Drift and Hall mobility of two-dimensional materials from first principles

Samuel Poncé,1,2,∗ Miquel Royo,3 Marco Gibertini,4,5 Nicola Marzari,2,6 and Massimiliano Stengel1,7

1Institute of Condensed Matter and Nanosciences (IMCN), Université catholique de Louvain, Chemin des Étoiles 8, B-1348 Louvain-la-Neuve, Belgium
2Theory and Simulation of Materials (THEOS), École Polytechnique Fédérale de Lausanne, CH-1015 Lausanne, Switzerland
3Institut de Ciència de Materials de Barcelona (ICMAB-CSIC), Campus UAB, 08193 Bellaterra, Spain
4Dipartimento di Scienze Fisiche, Informatiche e Matematiche, Università di Modena e Reggio Emilia, Via Campi 213/a I-41125 Modena, Italy
5Centro S3, Istituto Nanoscienze-CNR, Via Campi 213/a, I-41125 Modena, Italy
6National Centre for Computational Design and Discovery of Novel Materials (MARVEL), École Polytechnique Fédérale de Lausanne, CH-1015 Lausanne, Switzerland
7Institució Catalana de Recerca i Estudis Avançats (ICREA), Pg. Lluís Companys, 23, 08010 Barcelona, Spain
(Dated: July 22, 2022)

Despite considerable efforts, accurate computations of electron-phonon and carrier transport properties of low-dimensional materials from first principles have remained elusive. By building on recent advances in the description of long-range electrostatic interactions [Phys. Rev. X 11, 041027 (2021)], we develop a general approach to the calculation of electron-phonon couplings in two-dimensional materials with an accuracy that can be systematically improved to arbitrary multipolar orders. We showcase this approach for a MoS\textsubscript{2} monolayer and compute its fully converged intrinsic drift and Hall carrier mobilities exploiting efficient Fourier-Wannier interpolations. We find that the contribution of dynamical quadrupoles to the scattering potential is essential, and that their neglect leads to errors of 19% and 43% in the room temperature Hall electron and hole mobilities.

Charge transport in two-dimensional (2D) materials lies at the heart of many technological applications ranging from transistors [1], valleytronics [2, 3], solar cells [4, 5], emitters [6], photodetectors [7], and transparent conductors [8]. It is therefore desirable to understand the different scattering mechanisms that govern carriers transport. In the case of high-quality samples with low doping concentrations, lattice scattering is the dominant mechanism limiting carrier mobilities [9]. Indeed, electron-phonon interactions [10] occurring at reduced dimensionality lie behind some unique features reported in recent years: lattice thermal conductivity suppression [11], unusual photoelectric effects [12], and classical superconductivity [13–15]. Given the current challenges with the experimental probing of electron-phonon interactions in 2D, theoretical studies based on \textit{ab-initio} simulations are crucial for future progress.

Recent advances in first-principles calculations of mobility (see Ref. 9 for a review of the field) have made it possible to study electronic transport in semiconductors. At the root of such calculations are the electron-phonon matrix elements $g_{\text{mnp}}(\mathbf{k}, \mathbf{q})$, that describe the scattering amplitudes from an initial electron Bloch state $\mathbf{nk}$ to a final state $\mathbf{mk} + \mathbf{q}$ via the emission or absorption of a phonon with frequency $\omega_{\mathbf{q} \mathbf{p}}$. One of the major challenges lies in achieving a numerically correct mobility, which often requires samplings of $g_{\text{mnp}}(\mathbf{k}, \mathbf{q})$ on ultra-dense momentum grids [16]. To make the problem tractable, an efficient way forward consists in explicitly computing the electron-phonon matrix element on coarse grids via state-of-the-art density-functional perturbation theory (DFPT) [17, 18] methods, followed by Wannier-Fourier interpolation to these ultra-dense grids [19, 20]. Maximally localized Wannier functions [21] are typically used as an exact and minimal representation.

However, this approach requires further care when dealing with the long-range electrostatic fields that arise near the Brillouin-zone center in semiconductors and insulators. The leading Fröhlich interaction [22], together with higher-order multipolar contribution [23–27], makes the electron-phonon interactions nonanalytic in the long-wavelength limit, which precludes straightforward application of Fourier-Wannier interpolations [28, 29]. The strategy to tackle such problem rests on a formal analysis of the nonanalytic properties of the scattering potential [23, 24] and is now well established [30] in the context of 3D crystals. By contrast, the long-range electrostatic problem in 2D materials has not been thoroughly investigated, due to the challenge of reproducing the environment of a free-standing layer in periodic \textit{ab initio} codes. A major advance in this direction came from Sollier and co-workers [31, 32], with a formulation of the long-range electrostatic interactions that accounts for the effect of the in-plane dipoles and relies on 2D Coulomb truncation [33]. Nonetheless, the treatment of such interactions has been based on dielectric analogues, which correctly capture the leading Fröhlich-like term, but miss higher multipoles. Efforts in this direction have been reported recently [34], but a fundamental understanding of higher-order multipolar couplings in 2D is still missing.

Here, we build on recent advances in the description of dielectric screening of the interatomic force constants [35] and apply these to the electron-phonon interactions for 2D materials. We focus on drift and Hall carrier mobil-
ities, and take monolayer MoS$_2$ as a paradigmatic case study; such choice is motivated by its technological relevance, and the availability of many theoretical and experimental data for this system. Results for five additional representative 2D crystals are reported in our accompanying paper [36]. In particular, we show that mobilities are strongly affected by long-range effects, and that a correct treatment provides a room-temperature mobility 25% smaller than previous reports [34]. This highlights the importance of quadrupolar coupling in the description of electron-phonon interactions in semiconducting low-dimensional materials.

To deal correctly with these higher order terms, one decomposes $g_{mnν}(\mathbf{k}, q)$ into short- ($S$) and long-range ($L$) contributions, where the long-range part can be formally expressed in terms of a scattering potential $V^L_{q,α}$, referring to the displacement of atom $κ$ in the Cartesian direction $α$, as:

$$
g^L_{mnν}(\mathbf{k}, q) = \left[ \frac{ℏ}{2ω(\mathbf{q})} \right]^\frac{1}{2} \sum_{κα} \frac{ε_{καρ}(q)}{\sqrt{M_κ}} \sum_{sp} \epsilon_{καρ}(q) \times U_{ms,k+q}W^{W}_{sp,k+q}|V^L_{q,κα}|u^{W}_{p,k}|u^{W}_{p,k}. \tag{1}$$

Here we have changed the representation to the maximally localized Wannier gauge to guarantee a smooth behavior in $q$ of the cell-periodic part of the Bloch eigenstates ($|u_{nk}\rangle = ∑_p U_{np,k}|u^{W}_{p,k}\rangle$ and where the $ε_{καρ}$ are the dynamical matrix eigenstates corresponding to a phonon mode $ν$ of momentum $q$).

In its simplest form, the long-range scattering potential in 2D can be written (detailed derivation in 36) as

$$V^L_{q,κα}(r) = \frac{e}{S} \frac{2πf(q)}{q} \frac{iq·Z^∥_{q,κα}(q)}{ε_{κα}(q)} \varphi^∥_q(z)e^{-iq·τ_κ}. \tag{2}$$

Here, $S$ is the unit-cell area, $τ_κ$ stands for the position of atom $κ$ within the cell, $z$ is the out-of-plane coordinate and $q = |q|$. The range separation function $f(q) = 1 - tanh(qL/2)$ is a low-pass Fourier filter that ensures the macroscopic character of the potential, where the parameter $L$ defines the length scale [35]. Note that the choice of $L$ is, to a large extent, arbitrary and can be tuned to maximize the numerical efficiency of the interpolation [69]. Eq. (2) can be intuitively interpreted as the bare long-range Coulomb kernel in 2D, $ν^{2D}(q) = 2πf(q)/q$, multiplied by a screened surface polarization charge and by a form factor $\varphi^∥_q(z)$. The latter reflects the fact that the electrostatic potentials produced by a modulated plane of charge are nonuniform along the out-of-plane direction [35]. In turn, the screened charge is written as the divergence of the polarization field associated with the displacement of atom $κ$ along the in-plane Cartesian direction $α$, $Z^∥_{q,κα}(q)$, divided by the macroscopic in-plane dielectric function $ε^∥(q) = 1 + ν^{2D}(q)α^∥(q)$, where $α^∥(q)$ is the macroscopic in-plane polarizability [70]. In the long-wavelength limit, $Z^∥_{q,κα}(q)$ can be conveniently expressed in a multipole expansion as

$$Z^∥_{q,κα}(q) = Z_{καβ} - i ∑_{γ} \frac{2}{2π} (Q_{καβγ} - δ_{καβ}Q_{κzzz}) + O(q^2).$$

Here $Z_{καβ}$ is the dynamical Born effective charge tensor [17, 18], and $Q_{καβγ}$ is the dynamical quadrupole tensor, providing the first-order spatial dispersion correction to the Born charges [37, 38]. Note that Eq. (2) describes the mirror-even part of the long-range scattering potential, while mirror-odd contributions mediated by out-of-plane electric fields have been neglected. The latter vanish by symmetry when the initial and final Bloch states have the same parity, such as in Fig. 1 for MoS$_2$; a test on an hexagonal BN monolayer [36] suggests that their contribution is indeed small.

The explicit expression in Eq. (2) for the scattering potential allows to construct the corresponding matrix element in Eq. (1), which can be subtracted from the ab-initio results, making these short-ranged and analytic in $q$, and amenable to accurate Fourier-Wannier interpolations. Adding the non-analytic expressions back then allows to obtain the electron-phonon matrix elements over arbitrarily dense grids [28, 29]. We note in passing that, in order to recover the correct $q → 0$ expansion of Eq. (1), we also need the expansion to first order of the matrix elements of the form factor,

$$(u_{sk+q}^W|\varphi^∥_q|u_{p,k}^W) ≃ δ_{sp} + iq·(A^W_{sp;k} + (u_{sk+q}^W|V|u_{p,k}^W). \tag{3}$$

where we have exploited that the Wannier gauge is smooth everywhere in the Brillouin zone. The expansion of Eq. (3) therefore involves two terms beyond Ref. [28]: one involving the matrix elements of the self-consistent potential (“local fields”) response to a uniform electric field $E$ already identified in the 3D case [23, 24]—and a previously unreported contribution arising from the Berry connection $A^W_{sp;k}$—both occur at the same order in $q$ as the dynamical quadrupoles. Preliminary tests [36] and previous experience with 3D crystals [23, 24] suggest that these additional terms are small in the considered cases and therefore neglect them in the calculations presented here. With these capabilities, we are now in a position to obtain the low-field phonon-limited carrier mobility in the presence of a vanishing magnetic field $B$ by solving the Boltzmann transport equation (BTE) [9, 27, 39]. From this solution we can also compute the Hall factor $r_{αβ}(B)$ as the ratio between mobilities with/without magnetic field [40–42].

We perform our calculations using the EPW [19, 43], WANNIER90 [44], and QUANTUM ESPRESSO [45] packages, including spin-orbit coupling (SOC) and using fully converged computational parameters [71]. In addition, we use the long-wave driver of ABINIT [46, 47] to compute the quadrupoles [38] in absence of SOC. The linearly-independent quadrupoles obtained are $Q_{κyyyy} = 5.533$ e-Bohr for Mo, and $Q_{κyyyy} = 0.391$, $Q_{κyyz} = -0.174$, $Q_{κzzz} = 7.858$, and $Q_{κzzz} = 0.239$ e-Bohr for one of the two S atoms.
of Ref. 35. We find no visible differences between the distinct schemes for the two approaches in this particular calculation; obviously, this is not a general conclusion [35]. As mentioned, here the mirror-even phonon branches are crucial, as they mediate electronic transitions that are major contributions to the mobilities. In MoS$_2$ there are three sets of mirror-even branches: the zone-center infrared and Raman active E$'$ mode associated with an in-plane out-of-phase movement of the Mo and S atoms, which splits into LO$_2$ and TO$_2$ branches at finite momentum; the Raman active A$_1$ mode associated with the out-of-plane motion of the sulfur atoms while the molybdenum atoms remain fixed (ZO$_1$ branch); and the zone-center E$'$ acoustic mode that splits into the LA and TA branches at finite momentum.

In Fig. 1(b) and (c) we show the Wannier-interpolated deformation potentials [29, 52] for all the phonon branches, and compare these with direct DFPT calculations (black circles). As expected, the mirror-odd ZA, LO$_1$, TO$_1$, and ZO$_2$ modes are inactive along the high symmetry directions shown in the figure. In contrast to the phonon dispersion of Fig. 1(a), the addition of dynamical quadrupoles is essential to recover the correct ZO$_1$ deformation potential. In 36, we also show how the dipole approximation would yield deformation potentials with quantitatively and qualitatively incorrect long-wavelength dispersions for different phonon branches for each type of 2D material considered. In all these cases, the inclusion of the quadrupolar fields recovers the correct dispersions. We also compare our dipolar results (the orange lines in Fig. 1) with those provided by the simplified dipole-based formalism of Ref. 32, and find no appreciable differences (not shown). This means that our correct description of the in-plane screening, which enters the scattering potential via the dielectric function $\varepsilon(k)$ at the denominator of Eq. (2), has a small impact on the interpolation of MoS$_2$ deformation potentials. However, this is not always the case and we report these differences for other 2D materials in Ref. 36.

Finally, we show in Fig. 2 the intrinsic Hall electron and hole mobilities that are obtained considering only the dipoles, or dipoles and quadrupoles, on dense 500 $\times$ 500 $k$ and $q$ momentum grids. The latter yield room temperature Hall mobilities of 142 cm$^2$/Vs and 117 cm$^2$/Vs for electrons and holes, respectively. Importantly, the most noticeable result is the dramatic impact that the inclusion of the quadrupoles in the interpolation entails on the final results: the room-temperature Hall mobility is reduced by 19% for electrons and by 43% for holes, when compared to the dipoles-only case. The comparison with the intrinsic drift mobilities (not shown in the figure, and of 132 cm$^2$/Vs and 73 cm$^2$/Vs), reveals that the holes are more strongly affected by the presence of a magnetic field. Besides, in the right panels of Fig. 2 we show how the hole Hall factor strongly increases with temperature, doubling from 100 K to 500 K, while the electron Hall factor is almost unaffected. In the inset of Fig. 2(c) we show that 87% of the electron mobility of this MoS$_2$ monolayer

In Fig. 1(a) we present the phonon dispersions along two high-symmetry directions, using either only dipoles, or dipoles and quadrupoles in the Fourier interpolations of the IFC, which we carry out following the approach of Ref. 35. We find no visible differences between the two approaches in this particular calculation; obviously, this is not a general conclusion [35]. As mentioned, here the mirror-even phonon branches are crucial, as they mediate electronic transitions that are major contributions to the mobilities. In MoS$_2$ there are three sets of mirror-even branches: the zone-center infrared and Raman active E$'$ mode associated with an in-plane out-of-phase movement of the Mo and S atoms, which splits into LO$_2$ and TO$_2$ branches at finite momentum; the Raman active A$_1$ mode associated with the out-of-plane motion of the sulfur atoms while the molybdenum atoms remain fixed (ZO$_1$ branch); and the zone-center E$'$ acoustic mode that splits into the LA and TA branches at finite momentum.

In Fig. 1(b) and (c) we show the Wannier-interpolated deformation potentials [29, 52] for all the phonon branches, and compare these with direct DFPT calculations (black circles). As expected, the mirror-odd ZA, LO$_1$, TO$_1$, and ZO$_2$ modes are inactive along the high symmetry directions shown in the figure. In contrast to the phonon dispersion of Fig. 1(a), the addition of dynamical quadrupoles is essential to recover the correct ZO$_1$ deformation potential. In 36, we also show how the dipole approximation would yield deformation potentials with quantitatively and qualitatively incorrect long-wavelength dispersions for different phonon branches for each type of 2D material considered. In all these cases, the inclusion of the quadrupolar fields recovers the correct dispersions. We also compare our dipolar results (the orange lines in Fig. 1) with those provided by the simplified dipole-based formalism of Ref. 32, and find no appreciable differences (not shown). This means that our correct description of the in-plane screening, which enters the scattering potential via the dielectric function $\varepsilon(k)$ at the denominator of Eq. (2), has a small impact on the interpolation of MoS$_2$ deformation potentials. However, this is not always the case and we report these differences for other 2D materials in Ref. 36.

Finally, we show in Fig. 2 the intrinsic Hall electron and hole mobilities that are obtained considering only the dipoles, or dipoles and quadrupoles, on dense 500 $\times$ 500 $k$ and $q$ momentum grids. The latter yield room temperature Hall mobilities of 142 cm$^2$/Vs and 117 cm$^2$/Vs for electrons and holes, respectively. Importantly, the most noticeable result is the dramatic impact that the inclusion of the quadrupoles in the interpolation entails on the final results: the room-temperature Hall mobility is reduced by 19% for electrons and by 43% for holes, when compared to the dipoles-only case. The comparison with the intrinsic drift mobilities (not shown in the figure, and of 132 cm$^2$/Vs and 73 cm$^2$/Vs), reveals that the holes are more strongly affected by the presence of a magnetic field. Besides, in the right panels of Fig. 2 we show how the hole Hall factor strongly increases with temperature, doubling from 100 K to 500 K, while the electron Hall factor is almost unaffected. In the inset of Fig. 2(c) we show that 87% of the electron mobility of this MoS$_2$ monolayer...
FIG. 2: Temperature dependence of the Hall carrier mobility for (a) electrons and (c) holes in MoS$_2$ as well as the electron (b) and hole (d) Hall factor. The dashed (solid) lines represent the mobilities calculated using electron-phonon interactions calculated considering only the dipole or dipole and quadrupole contributions; the temperature exponent for the mobility is also reported. The gray symbols refer to experimental data from Refs. [48–51]. The inset in (c) provides the spectral decomposition of the electron (orange) and hole (green) scattering rates at 300 K as a function of phonon energy, and calculated as angular averages for carriers at an energy $3/2k_B T = 39$ meV from the band edge.

is limited by acoustic scattering, while 52% of the hole mobility is due to optical scattering. Since quadrupoles mostly affect the ZO$_1$ optical mode, this clarifies why the hole mobility is affected more than the electron mobility.

For the room-temperature electron mobility, experimental values range from 23 to 217 cm$^2$/Vs [48–50, 53–55], and typically rely on the use of high-permittivity gate dielectrics. The goal of such dielectric is to suppress Coulomb scattering by immersing the MoS$_2$ monolayer in a high-$\kappa$ dielectric environment to boost mobility. There is a large spread in the reported theoretical literature, with room-temperature electron mobilities in the range 97-410 cm$^2$/Vs [34, 56–64]. In the case of the hole mobilities instead, the only previous calculation did not consider SOC, yielding a room-temperature value of 26 cm$^2$/Vs [63], underestimating by a factor of three the 76 cm$^2$/Vs experimental value [51], and by a factor of four the present result.

We note that the decrease in hole mobility occurring for temperatures above 200 K in Fig. 2(c) is due to the $\Gamma$ valley being only 52 meV below the valence-band maximum and therefore becoming thermally populated at higher temperatures. In addition to the SOC, and as seen in Fig. 2, the role of dynamical quadrupoles is crucial and delivers predicted drift and Hall mobility close to experimental values, with also the exponent of the power-law temperature decrease in the electron mobility going from $-1.34$ to $-1.08$, closer to measurements.

In conclusion, we have developed and implemented a conceptual and numerical framework to accurately calculate electron-phonon couplings in 2D materials on ultra-dense momentum grids, including full 2D electrostatics, quadrupoles, and SOC, highlighting how those are critical for the electron and hole mobilities of 2D materials like MoS$_2$. Given the major role played by 2D semiconductors in the post-silicon roadmap [65], we believe that the understanding of the physical effects described here will be of great value in the characterization and engineering of these promising and challenging materials. From the point of view of the theory, our work opens interesting new avenues for further investigation. For example, the emergence of the Berry connection in Eq. (3) suggests a previously unsuspected breakdown of (Wannier) gauge covariance in the long-range part of the matrix elements. Further work will be necessary to clarify the formal and practical implications of this result.

S.P. acknowledges support from the F.R.S.-FNRS; N.M. acknowledges support from the Swiss National Science Foundation and the NCCR MARVEL; M.G. acknowledges support from the Italian Ministry for University and Research through the Levi-Montalcini program; M.S. and M.R. acknowledge support from Minis-
terio de Ciencia y Innovación (MICINN-Spain) through Grant No. PID2019-108573GB-C22; from Severo Ochoa FUNFUTURE center of excellence (CEX2019-000917-S); from Generalitat de Catalunya (Grant No. 2017 SGR1506); and from the European Research Council (ERC) under the European Union’s Horizon 2020 research and innovation program (Grant Agreement No. 724529). Computational resources have been provided by the PRACE-21 resources MareNostrum at BSC-CNS, the Supercomputing Center of Galicia (CESGA), and by the Consortium des Équipements de Calcul Intensif (CÉCI), funded by the Fonds de la Recherche Scientifique de Belgique (F.R.S.-FNRS) under Grant No. 2.5020.11 and by computational resources awarded on the Belgian share of the EuroHPC LUMI supercomputer.
L. Baguet, J.-M. Beuken, J. Bieder, F. Bottin, J. Bouchet, E. Bousquet, et al., Comput. Phys. Commun. 248, 107042 (2020).
[48] Z. Yu, Z.-Y. Ong, Y. Pan, Y. Cui, R. Xin, Y. Shi, B. Wang, Y. Wu, T. Chen, Y.-W. Zhang, et al., Advanced Materials 28, 547 (2016).
[49] X. Cui, G.-H. Lee, Y. D. Kim, G. Arefe, P. Y. Huang, C.-H. Lee, D. A. Chenet, X. Zhang, L. Wang, F. Ye, et al., Nature Nanotechnology 10, 534 (2015).
[50] Y. Liu, H. Wu, H.-C. Cheng, S. Yang, E. Zhu, Q. He, M. Ding, J. Li, D. Guo, W. N. O., Y. Huang, et al., Nano Lett. 15, 3030 (2015).
[51] T. Momose, A. Nakamura, M. Daniel, and M. Shiomura, AIP Advances 8, 025009 (2018).
[52] S. Zollner, S. Gopalan, and M. Cardona, Journal of Applied Physics 68, 1682 (1990).
[53] Z. Yu, Y. Pan, Y. Shen, Z. Wang, Z. Y. Ong, T. Xu, R. Xin, L. Pan, B. Wang, L. Sun, et al., Nature Communications 5, 5290 (2014).
[54] A. Sanne, R. Ghosh, A. Rai, H. C. P. Movva, A. Sharma, R. Rao, L. Mathew, and S. K. Banerjee, Applied Physics Letters 106, 062101 (2015).
[55] K. Kang, S. Xie, L. Huang, Y. Han, P. Y. Huang, K. F. Mak, C. J. Kim, D. Muller, and J. Park, Nature 520, 656 (2015).
[56] K. Kaasbjerg, K. S. Thygesen, and K. W. Jacobsen, Phys. Rev. B 85, 115317 (2012).
[57] X. Li, J. T. Mullen, Z. Jin, K. M. Borysenko, M. Buongiorno Nardelli, and K. W. Kim, Phys. Rev. B 87, 115418 (2013).
[58] K. Kaasbjerg, K. S. Thygesen, and A.-P. Jauho, Phys. Rev. B 87, 235312 (2013).
[59] W. Zhang, Z. Huang, W. Zhang, and Y. Li, Nano Research 7, 1731 (2014).
[60] W. Li, Phys. Rev. B 92, 075405 (2015).
[61] T. Gunst, T. Markussen, K. Stokbro, and M. Brandbyge, Phys. Rev. B 93, 035414 (2016).

[62] T. Sohier, D. Campi, N. Marzari, and M. Gibertini, Phys. Rev. Materials 2, 114010 (2018).
[63] F. Guo, Z. Liu, M. Zhu, and Y. Zheng, Phys. Chem. Chem. Phys. 21, 22879 (2019).
[64] G. Gaddemane, S. Gopalan, M. V. de Put, and M. V. Fischetti, J. Comput. Elect. 20, 49 (2021).
[65] International roadmap for devices and systems (irds).
[66] J. P. Perdew, K. Burke, and M. Ernzerhof, Phys. Rev. Lett. 77, 3865 (1996).
[67] D. R. Hamann, Phys. Rev. B 88, 085117 (2013).
[68] M. van Setten, M. Giantomassi, E. Bousquet, M. Verstraete, D. Hamann, X. Gonze, and G.-M. Rignanese, Computer Physics Communications 226, 39 (2018).
[69] Here we follow the prescriptions of Ref. 35, based on the analysis of the real-space interatomic force constants; for MoS$_2$, we find that the optimal range separation parameter is $L = 10.5$ Bohr.
[70] The macroscopic in-plane polarizability can be obtained from the in-plane dielectric constant $\varepsilon_{\alpha\beta}$ of an artificially periodic stack of monolayers with spacing $c$ through $\alpha^{\parallel}(q) = (c/4\pi) \sum_{\alpha\beta} q_\alpha (\varepsilon_{\alpha\beta} - \delta_{\alpha\beta}) q_\beta$.
[71] We use fully relativistic norm-conserving Perdew-Burke-Ernzerhof (PBE) [66] pseudopotentials generated using the ONCVPSP code [67] and optimized via the PSEUDOJO initiative [68] which includes the 4$s^2$, 4$p^6$, 4$d^5$, 5$s^1$ as valence states for Mo and 3$s^2$, 3$p^4$ as valence states for S and was generated fully relativistically allowing for the inclusion of spin-orbit coupling in the calculation. The electron wave functions are expanded in a plane-wave basis set with kinetic energy cutoff of 140 Ry, and the Brillouin zone is sampled using a homogeneous $\Gamma$-centred $18 \times 18 \times 1$ mesh. We perform response calculations using density functional perturbation theory (DFPT) [17, 18] including a 2D Coulomb truncation [32] on a $18 \times 18 \times 1$ electron and phonon grids to ensure good convergence of the dielectric properties.