

Guidelines for accurate and efficient calculations of mobilities in two-dimensional semiconductorsJiaqi Zhou^{1,*}, Samuel Poncé^{2,3,†} and Jean-Christophe Charlier^{1,‡}¹*Institute of Condensed Matter and Nanosciences (IMCN), Université catholique de Louvain (UCLouvain), 1348 Louvain-la-Neuve, Belgium*²*European Theoretical Spectroscopy Facility and Institute of Condensed Matter and Nanosciences (IMCN), Université catholique de Louvain (UCLouvain), 1348 Louvain-la-Neuve, Belgium*³*WEL Research Institute, Avenue Pasteur 6, 1300 Wavre, Belgium*

(Received 12 April 2024; revised 9 September 2024; accepted 13 September 2024; published 30 September 2024)

Emerging two-dimensional (2D) materials bring unprecedented opportunities for electronic applications. The design of high-performance devices requires an accurate prediction of carrier mobility in 2D materials, which can be obtained using state-of-the-art *ab initio* calculations. However, various factors impact the computational accuracy, leading to contradictory estimations for the mobility. Targeting accurate and efficient *ab initio* calculations for nonmagnetic materials, transport properties in III-V monolayers are reported using the Boltzmann transport equation, and the influences of the pseudopotential, quadrupole correction, Berry connection, and spin-orbit coupling (SOC) on mobilities are systematically investigated in this work. Our findings are as follows: (1) The inclusion of semicore states in pseudopotentials is important to obtain accurate calculations. (2) The variations induced by dynamical quadrupole and Berry connection when treating long-range fields can be 40% and 10%, respectively. (3) The impact of SOC can reach up to 100% for materials with multipeak bands. Importantly, although SOC notably modifies the electronic wave functions, it negligibly impacts the dynamical matrices and scattering potential variations. As a result, the combination of a fully relativistic electron calculation and a scalar-relativistic phonon calculation can strike a good balance between accuracy and cost. This work compares computational methodologies, providing guidelines for accurate and efficient calculations of mobilities in 2D semiconductors.

DOI: [10.1103/PhysRevB.110.125304](https://doi.org/10.1103/PhysRevB.110.125304)**I. INTRODUCTION**

Two-dimensional (2D) materials exhibit superior transport properties, leading to promising applications in electronic devices [1–3]. The atomic thickness of 2D semiconductors enables the efficient engineering of electronic properties through gate voltage control, making them promising for transistor applications [4,5]. Drift mobility quantifies the transport of carrier in a material in response to an electric field, and the prediction of carrier mobility is critical for the design of high-performance devices. A prevalent approach is to employ density functional theory (DFT) and density functional perturbation theory (DFPT) to evaluate the electron-phonon coupling (EPC) matrices [6]. However, the accurate calculation of mobility requires very dense momentum grids, which is generally too expensive to perform by direct DFPT calculations. To solve the problem of computational cost, Wannier functions can be used to interpolate EPC from coarse grids to fine grids [7]. Using this technique, efforts have been devoted to discovering 2D materials with high mobilities [8–13]. However, conclusions have been contradictory due to the various approximations used in different studies. Using *ab initio* calculations, Ref. [14] reported that the III-V

monolayers GaP and InP present electron mobilities of 935 and 1023 cm²/Vs, respectively. Interestingly, another work reported lower value of $\mu^e = 408$ and 542 cm²/Vs, respectively [15], and another one reported even lower values of 272 and 520 cm²/Vs [16] for the same materials. These discrepancies illustrate that the accurate prediction of carrier mobility can be complicated since many factors are involved, creating confusion on *ab initio* values of computed mobilities in 2D materials.

The atomic motions in 2D semiconductors generate long-range dynamical dipoles and quadrupoles that should be considered to accurately interpolate EPC. Moreover, spin-orbit coupling (SOC) can significantly modify the electronic structures in materials with heavy elements. Semicore states in pseudopotentials may also influence the hole effective mass. To understand and facilitate computation, it is necessary to elucidate the impacts on the mobilities in 2D materials caused by these factors in order to optimize the balance between the accuracy and the computational cost in *ab initio* calculations.

In this work, we focus on the III-V monolayers *MX* (*M* = Ga, In and *X* = P, As, Sb) while considering a moderate band gap and SOC strength. The influences on mobilities caused by pseudopotentials with different semicore states [17], the dynamical quadrupole [18–20], the Berry connection [12,21], and SOC are systematically studied. Their impacts are interpreted by investigating the momentum- and mode-resolved scattering rates. In addition, DFPT results are found to be negligibly affected by SOC; thus, it can

*Contact author: jiaqi.zhou@uclouvain.be†Contact author: samuel.ponce@uclouvain.be‡Contact author: jean-christophe.charlier@uclouvain.be

be neglected to accelerate computation. After we discuss methodologies, the temperature-dependent drift and Hall mobilities are computed. Rather than unity, which is commonly assumed in experiments, we find that the Hall factors range from 1.0 to 1.7.

II. DRIFT MOBILITY

The phonon-limited drift mobility of a carrier in a 2D semiconductor is calculated as [22,23]

$$\mu_{\alpha\beta} = \frac{-1}{S^{uc}n^c} \sum_n \int \frac{d^2\mathbf{k}}{\Omega^{BZ}} v_{n\mathbf{k}\alpha} \partial_{E_\beta} f_{n\mathbf{k}}, \quad (1)$$

where α and β are Cartesian directions and $\alpha = \beta$ in the plane for III-V monolayers. $\partial_{E_\beta} f_{n\mathbf{k}} = (\partial f_{n\mathbf{k}} / \partial E_\beta)|_{E=0}$ indicates the linear variation of the electronic occupation function $f_{n\mathbf{k}}$ in response to the electric field $\mathbf{E}$. S^{uc} is the unit cell area, and Ω^{BZ} is the first Brillouin zone (BZ) area. The band velocity of state $\varepsilon_{n\mathbf{k}}$ is given by $v_{n\mathbf{k}\alpha} = \hbar^{-1} \partial \varepsilon_{n\mathbf{k}} / \partial k_\alpha$, and n^c denotes the carrier concentration, which should be properly chosen since a low value will lead to a difficult convergence, while a too high value will not be intrinsic and will break the scaling relation between σ^c and n^c , as shown in Fig. S4 in the Supplemental Material (SM) [24]. In this work, we set $n^c = 10^{10} \text{ cm}^{-2}$, which we find to be a good compromise. $\partial_{E_\beta} f_{n\mathbf{k}}$ can be obtained by solving the linearized Boltzmann transport equation (BTE) [23]:

$$\begin{aligned} \partial_{E_\beta} f_{n\mathbf{k}} = & e v_{n\mathbf{k}\beta} \frac{\partial f_{n\mathbf{k}}^0}{\partial \varepsilon_{n\mathbf{k}}} \tau_{n\mathbf{k}} + \frac{2\pi \tau_{n\mathbf{k}}}{\hbar} \sum_{mv} \int \frac{d^2\mathbf{q}}{\Omega^{BZ}} |g_{m\nu}(\mathbf{k}, \mathbf{q})|^2 \\ & \times [(n_{\mathbf{q}\nu} + 1 - f_{n\mathbf{k}}^0) \delta(\varepsilon_{n\mathbf{k}} - \varepsilon_{m\mathbf{k}+\mathbf{q}} + \hbar\omega_{\mathbf{q}\nu}) \\ & + (n_{\mathbf{q}\nu} + f_{n\mathbf{k}}^0) \delta(\varepsilon_{n\mathbf{k}} - \varepsilon_{m\mathbf{k}+\mathbf{q}} - \hbar\omega_{\mathbf{q}\nu})] \partial_{E_\beta} f_{m\mathbf{k}+\mathbf{q}}, \end{aligned} \quad (2)$$

where $\tau_{n\mathbf{k}}$ is the total scattering lifetime and its inverse $\tau_{n\mathbf{k}}^{-1}$ is the scattering rate, given as

$$\begin{aligned} \tau_{n\mathbf{k}}^{-1} = & \frac{2\pi}{\hbar} \sum_{mv} \int \frac{d^2\mathbf{q}}{\Omega^{BZ}} |g_{m\nu}(\mathbf{k}, \mathbf{q})|^2 \\ & \times [(n_{\mathbf{q}\nu} + 1 - f_{n\mathbf{k}}^0) \delta(\varepsilon_{n\mathbf{k}} - \varepsilon_{m\mathbf{k}+\mathbf{q}} - \hbar\omega_{\mathbf{q}\nu}) \\ & + (n_{\mathbf{q}\nu} + f_{n\mathbf{k}}^0) \delta(\varepsilon_{n\mathbf{k}} - \varepsilon_{m\mathbf{k}+\mathbf{q}} + \hbar\omega_{\mathbf{q}\nu})], \end{aligned} \quad (3)$$

where $g_{m\nu}(\mathbf{k}, \mathbf{q})$ is the EPC matrix element denoting the amplitude of scattering between the $|n\mathbf{k}\rangle$ state and the $|m\mathbf{k} + \mathbf{q}\rangle$ state via a phonon of frequency $\omega_{\mathbf{q}\nu}$, $n_{\mathbf{q}\nu}$ is the Bose-Einstein distribution, and $f_{n\mathbf{k}}^0$ is the Fermi-Dirac occupation function.

III. COMPUTATIONAL DETAILS

To solve these equations, we compute wave functions and potential variations using the QUANTUM ESPRESSO package [25]. The norm-conserving pseudopotentials PSEUDO DOJO [17] were employed within the Perdew-Burke-Ernzerhof (PBE) parametrization of the generalized gradient approximation. The projector augmented wave and ultrasoft pseudopotentials from PSLIBRARY [26] were also tested, and both present consistent results within 5% with PSEUDO DOJO [17],

as shown in Fig. S14. Variable cell relaxation is performed with total energy convergence of 10^{-8} Ry, force convergence of 10^{-4} Ry/bohr, and pressure convergence of 0.1 kbar. Relaxed structures are given in Sec. S1 in the SM. The cut-off energy for wave functions is set to 120 Ry. We use the 2D Coulomb truncation scheme in Ref. [27] with a vacuum distance over 19 Å. Note that in the calculation at the zone center $\mathbf{q} = \Gamma$, a denser $\mathbf{k}$ grid is used in DFPT to get convergent electrostatic properties. Electron and hole mobilities are calculated using the EPW package [28,29], in which the electron-phonon coupling is interpolated from coarse $\mathbf{k}/\mathbf{q}$ grids to fine $\mathbf{k}/\mathbf{q}$ grids using Wannier functions while considering the dipole, quadrupole, and Berry connection [12,21]. A coarse $12 \times 12 \times 1$ $\mathbf{k}/\mathbf{q}$ grid is adopted in *ab initio* calculations; then a fine $\mathbf{k}/\mathbf{q}$ grid of $720 \times 720 \times 1$ is used for electron mobility, which involves a single valley in the BZ, and a $\mathbf{k}/\mathbf{q}$ grid of $360 \times 360 \times 1$ is used for hole mobility, which involves the whole BZ. A Fermi surface window of 0.3 eV is used in all calculations. Convergence tests were performed with respect to the grid density and Fermi surface window, as shown in Sec. S2. The temperature is set at 300 K if not otherwise specified. A phonon frequency cutoff of 1 cm^{-1} is employed. Parameter-free adaptive broadening [30] with a fixed floor value is applied in the energy-conserving δ functions as an approximation for the exact self-consistent broadening [31]. Since the broadening values can impact the results [14,15,32,33], fixed broadening values of 1, 2, and 5 meV are also tested (see details in Fig. S15). In short, all types of broadening can reach a convergence of 5%, suggesting that broadening around 5 meV can be a good choice. However, this value should be carefully tested to eliminate divergence and oversmoothing. The dynamical quadrupole is computed using linear response as implemented in ABINIT [34,35] with a $\mathbf{k}$ grid of $32 \times 32 \times 1$, together with the PSEUDO DOJO pseudopotentials without nonlinear core corrections or spin-orbit coupling [36]. Many-body corrections such as the GW approach can be further employed to improve the accuracy of mobility prediction [37,38]. However, the computations can be rather expensive, and the change in effective mass is limited for materials with moderate to large band gaps [14,23,39,40]. It should be noted that for materials with a small band gap such as BSb with an underestimated PBE gap of 0.3 eV, its effective mass can be greatly underestimated [14], making GW corrections essential for accurate mobility calculations. Nevertheless, the guideline proposed in this work should be negligibly affected by such corrections for materials with a sufficient band gap. In this work, all materials present DFT band gaps above 0.5 eV.

IV. METHODOLOGY

In this section, we focus on the impacts on mobilities induced by pseudopotentials, the dynamical quadrupole, the Berry connection, and SOC. Using the stringent pseudopotential table from PSEUDO DOJO [17] and considering SOC, the atomic structures of all the materials have been relaxed. We find that all the III-V monolayers present a buckled structure, in agreement with Refs. [16,41–43]. This buckling breaks the inversion symmetry, leading to a Rashba splitting when SOC is considered. Details related to atomic structures are given

TABLE I. Methodologies employed in this work. PP is the abbreviation for pseudopotential. Quad and Berry indicate whether the dynamical quadrupole and Berry connection are considered, respectively. FR denotes the fully relativistic calculation with SOC, while SR denotes the scalar-relativistic calculation without SOC. e denotes the treatment of the electronic calculation for wave functions, and ph denotes the treatment of the phonon calculation for scattering potential variations. Methodology 7 strikes a good balance between accuracy and cost when a “mix” approach is used for the treatment of e and ph, thus this methodology is suggested to be employed in calculations.

Number	Methodology notation	PP	Quad	Berry	e	ph
1	valencePP+Quad+Berry+FR	valence	yes	yes	FR	FR
2	standardPP+Quad+Berry+FR	standard	yes	yes	FR	FR
3	stringentPP+Quad+Berry+FR	stringent	yes	yes	FR	FR
4	standardPP+Quad+FR	standard	yes	no	FR	FR
5	standardPP+FR	standard	no	no	FR	FR
6	standardPP+Quad+Berry+SR	standard	yes	yes	SR	SR
7	standardPP+Quad+Berry+mix	standard	yes	yes	FR	SR

in Sec. S1 in the SM [24]. The InAs monolayer is taken as a representative material due to its moderate band gap and SOC strength. Results for the other monolayers can be found in the SM [24]. Methodologies are listed in Table I. Three types of control experiments are implemented for the drift mobility calculations, evaluating the impacts of pseudopotentials (methodologies 1–3), the dynamical quadrupole and Berry connection (2, 4, and 5), and the fully relativistic SOC effect (2 and 6). Another evaluation is performed for the computational efficiency (2 and 7) by employing the wave functions from the fully relativistic calculation and the scattering potential variation from the scalar-relativistic calculation.

A. Impact of pseudopotential

Three types of norm-conserving pseudopotentials from PSEUDO DOJO are used in this work. For elements with the principal quantum number $n \geq 3$, the valence pseudopotential includes only the valence electrons, the standard pseudopotential includes one full d orbital, and the stringent pseudopotential includes full s , p , and d orbitals. The details of the included orbitals can be found in Table S3 in the SM [24].

Figure 1 shows the electron and phonon dispersions of the InAs monolayer computed with valence, standard, and stringent pseudopotentials, respectively. SOC lifts the spin degeneracy of the top valence band states; thus, the coupling between the spin and orbital momentum leads to energy splitting around the Γ and K points, which tends to increase the mobility due to the reduction in available scattering channel [23]. Different pseudopotentials introduce small changes to the valence bands. The valence band maximum (VBM) is always located at the Γ point. For the valence pseudopotential, $\varepsilon_K = E_F - 140$ meV. However, for the standard and stringent pseudopotentials, the highest valence band presents $\varepsilon_K \approx E_F - 90$ meV, demonstrating that the semicore states can cause a discrepancy of 50 meV on the valence bands, and the standard pseudopotential is consistent with the stringent one. More intriguingly, phonon dispersions are strongly affected, as illustrated in Figs. 1(d)–1(f). Imaginary frequencies emerge around the Γ point in the valence pseudopotential calculation. In addition, the translational invariance is violated by the stringent pseudopotential, and the former can be recovered

by the acoustic sum rule, as demonstrated in Fig. S12 in the SM [24]. Overall, the quadratic ZA mode is derived with the standard pseudopotential.

The convergence of mobility requires a fine grid to sample the BZ, making DFPT calculation too expensive. Thus, maximally localized Wannier functions are used for the EPC calculation. To assess the quality of the Wannier interpolation, we compare the interpolated band structures in Figs. 1(a)–1(c) and the EPC matrix element with those obtained from a direct DFPT calculation. For simplicity, we compute the total

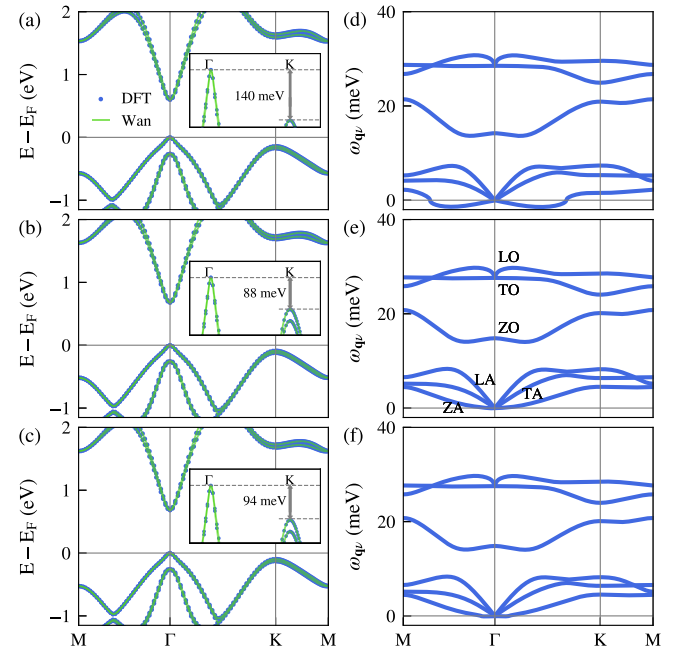


FIG. 1. InAs monolayer (a)–(c) electronic band structures and (d)–(f) phonon dispersions for fully relativistic calculations of valence, standard, and stringent pseudopotentials, respectively. The insets in (a)–(c) show a zoomed-in plot around the Fermi energy. Phonon modes are marked by ZA for the out-of-plane acoustic mode, TA for the transverse acoustic mode, LA for the longitudinal acoustic mode, ZO for the out-of-plane optical mode, TO for the transverse optical mode, and LO for the longitudinal optical mode.

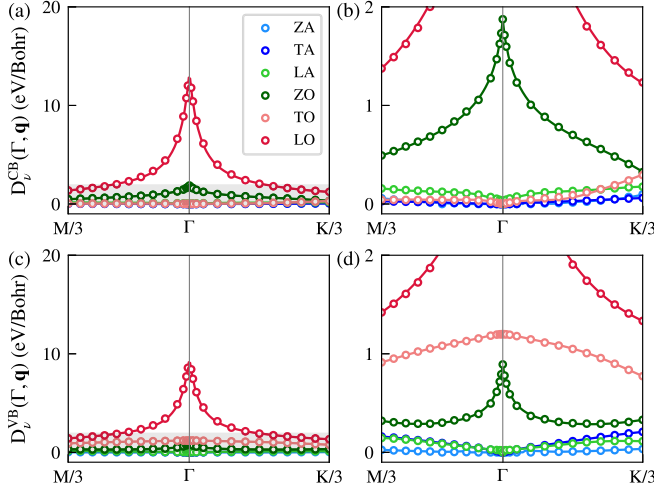


FIG. 2. Comparison between the deformation potentials of the InAs monolayer with the initial state $\mathbf{k} = \Gamma$ along high-symmetry lines; the DFPT and Wannier interpolation results are respectively denoted by circles and lines. The standard pseudopotential is employed. (a) Conduction and (c) valence bands are given in the left column; (b) and (d) the corresponding zoomed-in plots of the shaded region from 0 to 2 eV/bohr are given in the right column.

deformation potential [44] in the BZ center zone:

$$D_v(\Gamma, \mathbf{q}) = \frac{1}{\hbar N^w} \left[2\rho \Omega^{\text{BZ}} \hbar \omega_{\mathbf{q}v} \sum_{nm} |g_{mnv}(\Gamma, \mathbf{q})|^2 \right]^{1/2}, \quad (4)$$

where the $\mathbf{k} = \Gamma$ point is chosen, the sum over the bands is carried over the N^w states of the Wannier manifold, and ρ is the mass density of the crystal. It is found that deformation potentials calculated with different pseudopotentials are quite similar. Thus, only the result for the standard pseudopotential is presented in Fig. 2; the results for the valence and stringent pseudopotentials are given in Fig. S5 in the SM [24]. The Wannier interpolation reproduces the direct DFPT calculation quite well, hence validating its quality as well as the following computed transport properties. It should be noted that the convergence of the DFPT calculations of $\mathbf{q}$ points around Γ requires a denser $\mathbf{k}$ grid of DFT calculations. Here, a $32 \times 32 \times 1$ $\mathbf{k}$ grid is employed at $\mathbf{q} = \Gamma$, an $18 \times 18 \times 1$ $\mathbf{k}$ grid is used for $\mathbf{q}$ points around Γ , and a $12 \times 12 \times 1$ $\mathbf{k}$ grid is employed for $\mathbf{q}$ points around $M/3$ and $K/3$.

Figure 3 displays the mobilities of the III-V monolayers. Large electron mobilities around $1000 \text{ cm}^2/\text{Vs}$ are presented; notably, GaSb shows an ultrahigh μ^e around $1500 \text{ cm}^2/\text{Vs}$. The hole mobilities of GaP, InP, and GaAs are significantly suppressed, and relatively large μ^h are found in InAs, GaSb, and InSb. To make a more intuitive comparison, the ratios of the valence and stringent pseudopotential results to the standard pseudopotential result are displayed in Figs. 3(c) and 3(d), respectively, for electron and hole mobilities. For the electron mobility, the variation is limited to $\sim 10\%$. For the hole mobility, considerable variations are observed: The InAs monolayer has $\eta^h = 151\%$, and two phosphides even have η^h over 200%. It should be noted that in the valence pseudopotential results, imaginary frequencies appear in the phonon dispersions of the InAs, GaP, and InP monolayers, as

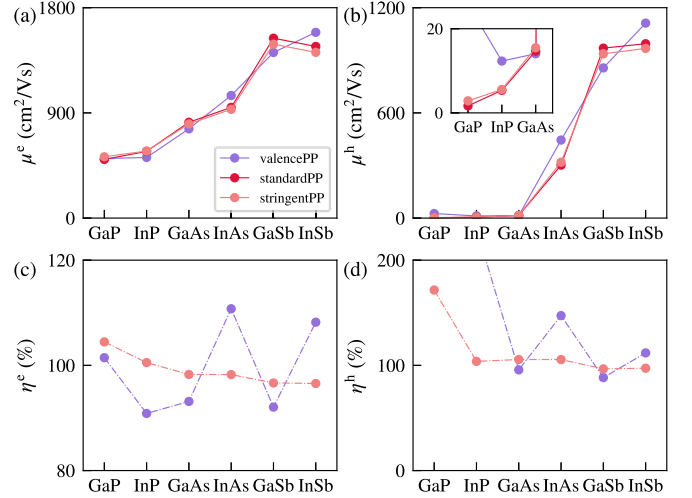


FIG. 3. (a) Electron and (b) hole mobilities calculated using different pseudopotentials according to methodologies 1–3. Taking the result of the standard pseudopotential as a benchmark, variations of the results of valence and stringent pseudopotentials are given as ratios in (c) for electrons and (d) for holes, respectively.

shown in Fig. 1(d) and Figs. S6 and S8 in the SM [24]. In our calculations, the modes $\mathbf{q}v$ with $\omega_{\mathbf{q}v} < 0$ have $g_{mnv}(\mathbf{k}, \mathbf{q}) = 0$; thus, the calculated result is an upper bound of mobility. In addition, the standard and stringent pseudopotentials present consistent behaviors in most cases, except for the μ^h of GaP since it is minimal and thus vulnerable to numerical errors in the ratio. The above discussion shows that the semicore states will influence the electronic structure and phonon dispersions, leading to variations in mobilities. Considering that standard and stringent pseudopotentials present consistent behaviors and applying the former can produce the correct phonon dispersion without an acoustic sum rule, we conclude that the standard pseudopotential is the most appropriate for the mobility calculation.

B. Impacts of the dynamical quadrupole and Berry connection

In the case of infrared active 2D materials, the Fröhlich interaction leads to a direction-dependent $g_{mnv}(\mathbf{k}, \mathbf{q})$ for $|\mathbf{q}| \rightarrow 0$ [45], hindering the application of Wannier interpolation. This nonanalytic behavior can be solved by separating $g_{mnv}(\mathbf{k}, \mathbf{q})$ into a short-range (S) and long-range (L) contributions [46] as

$$g_{mnv}(\mathbf{k}, \mathbf{q}) = g_{mnv}^S(\mathbf{k}, \mathbf{q}) + g_{mnv}^L(\mathbf{k}, \mathbf{q}), \quad (5)$$

where the short-range part is smooth and analytic in $\mathbf{q}$. The nonanalyticity is included in the long-range part, which is given by an explicit expression. Once it is obtained, $g_{mnv}(\mathbf{k}, \mathbf{q})$ can be accurately calculated by first subtracting $g_{mnv}^L(\mathbf{k}, \mathbf{q})$ to obtain $g_{mnv}^S(\mathbf{k}, \mathbf{q})$ on the coarse grid, then applying the Wannier interpolation to $g_{mnv}^S(\mathbf{k}, \mathbf{q})$, and finally adding $g_{mnv}^L(\mathbf{k}, \mathbf{q})$ back into the interpolated $g_{mnv}^S(\mathbf{k}, \mathbf{q})$ on the fine grid to obtain a complete $g_{mnv}(\mathbf{k}, \mathbf{q})$. Using the long-range scattering potential $V_{\mathbf{q}\kappa\alpha}^L$, which refers to the displacement of atom κ in direction α along a phonon mode $\mathbf{q}$, the expression of

TABLE II. Dynamical dipole Z (e) and quadrupole Q (e bohr), separation length L (bohr), dielectric and polarizability tensors (ϵ and α) of all the monolayers. Only independent components are presented. Note that the crystal symmetry constrains $Z_{\kappa xx} = Z_{\kappa yy}$, $Q_{\kappa xxz} = Q_{\kappa yyz}$, $Q_{\kappa xxy} = Q_{\kappa yxx} = -Q_{\kappa yyx}$, $Q_{\kappa zxx} = Q_{\kappa zyy}$, $\epsilon_{xx}^{\parallel} = \epsilon_{yy}^{\parallel}$, and $\alpha_{xx}^{\parallel} = \alpha_{yy}^{\parallel}$. Z and Q are atom-resolved thus occupying two columns per material. L , ϵ and α are material-resolved, thus they are merged into one column for one material.

Monolayer	Ga	P	In	P	Ga	As	In	As	Ga	Sb	In	Sb
$Z_{\kappa xx}$	2.956	-2.956	3.223	-3.223	2.946	-2.946	3.222	-3.222	2.765	-2.765	3.039	-3.039
$Z_{\kappa zz}$	0.132	-0.132	0.153	-0.153	0.089	-0.089	0.118	-0.118	0.046	-0.046	0.075	-0.075
$Q_{\kappa xxz}$	-0.858	-1.209	-1.007	-1.809	-1.444	-1.650	-0.260	-5.396	-1.996	-1.578	-2.139	-2.110
$Q_{\kappa xxy}$	14.158	-4.579	15.039	-4.345	16.900	-7.106	18.077	-6.348	21.026	-10.996	21.485	-10.062
$Q_{\kappa zxx}$	-11.905	7.813	-17.423	11.754	-26.850	20.669	-34.642	26.327	-41.400	34.332	-45.725	37.248
$Q_{\kappa zzz}$	0.638	-0.569	0.775	-0.666	1.150	-1.070	2.599	-6.637	1.454	-1.352	1.553	-1.434
L	9.852		12.257		16.218		18.973		27.624		26.270	
$\epsilon_{xx}^{\parallel}$	3.034		3.408		3.874		4.449		5.372		5.768	
$\epsilon^{\perp}$	1.152		1.160		1.161		1.169		1.174		1.180	
$\alpha_{xx}^{\parallel}$	6.117		7.245		8.644		10.375		13.150		14.341	
$\alpha^{\perp}$	0.458		0.483		0.486		0.509		0.525		0.544	

$g_{mnv}^{\mathcal{L}}(\mathbf{k}, \mathbf{q})$ is given as [21,46]

$$g_{mnv}^{\mathcal{L}}(\mathbf{k}, \mathbf{q}) = \left[\frac{\hbar}{2\omega_v(\mathbf{q})} \right]^{\frac{1}{2}} \sum_{\kappa\alpha} \frac{e_{\kappa\alpha v}(\mathbf{q})}{\sqrt{M_{\kappa}}} \sum_{sp} U_{ms\mathbf{k}+\mathbf{q}} \times \langle u_{s\mathbf{k}+\mathbf{q}}^{\mathbf{W}} | V_{\mathbf{q}\kappa\alpha}^{\mathcal{L}} | u_{p\mathbf{k}}^{\mathbf{W}} \rangle U_{pn\mathbf{k}}^{\dagger}, \quad (6)$$

where $e_{\kappa\alpha v}$ is the phonon eigenvector, M_{κ} is the atomic mass, and $U_{ms\mathbf{k}}$ denotes the Wannier rotation matrices applied to the periodic part of the wave functions expressed in the Wannier basis $|u_{n\mathbf{k}}^{\mathbf{W}}\rangle$.

Truncating the expansion at the order of $O(\mathbf{q})$, the long-range scattering potential is given as [12]

$$V_{\mathbf{q}\kappa\alpha}^{\mathcal{L}}(\mathbf{r}) = \frac{\pi e}{S_{uc}} \frac{f(|\mathbf{q}|)}{|\mathbf{q}|} e^{-i\mathbf{q}\cdot\boldsymbol{\tau}_{\kappa}} \left[\frac{1}{\tilde{\epsilon}^{\parallel}(\mathbf{q})} \{ 2i\mathbf{q} \cdot \mathbf{Z}_{\kappa\alpha} + \mathbf{q} \cdot \mathbf{q} \cdot \mathbf{Q}_{\kappa\alpha} - |\mathbf{q}|^2 Q_{\kappa\alpha zz} - 2\mathbf{q} \cdot \mathbf{Z}_{\kappa\alpha} \mathbf{q} \cdot V^{\text{Hxc}, \mathcal{E}}(\mathbf{r})/e \} + \frac{1}{\tilde{\epsilon}^{\perp}(\mathbf{q})} \{ 2|\mathbf{q}|^2 Z_{\kappa\alpha z} [z + V^{\text{Hxc}, \mathcal{E}}(\mathbf{r})/e] \} \right], \quad (7)$$

where $V^{\text{Hxc}, \mathcal{E}}$ denotes the self-consistent potential change induced by the electric field perturbation, $\boldsymbol{\tau}_{\kappa}$ denotes the position of atom κ within the cell, $\mathbf{Z}_{\kappa\alpha}$ is the dynamical dipole along the α direction, and $\mathbf{Q}_{\kappa\alpha}$ is the dynamical quadrupole. The range separation function $f(|\mathbf{q}|) = 1 - \tanh(|\mathbf{q}|L/2)$ is a low-pass Fourier filter that ensures the macroscopic character of the potential, where the parameter L defines the length scale. L is optimized by minimizing the sum of the real-space short-range interatomic force constants, see details in Eq. (63) in Ref. [12]. $\tilde{\epsilon}^{\parallel}$ and $\tilde{\epsilon}^{\perp}$ are the dielectric functions given by Eqs. (38) and (39) in Ref. [12]. The above parameters of III-V monolayers are given in Table II.

In addition to the expansion of $V_{\mathbf{q}\kappa\alpha}^{\mathcal{L}}$, the Wannier-gauge eigenstates are also expanded to the first order of Taylor series as

$$\langle u_{s\mathbf{k}+\mathbf{q}}^{\mathbf{W}} | = \langle u_{s\mathbf{k}}^{\mathbf{W}} | + \sum_{\alpha} q_{\alpha} \left\langle \frac{\partial u_{s\mathbf{k}}^{\mathbf{W}}}{\partial k_{\alpha}} \right|. \quad (8)$$

By introducing the Berry connection $A_{sp\mathbf{k},\alpha}^{\mathbf{W}} \equiv -i \langle \frac{\partial u_{s\mathbf{k}}^{\mathbf{W}}}{\partial k_{\alpha}} | u_{p\mathbf{k}}^{\mathbf{W}} \rangle$ and $\varphi_{\mathbf{q}}$ as the form factor for $V_{\mathbf{q}\kappa\alpha}^{\mathcal{L}}$, we obtain

$$\langle u_{s\mathbf{k}+\mathbf{q}}^{\mathbf{W}} | \varphi_{\mathbf{q}} | u_{p\mathbf{k}}^{\mathbf{W}} \rangle \approx \delta_{sp} + i\mathbf{q} \cdot [\langle u_{s\mathbf{k}}^{\mathbf{W}} | V^{\mathcal{E}} | u_{p\mathbf{k}}^{\mathbf{W}} \rangle + \mathbf{A}_{sp\mathbf{k}}^{\mathbf{W}}]. \quad (9)$$

The term $V^{\mathcal{E}}$ in Eq. (9) is omitted in our calculations due to its negligible contribution [18,21]. The term $\mathbf{A}_{sp\mathbf{k}}^{\mathbf{W}}$ involves the Berry connection, which is crucial since it ensures the smoothness of $g_{mnv}^{\mathcal{L}}(\mathbf{k}, \mathbf{q})$ and restores gauge covariance to the lowest order in $\mathbf{q}$ [12,21].

The effects of the two corrections mentioned above, namely, the dynamical quadrupole and the Berry connection, on the carrier mobility in III-V 2D semiconductors are investigated in this section. The standard pseudopotential and SOC are applied to the calculations. The quality of the Wannier interpolations is checked with the deformation potentials of the InAs monolayer. Figure 4 shows the result of the Wannier interpolation without Berry connection, which leads to slight deviations from the DFPT results for the LO modes. Figure 5 presents the Wannier result in the absence of both the Berry connection and dynamical quadrupole. For the ZO mode, neglecting quadrupole introduces large deviations from the DFPT results for both conduction and valence bands.

Figure 6 shows the impacts on the mobilities caused by the two types of corrections. For μ^e in phosphides, the impact of the Berry connection is large since the ZA, ZO, and TO modes are not well interpolated (details are given in Figs. S7 and S9 in the SM [24]). In other cases, the impact of the Berry connection is limited to 10%, attributed to the relatively good fittings as manifested by the InAs result in Fig. 4. Further neglecting dynamical quadrupole leads to a mobility underestimation for both electrons and holes for all III-V monolayers. The underestimation can reach up to 40% for the electron mobilities of all the materials, as well as for the hole mobilities in InAs and antimonides. Regarding phosphides and GaAs in which the hole mobilities are strongly suppressed by band structures with multiple peaks in the BZ, the effect of the quadrupole is also suppressed. Note that this conclusion, i.e., neglecting the quadrupole will underestimate the mobility, cannot be generalized for all the materials since scattering rates depend on not only the EPC matrix elements but also on

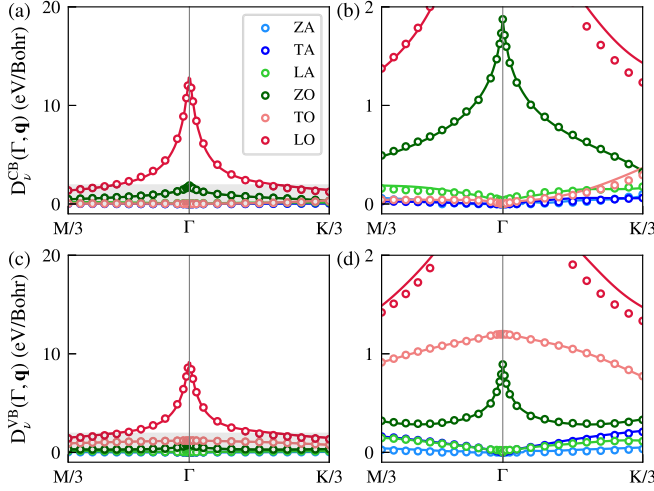


FIG. 4. Comparison between the InAs deformation potentials of DFPT and Wannier interpolations. Dynamical quadrupoles are considered, while the Berry connection is neglected for the Wannier interpolation. (a) Conduction and (c) valence bands are given in the left column; (b) and (d) the corresponding zoomed-in plots of the shaded region from 0 to 2 eV/bohr are given in the right column.

the conservation of energies and momenta. Moreover, Fig. 5 shows that the behavior of the Wannier deformation potential of the ZO mode is not systematic; namely, the Wannier deformation potential is higher than the DFPT around Γ but lower away from Γ . Thus, the influence of neglecting the quadrupole can be given only by calculations. In any case, neglecting the quadrupole will lower the quality of the Wannier interpolation, leading to unconvincing computational results.

The behavior of the carrier mobility can be interpreted by the scattering rates, which can be decomposed by phonon energy. Here, we focus on the electron mobility in the InAs

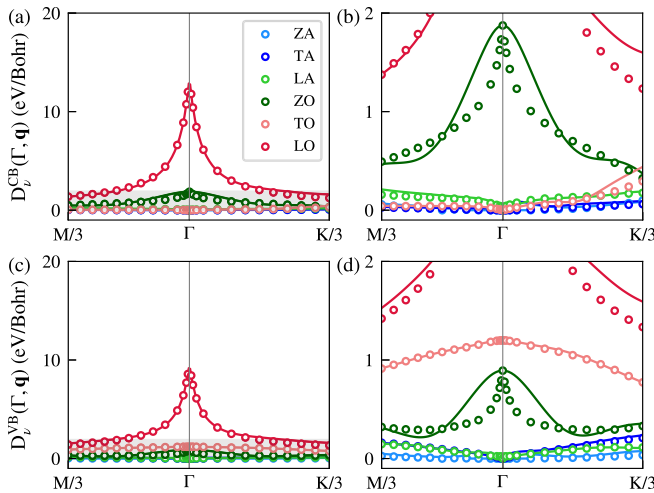


FIG. 5. Comparison between the InAs deformation potentials of DFPT and Wannier interpolations in the absence of both the dynamical quadrupole and Berry connection. (a) Conduction and (c) valence bands are given in the left column; (b) and (d) the corresponding zoomed-in plots of the shaded region from 0 to 2 eV/bohr are given in the right column.

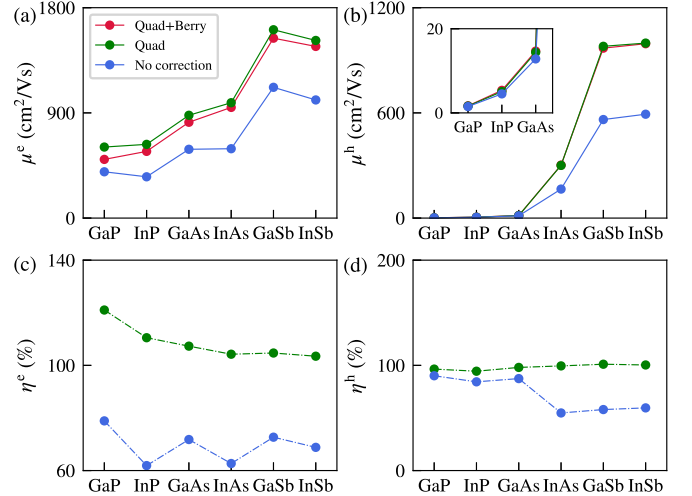


FIG. 6. (a) Electron and (b) hole mobilities calculated with and without corrections according to methodologies 2, 4, and 5. Taking the result with the quadrupole and Berry connection as a benchmark, variations of the results of neglecting the Berry connection and neglecting both corrections are given as ratios in (c) for electrons and (d) for holes, respectively.

monolayer. Considering an energy of $3/2k_B T = 39$ meV away from the band edge (see details in Ref. [47]), the phonon energy resolved scattering rates τ^{-1} computed in the presence or in the absence of both corrections are presented in Figs. 7(a) and 7(b), respectively. The total scattering rate is enhanced by the absence of corrections from 48 to 68 THz, increasing by $\sim 40\%$, demonstrating the main influence is caused by

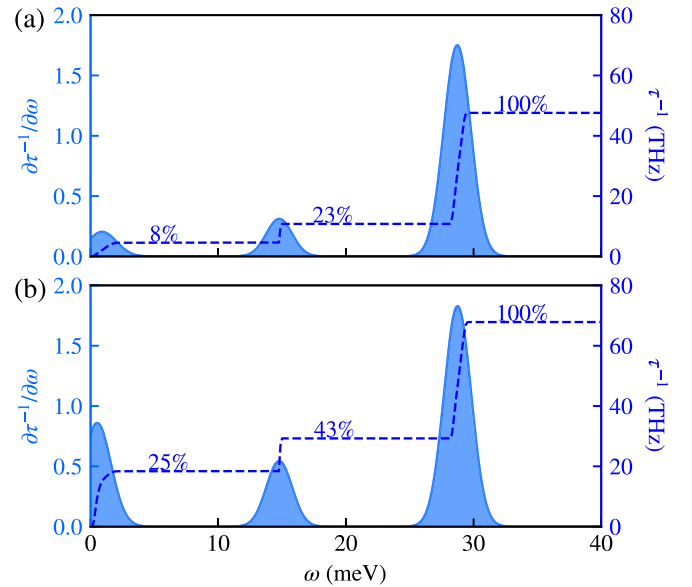


FIG. 7. Spectral decomposition of the electron scattering rates as a function of phonon energy of the InAs monolayer (a) in the presence and (b) in the absence of both the dynamical quadrupole and Berry connection. The peaks represent $\partial\tau^{-1}/\partial\omega$ (left axis), and the dashed lines represent the cumulative integral τ^{-1} (right axis). The percentages indicate the cumulative contribution to the total value of τ^{-1} .

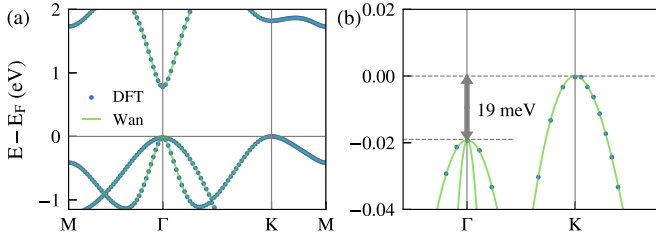


FIG. 8. (a) InAs monolayer electronic band structure calculated without SOC and (b) the corresponding zoomed-in plot around Fermi energy.

the dynamical quadrupole. In both cases, the scatterings are determined by the high-frequency phonons, i.e., the LO mode. Indeed, InAs is a polar material with one isotropic and parabolic conduction band, and its behavior can be described by the Fröhlich model [48]. Since the quadrupole has a negligible influence on the LO mode, the scatterings contributed by the LO mode (the area of the right peak) are nearly unchanged. On the other hand, the quadrupole strongly affects the ZO mode. When the quadrupole is neglected, the scattering contributed by the ZO mode is increased from 7.2 to 12.2 THz, given by the areas of the middle peaks, explaining the mobility underestimation in the absence of the quadrupole. The above discussion illustrates that the impacts of Berry connection are materials dependent. More importantly, the dynamical quadrupole can strongly affect the computed mobility by up to 40% for materials with high mobilities, illustrating the necessity to include quadrupole for their accurate predictions.

C. Impact of spin-orbit coupling

Arising from the interaction between the spin and orbital angular momenta, SOC impacts the motion of electrons and modifies the electronic structures, especially for materials with heavy atoms. In the fully relativistic calculation including SOC [see Fig. 1(b)], the conduction band minimum presents a Rashba splitting at the Γ point, analogous to a single valley in the BZ. In the InAs monolayer, the VBM is also located at the Γ point where the SOC splitting occurs, and $\varepsilon_K = E_F - 88$ meV. In the scalar-relativistic case (see Fig. 8), the edge of the conduction band becomes a single valley at the Γ point, and the valence bands are changed: The VBM is moved to the K point, the degeneracy at the Γ point is recovered due to crystal symmetry, and $\varepsilon_\Gamma = E_F - 19$ meV. The modifications are even more striking in GaSb and InSb (see Figs. S16 and S18 in the SM [24]). In contrast to the electronic structure, SOC induces a negligible change to the phonon dispersion, as shown in Fig. S12 in the SM [24], since the phonon dispersion is associated with lattice vibrations which are primarily determined by the masses of the nuclei and the interatomic forces in the lattice. The deformation potential from the scalar-relativistic (SR) calculation is very similar to that from the fully relativistic (FR) calculation (see Fig. S13 in the SM [24]).

Comparisons of the FR and SR calculated mobilities are shown in Fig. 9. The absence of SOC causes limited changes in electron mobilities for all materials, while more prominent variations emerge in hole mobilities. For phos-

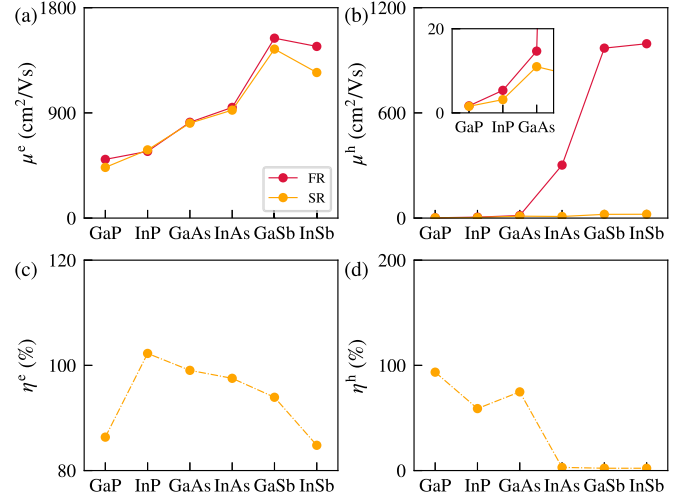


FIG. 9. (a) Electron and (b) hole mobilities calculated with FR and SR according to methodologies 2 and 6. Taking the FR result as a benchmark, variations of the SR result are given as ratios in (c) for electrons and (d) for holes, respectively.

phides and GaAs, the hole mobilities are strongly suppressed in both cases since the VBMs are always located at the K points in these materials. For InAs and antimonides, the hole mobilities are greatly suppressed by the absence of SOC.

The significant influence caused by SOC can be explained by the scattering rates of holes. The $\mathbf{k}$ -resolved scattering rates in FR and SR calculations of the InAs monolayer are, respectively, presented in Figs. 10(a) and 10(b). Compared with the FR scattering rates, SR scattering rates are enhanced, especially around the K points. This is because in the FR calculation, the VBM is located at the Γ point, which is a single scattering peak. In addition, eigenstates at the Γ and K points are separated by a large energy of 88 meV, increasing the barrier of scattering between Γ and K. In contrast, in the SR case, the VBMs are located at the K points which present multiple peaks of scattering in the BZ. Moreover, the energy barrier between eigenstates at the Γ and K points is reduced to 19 meV, increasing the interpeak scattering. Consequently, the hole mobility in the SR calculation is notably reduced.

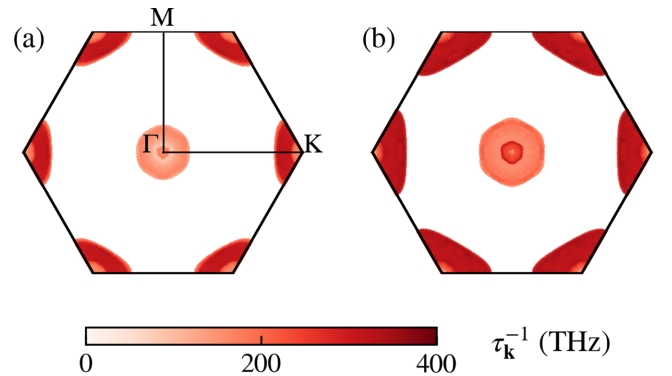


FIG. 10. Momentum-resolved scattering rates of holes in the InAs monolayer in (a) FR and (b) SR calculations, given by $\tau_k^{-1} = \frac{1}{N^w} \sum_n \tau_{nk}^{-1}$.

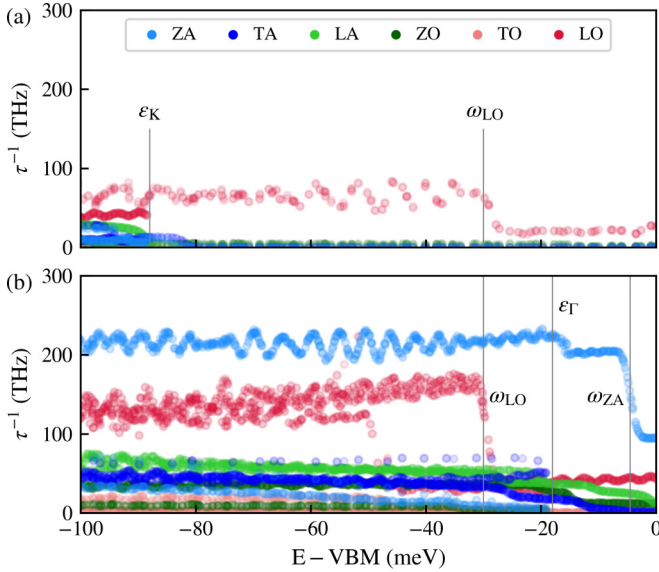


FIG. 11. Mode-resolved scattering rates of hole mobility in the InAs monolayer in the (a) FR and (b) SR calculations.

Additional understanding of the hole mobility can be gained through analysis of the mode-resolved scattering, whose behavior is determined by the electron and phonon dispersions due to the energy and momentum conservations. Figure 11(a) shows that in the FR calculation, the LO mode makes the dominant contribution in the energy range from the VBM to $\varepsilon_K = -88$ meV since there is only one band peak at the Γ point, analogous to a Fröhlich system. The increase at -30 meV is attributed to the LO mode with $\omega_{LO} \approx 30$ meV. Below ε_K , multiple peaks at K points are involved in the Fermi surface window, and the ZA mode starts to contribute to the intrapeak and interpeak scatterings with small phonon energies. As indicated by Fig. 11(b), in the FR case, ZA phonons dominate the scattering due to the intrapeak scattering at the K point. A sharp increase of the ZA contribution occurs around -4 meV since more phonons with $\omega_{ZA} \approx 4$ meV at the $\mathbf{q} = \mathbf{K}$ point [see Fig. 1(e)] are enabled for interpeak scatterings between K points in $\mathbf{k}$ space. When $\varepsilon_\Gamma = -19$ meV enters the Fermi surface window, the ZA contribution is further enhanced by the interpeak scattering between the Γ and K points. Furthermore, expanding the Fermi surface window allows the LO mode with $\omega_{LO} \approx 30$ meV to participate in the scattering. Since more phonons are involved, the scattering is enhanced in the SR calculation. The above discussion shows that SOC can significantly modify the electronic structure and lead to large mobility variations $\sim 100\%$, demonstrating the importance of considering SOC, especially for materials with heavy atoms and multiband edges.

D. Balance between accuracy and cost

Although SOC induces significant changes in the electronic structures, it negligibly affects the phonon dispersion and scattering potential variation. Considering the cost of DFPT calculations, we can use the electronic wave functions from the FR calculation and the scattering potential variations from the SR calculation to perform the Wannier interpolation

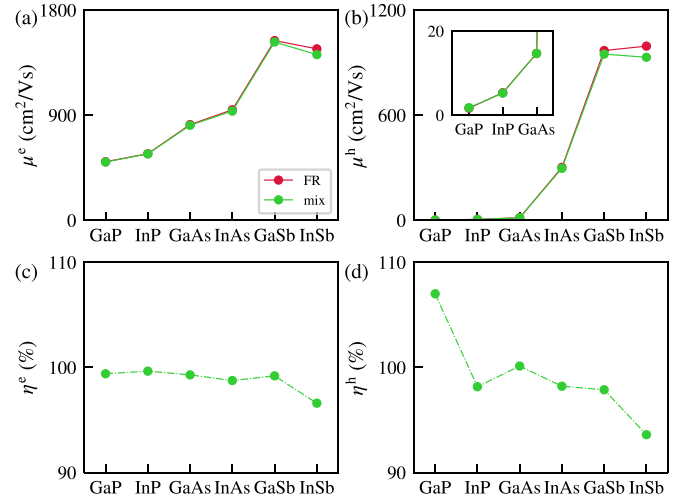


FIG. 12. (a) Electron and (b) hole mobilities calculated with the FR and the mix scheme according to methodologies 2 and 7. Taking the FR result as a benchmark, variations of the mix methodology are given as ratios in (c) for electrons and (d) for holes, respectively.

for EPC computation. The quality of the interpolation was validated by the corresponding Wannier and DFPT deformation potentials, as shown in Fig. S13 in the SM [24].

Figure 12 presents a comparison of the complete FR calculation and the mix methodology. For the electron mobility, the discrepancies are limited to less than 3%, while for the hole mobility, the discrepancies can be up to 7% in GaP and InSb. Still, the mix methodology produces a result consistent with the complete FR calculation, demonstrating its effectiveness.

Although causing a small loss in accuracy, the mix methodology can significantly enhance the calculation efficiency. The computational costs of the various methodologies mentioned above are summarized in Fig. 13, where the dynamical quadrupole and Berry connection are always considered. DFPT is always the most expensive calculation for all methodologies. The valencePP + FR and standardPP + SR methodologies are inexpensive but unreliable. Both

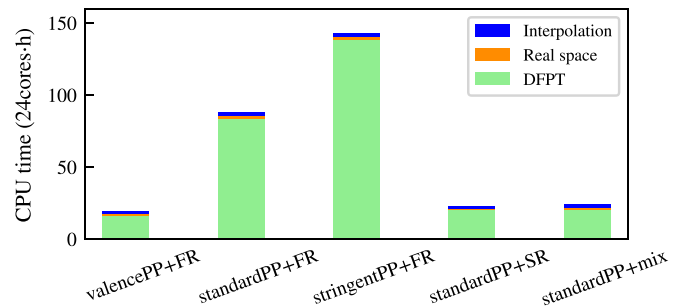


FIG. 13. Computational costs for different methodologies in the InAs monolayer. “DFPT” denotes the phonon calculation for potential variation with a self-consistency threshold of 10^{-18} . “Real space” denotes the EPW calculation for the representation transform from reciprocal to real space on the coarse grid. “Interpolation” denotes the interpolation from real to reciprocal space for transport properties on a fine grid. All the calculations are performed using 24 AMD EPYC 7H12 cores with a base clock speed of 2.6 GHz.

TABLE III. Electron and hole mobilities calculated using the standardPP+Quad+Berry+mix methodology (in cm^2/Vs).

Materials	μ^e	μ^h
GaP	499	1
InP	568	5
GaAs	813	14
InAs	935	296
GaSb	1526	948
InSb	1418	930

standardPP + FR and stringentPP + FR produce consistent results, while the latter is more expensive since more semicore states are included. The standardPP + mix methodology exhibits the same computational cost as the standardPP + SR in the DFPT calculation and a computational cost similar to that of the standardPP + FR in the EPW calculation. Consequently, the total cost of the standardPP + mix methodology significantly decreases and is 28% of the standardPP + FR cost and 17% of the stringentPP + FR cost. The above discussion suggests that the standardPP + mix methodology can strike a good balance between accuracy and cost. The mobilities of all the materials calculated with this methodology are listed in Table III, and the results for all the methodologies are given in Table S2 of the SM [24].

V. TEMPERATURE-DEPENDENT MOBILITIES

To show the most accurate results, we present the temperature-dependent mobilities calculated with the standardPP + Quad + Berry + FR methodology in Fig. 14. All the mobilities go down with increasing temperature. For the electron mobilities, all materials present a high μ^e greater than $100 \text{ cm}^2/\text{Vs}$, attributed to the single valley of the conduction bands. The mobilities increase with atomic number due to the decreasing effective mass, and the antimonides can reach $\mu^e \approx 10^4 \text{ cm}^2/\text{Vs}$ at 100 K. The decay of μ^e can be 1 order of magnitude from 100 to 500 K. For the hole mobilities, μ^h decays much faster with temperature. Still, antimonides can present high hole mobilities thanks to their single-peak VBMs.

Instead of drift mobility, Hall mobility is more commonly measured experimentally since it can be determined using a well-established setup, i.e., the Hall effect measurement. Indeed, such a transport measurement is performed under an external magnetic field, which induces a Lorentz force on the carriers and then changes the drift mobility μ to the Hall

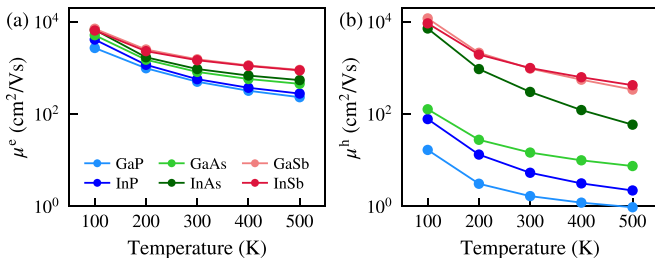


FIG. 14. Temperature-dependent (a) electron and (b) hole drift mobilities.

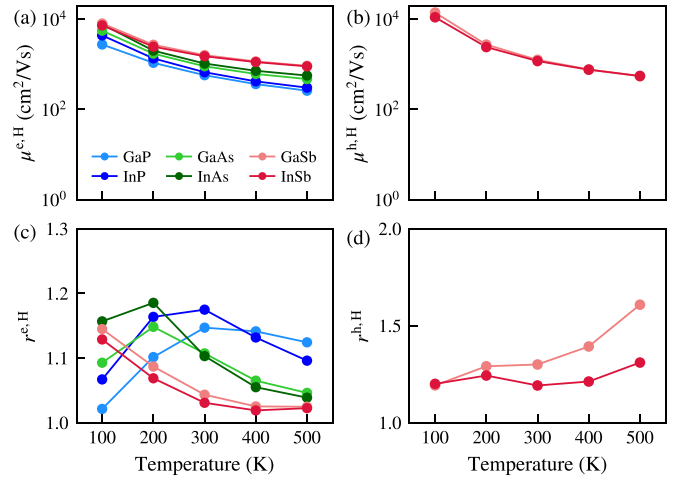


FIG. 15. Temperature-dependent (a) electron and (b) hole Hall mobilities, as well as (c) electron and (d) hole Hall factors.

mobility μ^H . Denoting the ratio between the Hall mobility and the drift mobility, the Hall factor is commonly assumed to be unity [49]. However, the suitability of this assumption should be carefully evaluated. The Hall mobility can be calculated by adding the external magnetic field to the BTE; for details see Eqs. (6)–(9) in Ref. [29]. In the present computations of the Hall factors, we apply an external magnetic field along the z direction with $B_z = 10^{-10} \text{ T}$, and we focus on the tensor element r_{xy}^H , which indicates the ratio of μ_{xy}^H over μ_{xx} ; the subscripts will be omitted in the following discussions.

Figure 15 presents the temperature-dependent Hall mobilities and Hall factors, depicting the materials with high mobilities which are more promising for experiments. For the electron transport, the magnetic field introduces limited changes to the mobility, as demonstrated by $r^{e,H}$, which is below 1.3 in all the materials. However, for the hole transport, $r^{h,H}$ can be as large as 1.7, as shown by the GaSb monolayer at high temperature. These theoretical results demonstrate that the unitary Hall factor can be a rough approximation, which may sometimes lead to the overestimation of the drift mobility in experiments.

VI. SUMMARY

Carrier mobilities investigated using DFPT and Wannier functions were reported for six semiconductors in the III-V monolayer family. The quality of the Wannier interpolation was validated by the comparison with DFPT calculations. To show the impacts induced by different approximations, several methodologies were proposed to investigate the influences on mobilities caused by semicore states, the dynamical quadrupole, the Berry connection, and spin-orbit coupling. Semicore states in pseudopotentials were found to be essential for accurate mobility calculations, the dynamical quadrupole can induce a variation of $\sim 40\%$, the Berry connection causes an impact of $\sim 10\%$, and SOC can even lead to a variation of $\sim 100\%$ for materials with multipeak electronic structures. The different mechanisms were interpreted by the momentum- and mode-resolved scattering rates. In addition, DFPT results were negligibly affected by SOC, which can be

neglected to accelerate computation with a precision loss of less than 7%. After we evaluated the various methodologies, temperature-dependent drift mobilities were computed, illustrating that the temperature can change the mobilities by 1 or 2 orders of magnitude, and the Hall factors range from 1.0 to 1.7. From the point of view of *ab initio* modeling, implementing certain approximations is mandatory and commonly accepted. Consequently, it is crucial to understand the extent of their impacts and evaluate their suitability. Striking the balance between accuracy and cost, this research can definitely provide guidelines for accurate and efficient calculations of carrier mobilities in 2D semiconductors. As a prospect, we hope this work can inspire experimental works to reveal the high mobilities in III-V monolayers.

For reproducibility, all information, including input files, software, pseudopotentials, and additional details, are provided in the Materials Cloud Archive [50].

ACKNOWLEDGMENTS

The authors would like to thank M. Giantomassi for fruitful discussions. S.P. acknowledges the support from the Fonds de la Recherche Scientifique de Belgique (F.R.S.-FNRS). J.Z. and J.-C.C. acknowledge financial support from the Fédération Wallonie-Bruxelles through the ARC Grant “DREAMS” (Grant No. 21/26-116), from the EOS project “CONNECT” (Grant No. 40007563), and from the Belgium F.R.S.-FNRS through the research project (Grant No. T.029.22F). Computational resources were provided by the PRACE award granting access to MareNostrum4 at the Barcelona Supercomputing Center (BSC), Spain, and Discoverer in SofiaTech, Bulgaria (OptoSpin Project ID No. 2020225411), and by the Consortium des Équipements de Calcul Intensif (CÉCI), funded by the F.R.S.-FNRS under Grant No. 2.5020.11 and by the Walloon Region, as well as computational resources awarded on the Belgian share of the EuroHPC LUMI supercomputer.

- [1] C. Zhang and Y. Liu, Phonon-limited transport of two-dimensional semiconductors: Quadrupole scattering and free carrier screening, *Phys. Rev. B* **106**, 115423 (2022).
- [2] X. Liu and M. C. Hersam, 2D materials for quantum information science, *Nat. Rev. Mater.* **4**, 669 (2019).
- [3] J. Zhou, S. Poncé, and J.-C. Charlier, Enhanced spin Hall ratio in two-dimensional semiconductors, *arXiv:2308.13692*.
- [4] F. Wu, H. Tian, Y. Shen, Z. Hou, J. Ren, G. Gou, Y. Sun, Y. Yang, and T.-L. Ren, Vertical MoS₂ transistors with sub-1-nm gate lengths, *Nature (London)* **603**, 259 (2022).
- [5] Y. Shen, Z. Dong, Y. Sun, H. Guo, F. Wu, X. Li, J. Tang, J. Liu, X. Wu, H. Tian, and T.-L. Ren, The trend of 2D transistors toward integrated circuits: Scaling down and new mechanisms, *Adv. Mater.* **34**, 2201916 (2022).
- [6] F. Giustino, Electron-phonon interactions from first principles, *Rev. Mod. Phys.* **89**, 015003 (2017).
- [7] F. Giustino, M. L. Cohen, and S. G. Louie, Electron-phonon interaction using Wannier functions, *Phys. Rev. B* **76**, 165108 (2007).
- [8] C.-H. Park, N. Bonini, T. Sohier, G. Samsonidze, B. Kozinsky, M. Calandra, F. Mauri, and N. Marzari, Electron-phonon interactions and the intrinsic electrical resistivity of graphene, *Nano Lett.* **14**, 1113 (2014).
- [9] L. Cheng, C. Zhang, and Y. Liu, The optimal electronic structure for high-mobility 2D semiconductors: Exceptionally high hole mobility in 2D antimony, *J. Am. Chem. Soc.* **141**, 16296 (2019).
- [10] T. Sohier, D. Campi, N. Marzari, and M. Gibertini, Mobility of two-dimensional materials from first principles in an accurate and automated framework, *Phys. Rev. Mater.* **2**, 114010 (2018).
- [11] T. Sohier, M. Gibertini, D. Campi, G. Pizzi, and N. Marzari, Valley-engineering mobilities in two-dimensional materials, *Nano Lett.* **19**, 3723 (2019).
- [12] S. Poncé, M. Royo, M. Stengel, N. Marzari, and M. Gibertini, Long-range electrostatic contribution to electron-phonon couplings and mobilities of two-dimensional and bulk materials, *Phys. Rev. B* **107**, 155424 (2023).
- [13] W.-H. Xiao, B. Zeng, Z.-K. Ding, H. Pan, W.-W. Liu, Q.-Q. Li, K. Yang, N. Luo, J. Zeng, K.-Q. Chen, and L.-M. Tang, Exceptionally high hole mobilities in monolayer group-IV monochalcogenides GeTe and SnTe, *Appl. Phys. Lett.* **123**, 013101 (2023).
- [14] S. Song, Y. Sun, S. Liu, J.-H. Yang, and X.-G. Gong, General rules and applications for screening high phonon-limited mobility in two-dimensional semiconductors, *Phys. Rev. B* **107**, 155101 (2023).
- [15] C. Zhang, R. Wang, H. Mishra, and Y. Liu, Two-dimensional semiconductors with high intrinsic carrier mobility at room temperature, *Phys. Rev. Lett.* **130**, 087001 (2023).
- [16] N. Hasani, A. Rajabi-Maram, and S. B. Touski, Effects of spin-orbit coupling on the electronic properties of the buckled III-V monolayers, *J. Magn. Magn. Mater.* **543**, 168638 (2022).
- [17] M. J. van Setten, M. Giantomassi, E. Bousquet, M. J. Verstraete, D. R. Hamann, X. Gonze, and G.-M. Rignanese, The PseudoDojo: Training and grading a 85 element optimized norm-conserving pseudopotential table, *Comput. Phys. Commun.* **226**, 39 (2018).
- [18] G. Brunin, H. P. C. Miranda, M. Giantomassi, M. Royo, M. Stengel, M. J. Verstraete, X. Gonze, G.-M. Rignanese, and G. Hautier, Electron-phonon beyond Fröhlich: Dynamical quadrupoles in polar and covalent solids, *Phys. Rev. Lett.* **125**, 136601 (2020).
- [19] G. Brunin, H. P. C. Miranda, M. Giantomassi, M. Royo, M. Stengel, M. J. Verstraete, X. Gonze, G.-M. Rignanese, and G. Hautier, Phonon-limited electron mobility in Si, GaAs, and GaP with exact treatment of dynamical quadrupoles, *Phys. Rev. B* **102**, 094308 (2020).
- [20] V. A. Jhalani, J.-J. Zhou, J. Park, C. E. Dreyer, and M. Bernardi, Piezoelectric electron-phonon interaction from *ab initio* dynamical quadrupoles: Impact on charge transport in wurtzite GaN, *Phys. Rev. Lett.* **125**, 136602 (2020).
- [21] S. Poncé, M. Royo, M. Gibertini, N. Marzari, and M. Stengel, Accurate prediction of Hall mobilities in two-dimensional materials through gauge-covariant quadrupolar contributions, *Phys. Rev. Lett.* **130**, 166301 (2023).
- [22] S. Poncé, W. Li, S. Reichardt, and F. Giustino, First-principles calculations of charge carrier mobility and conductivity in

- bulk semiconductors and two-dimensional materials, *Rep. Prog. Phys.* **83**, 036501 (2020).
- [23] S. Poncé, E. R. Margine, and F. Giustino, Towards predictive many-body calculations of phonon-limited carrier mobilities in semiconductors, *Phys. Rev. B* **97**, 121201(R) (2018).
- [24] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/PhysRevB.110.125304> for atomic structure and mobility data, convergence tests, electronic band structures, phonon bands, deformation potentials, scattering rates, and velocities of all 6 III-V monolayer semiconductors.
- [25] P. Giannozzi *et al.*, Advanced capabilities for materials modelling with Quantum ESPRESSO, *J. Phys.: Condens. Matter* **29**, 465901 (2017).
- [26] A. Dal Corso, Pseudopotentials periodic table: From H to Pu, *Comput. Mater. Sci.* **95**, 337 (2014).
- [27] T. Sohler, M. Calandra, and F. Mauri, Density functional perturbation theory for gated two-dimensional heterostructures: Theoretical developments and application to flexural phonons in graphene, *Phys. Rev. B* **96**, 075448 (2017).
- [28] S. Poncé, E. Margine, C. Verdi, and F. Giustino, EPW: Electron-phonon coupling, transport and superconducting properties using maximally localized Wannier functions, *Comput. Phys. Commun.* **209**, 116 (2016).
- [29] H. Lee, S. Poncé, K. Bushick, S. Hajinazar, J. Lafuente-Bartolome, J. Leveille, C. Lian, J.-M. Lihm, F. Macheda, H. Mori, H. Paudyal, W. H. Sio, S. Tiwari, M. Zacharias, X. Zhang, N. Bonini, E. Kioupakis, E. R. Margine, and F. Giustino, Electron-phonon physics from first principles using the EPW code, *npj Comput. Mater.* **9**, 156 (2023).
- [30] W. Li, J. Carrete, N. A. Katcho, and N. Mingo, ShengBTE: A solver of the Boltzmann transport equation for phonons, *Comput. Phys. Commun.* **185**, 1747 (2014).
- [31] J.-M. Lihm, S. Poncé, and C.-H. Park, Self-consistent electron lifetimes for electron-phonon scattering, *Phys. Rev. B* **110**, L121106 (2024).
- [32] J.-J. Zhou and M. Bernardi, *Ab initio* electron mobility and polar phonon scattering in GaAs, *Phys. Rev. B* **94**, 201201(R) (2016).
- [33] S. Li, Z. Tong, X. Zhang, and H. Bao, Thermal conductivity and Lorenz ratio of metals at intermediate temperatures with mode-level first-principles analysis, *Phys. Rev. B* **102**, 174306 (2020).
- [34] X. Gonze *et al.*, The ABINIT project: Impact, environment and recent developments, *Comput. Phys. Commun.* **248**, 107042 (2020).
- [35] M. Royo and M. Stengel, First-principles theory of spatial dispersion: Dynamical quadrupoles and flexoelectricity, *Phys. Rev. X* **9**, 021050 (2019).
- [36] <https://github.com/PseudoDojo/ONCVSP-PBE-SR-noCCv0.4>.
- [37] H. Jiang, Electronic band structures of molybdenum and tungsten dichalcogenides by the GW approach, *J. Phys. Chem. C* **116**, 7664 (2012).
- [38] J. Ma, A. S. Nissimagoudar, and W. Li, First-principles study of electron and hole mobilities of Si and GaAs, *Phys. Rev. B* **97**, 045201 (2018).
- [39] N. R. Knøsgaard and K. S. Thygesen, Representing individual electronic states for machine learning GW band structures of 2D materials, *Nat. Commun.* **13**, 468 (2022).
- [40] R. Rodrigues Pela, C. Vona, S. Lubeck, B. Alex, I. Gonzalez Oliva, and C. Draxl, Critical assessment of G0W0 calculations for 2D materials: The example of monolayer MoS₂, *npj Comput. Mater.* **10**, 77 (2024).
- [41] K. Wu, J. Chen, H. Ma, L. Wan, W. Hu, and J. Yang, Two-dimensional giant tunable Rashba semiconductors with two-atom-thick buckled honeycomb structure, *Nano Lett.* **21**, 740 (2021).
- [42] H. L. Zhuang, A. K. Singh, and R. G. Hennig, Computational discovery of single-layer III-V materials, *Phys. Rev. B* **87**, 165415 (2013).
- [43] H. Şahin, S. Cahangirov, M. Topsakal, E. Bekaroglu, E. Akturk, R. T. Senger, and S. Ciraci, Monolayer honeycomb structures of group-IV elements and III-V binary compounds: First-principles calculations, *Phys. Rev. B* **80**, 155453 (2009).
- [44] S. Poncé, F. Macheda, E. R. Margine, N. Marzari, N. Bonini, and F. Giustino, First-principles predictions of Hall and drift mobilities in semiconductors, *Phys. Rev. Res.* **3**, 043022 (2021).
- [45] P. Vogl, Microscopic theory of electron-phonon interaction in insulators or semiconductors, *Phys. Rev. B* **13**, 694 (1976).
- [46] C. Verdi and F. Giustino, Fröhlich electron-phonon vertex from first principles, *Phys. Rev. Lett.* **115**, 176401 (2015).
- [47] S. Poncé, M. Schlipf, and F. Giustino, Origin of low carrier mobilities in halide perovskites, *ACS Energy Lett.* **4**, 456 (2019).
- [48] F. Herbert, Interaction of electrons with lattice vibrations, *Proc. R. Soc. London, Ser. A* **215**, 291 (1952).
- [49] D. K. Schroder, *Semiconductor Material and Device Characterization* (Wiley-IEEE Press, Hoboken, New Jersey, USA, 2015).
- [50] J. Zhou, S. Poncé, and J.-C. Charlier, Materials Cloud Archive, <https://archive.materialscloud.org/record/2024.137>.