease the momentum relaxation time prevail so that the momentum relaxation time is smaller in HSE06 calculation when the effective mass is larger. Larger effective mass also yields smaller band velocities. Therefore, given the fact that both the momentum relaxation times and the band velocities are smaller for larger effective mass, the mobility is also smaller for larger effective mass. These results confirm the well-known fact that small effective masses are desirable to have large mobility.

Next possible simplification in the evaluation of the mobility could be to even avoid the electronic structure and phonon band structure calculation and assume a parabolic band structure, a dispersionless phonon mode and Fröhlich coupling between electrons and phonons. This is a textbook model of electrons, phonons, and their coupling in polar semiconductors. The results obtained within such a model are shown as well in Fig. 8 (labeled as “Fröhlich parab.”). These results differ very little from the previous model (where full electronic and LO phonon dispersions are considered), suggesting that these are a viable alternative for mobility calculation at higher temperatures.

However, if one is interested to obtain reasonably accurate results for phonon-limited mobility at lower temperatures, the approach where only the LO phonon mode is considered is clearly not appropriate. To obtain a simpler model, which could be accurate in the broader range of temperatures, we consider the possibility of including only the short-ranged or the long-ranged term in Eq. (18) in Supplemental Material [29] for electron-phonon scattering elements. The results

obtained when only the short-ranged term is included are presented in Fig. 8 (labeled as “SR”), while the results obtained when only the long-ranged term is included fully coincide with the full mobility result (labeled as “all modes”). We therefore conclude that the long-ranged part of electron-phonon interaction fully determines the mobility, while it can be evaluated directly from Eq. (17) in Supplemental Material [29] without the need to previously obtain electron-phonon coupling constants from DFPT and without the need to perform their interpolation.

IV. CONCLUSION

We conclude by pointing out several lessons learned from our *ab initio* calculations of electron mobility in representative II-VI semiconductors. Our study points to the necessity of having accurate band structure and dielectric constants as input to the calculation. For example, in the case of ZnSe one obtains twofold larger mobility at room temperature if the values obtained from PBE are directly used. While previous studies of other materials have also considered the issue of band structure and dielectric constants accuracy, the changes of mobility when local density approximation (LDA) or PBE results were replaced by more accurate results were relatively small (around 15% in the case of silicon in Ref. [9] and around 25% in the case of GaAs in Ref. [8]).

Since the whole approach that includes the interpolation of electron-phonon coupling constants to a dense grid is rather computationally demanding, we used the results obtained to benchmark possible simpler models that should yield compa-

table accuracy. Such models have the potential to be used in high-throughput screening of materials with desirable properties, where fast calculation of material properties in the screening process is needed.

We found that the Fröhlich model of electron-phonon coupling constants can give reasonably accurate results at temperatures around room temperature for these direct band gap materials where all relevant electronic states are at the vicinity of the Γ point and no intervalley scattering effects are present. Additional simplification of such model where parabolic bands are assumed gives essentially the same results. These two results stem from the fact that the band structure of the materials considered in the relevant range of energies at the bottom of the conduction band is nearly isotropic.

We also found that the long-ranged part of the electron-phonon interaction fully determines the mobility. Therefore, the approach where only this part is calculated is a viable alternative that avoids the need to perform interpolation of electron-phonon coupling constants.

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