Fröhlich electron-phonon interaction and LO-TO splitting in doped semiconductors

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We develop a method to compute the electron-phonon interaction and the phonon frequencies in doped 3-dimensional semiconductors. The present formalism goes beyond the state-of-the-art description of the Fröhlich-like vertex and of the long-range non-analytical part of the dynamical matrix, taking into account the change of the inverse dielectric function $\epsilon^{-1}(q)$ in presence of free carriers at finite temperature. The method is computationally fast thanks to the use of interpolation based on maximally localized Wannier functions. We apply our approach to cubic silicon carbide 3C-SiC, where we find that in presence of doping the Fröhlich coupling and the LO-TO splitting are strongly reduced, thereby influencing observable properties such as the electronic lifetime.

The electron-phonon interaction (EPI) plays a fundamental role in a variety of physical properties, thus making it one of the most thoroughly studied topics in solid state physics [1–5]. The accurate theoretical calculation of the EPI is of crucial importance in order to predict and interpret, for example, transport experiments [6–11], excited carriers relaxation [12–14] and superconductivity [3, 15]. Such a precise evaluation of the EPI is of crucial importance in order to make it one of the most thoroughly studied topics in solid state physics [1–5].

The electron-phonon interaction (EPI) plays a fundamental role in a variety of physical properties, thus making it one of the most thoroughly studied topics in solid state physics [1–5]. The accurate theoretical calculation of the EPI is of crucial importance in order to predict and interpret, for example, transport experiments [6–11], excited carriers relaxation [12–14] and superconductivity [3, 15]. Such a precise evaluation of the EPI has become possible with the development of the theoretical frameworks of ab-initio density functional theory (DFT) [16, 17] and density functional perturbation theory (DFPT) [18], in conjunction with technical advances in the field of Wannier interpolation technique [19–21] and the progress of ab-initio computational infrastructures [22–24].

Despite the abundance of recent advances, the realistic description of the EPI on the full Brillouin zone (BZ) for three-dimensional doped semiconductors has been hindered by the lack of a well-defined and fast method to compute the interaction between electrons and phonons on fine reciprocal space grids. In particular, the long-range non-analytical part of the EPI (Fröhlich-like term), which, as it will be shown here, is the most sensitive to doping, is commonly evaluated from calculations performed in the undoped setup [25–28] relying on the microscopic model firstly developed by Vogl [29]. For doped polar materials, this procedure may lead to a very large understimation of the dielectric screening and a proportional overestimation of the Fröhlich-like interaction and, hence, to an incorrect evaluation of the electronic lifetimes. The same approach is usually adopted also to compute, in 3-dimensional doped semiconductors, the LO-TO splitting of optical modes originating from the non-analytical long-range components of the dynamical matrix [30]. In general, this type of calculations does not take into account the large reduction of the splitting that is induced by doping; this neglect in turn affects the EPI evaluation, since the phonon displacement amplitude for LO modes is underestimated. Moreover, if the LO-TO splitting is large enough in the undoped setup, the lowering of the LO frequency may contribute to an earlier activation, in temperature, of the Fröhlich scattering mechanism.

In this work we overcome the above described limitations and propose a precise, stable and fast technique based on first principles calculations and Wannier interpolation in order to obtain the correct phonon frequencies and EPI on fine grids in the BZ for the case of doped 3-dimensional semiconductors. We also show that the evaluation of the intraband contribution to the inverse dielectric screening via a random-phase approach (RPA) may lead to a qualitatively correct estimate of the long-range part of the dynamical matrix and EPI in the doped case, even though the degree of accuracy of such approximation is of course material dependent. Since only the intraband contributions depend relevantly on doping, while the interband terms may be evaluated in the undoped setup, it follows that when RPA is a good approximation the correct description of phonon frequencies and EPI requires first-principle calculations performed only in the undoped setup. Importantly, the technologies developed in this work can be used also to shed light on the interpretation of EELS cross-sections [31] and other scattering experiments [32].

The starting point of our methodology is based on the splitting of the dynamical matrix and the EPI as the sum of an analytical, short-range component (ASRC or A) and a non-analytical, long-range one (NALRC or NA) following the seminal works of Pick, Cohen and Martin [32], Vogl [29] and Stengel [34]. Here it is shown that the NALRC of the dynamical matrix $C$ and of the EPI $g$ can be expressed as

$$C^\text{NA}_{s,s',\alpha,\beta}(q) = \frac{4\pi e^2}{V} Z^*_s,\alpha(q) \tilde{Z}_{s',\beta}(q), \quad (1)$$

$$g^\text{NA}_{\nu,\nu',\alpha,\beta}(q, k) = i \frac{4\pi e^2}{Vq} \left( \sum_{s,s'} Z_{s,\alpha}(q) c^{\nu,\alpha}_{s,s'}(q)^* \left( \frac{M_0}{M_s} \right)^{1/2} \times \langle u_{m,k+q} | u_{m',k} \rangle \right)^2, \quad (2)$$

where $\nu, \nu' \in \{\sigma, \pi\}$ are the electronic branches, $\alpha, \beta = \{\sigma, \pi\}$ label the spin states, $s, s' \in \{1, 2, \ldots, N\}$ denote the $N$ inequivalent crystallographic sublattices, $V$ is the unit cell volume, $q \in \Gamma$ is the reciprocal lattice vector, $M_0$ is the mass of the free electron, and $u_{m,k}$ is the atomic orbital of $m$th atom at $k$th point of the BZ.
where $V$ is the volume of the unit cell, $s, s'$ are atomic indexes, $\alpha, \beta$ are Cartesian indexes, $\nu$ is the phonon branch index, $m', m$ are electronic band indexes, $k$ and $q$ are the electronic and phonon wavevectors respectively, $\mathbf{e}_s^\nu(q)$ is the phonon eigenvector connected to the displacement of the atom $s$ belonging to the cell $\mathbf{R}_p$ in the position $\mathbf{r}_s$ via $\mathbf{u}_s^\nu(q) = \mathbf{e}_s^\nu(q)e^{iq\mathbf{R}_p+\mathbf{r}_s}$, $\nu_q = [\hbar/(2M_0\omega_q)]^{1/2}$ is the mean phonon displacement amplitude of the phonon $\omega_q$. $M_0$ is an arbitrary reference mass, $M_s$ is the mass of the atom $s$ and $u$ are the periodic part of the Bloch functions; $Z_{s,\alpha}(q)$ are the screened effective charges defined as \[ Z_{s,\alpha}(q) = -\frac{V}{e} \delta_{s,\alpha}^\text{ind}(q) + \frac{q_0}{q} Z_s, \] where $Z_s$ is the ionic charge and $\delta_{s,\alpha}^\text{ind}(q)$ is the induced charge density produced by a collective displacement of wavevector $q$ of the atoms of kind $s$ along the Cartesian direction $\alpha$, which we denote with $u_{s,\alpha}(q)$, while the absence of the phase factor $e^{iq\mathbf{r}_s}$ with respect to Eq. 2 of Ref. \[ \text{[26]} \] comes from the different definition of the phonon eigenvectors; the second term of Eq. \[ \text{[3]} \] is the trivial charge density change due to the bare collective atomic displacement. Lastly, we have defined the unscreened effective charges $Z_{s,\alpha}(q)$ as $Z_{s,\alpha}(q) = e^{-\mathbf{q}}(q)Z_{s,\alpha}(q)$ where $e^{-\mathbf{q}}(q)$ is the head of the inverse of the dielectric matrix; $Z_{s,\alpha}(q)$ can be obtained by means of DFPT by putting to zero the change of the Kohn-Sham potential at $\mathbf{G} = 0$ (we denote a generic reciprocal lattice vector with $\mathbf{G}$) during the self-consistent solution of the Sternheimer equation in presence of the external perturbation connected to $u_{s,\alpha}(q)$ \[ \text{[27]} \text{[32]}. \] It can be shown \[ \text{[35]} \] that $Z_{s,\alpha}(q)$ is analytical and enjoys a Taylor expansion of the form

\[ Z_{s,\alpha}(q) = \frac{q_0}{q} Z_{s,\alpha}^* + \frac{i}{2} \frac{q_0}{q} q_1 Q_{s,\alpha\beta} + \ldots \] (4)

where the various terms of the expansion, which we will refer to as effective charge tensors, can be identified with the Born effective charges, dynamical effective quadrupoles and so on—sums over repeated indexes are intended. For an undoped 3-dimensional semiconductor, the non-analyticity of Eqs. \[ \text{1} \text{ and } \text{2} \] stems from the long wavelength limit $\lim_{q \to 0} e^{-\mathbf{q}}(q) = q^2/(\mathbf{q}, \mathbf{q}^2)\mathbf{q}$, where $\mathbf{q}^2\infty$ is the high-frequency static macroscopic dielectric tensor \[ \text{[36]}. \] In this case, the form of Eqs. \[ \text{1} \text{ and } \text{2} \] would apparently invalidate any attempt of interpolation of the dynamical matrix or of the EPI near the BZ center. To prevent this, for a general $(\mathbf{k}, \mathbf{q})$ one first interpolates the differences $C^A = C - C^{\text{NA}}$ and $g^A = g - g^{\text{NA}}$ via Fourier or Wannier interpolation \[ \text{[29]}; \] since usually the NALRCs are explicitly known only for $q \to 0$, their expression is extended on the whole BZ applying Ewald summation techniques on proper real space expressions that lead to the correct limiting values \[ \text{[25, 37]} \]. After this step, the NALRCs of Eqs. \[ \text{1} \text{ and } \text{2} \] are separately evaluated at $(\mathbf{k}, \mathbf{q})$ truncating Eq. \[ \text{1} \] at a certain order (consistently with what done in the previous steps—commonly the first, apart from refined recent advances where also the second order is considered, see Refs. \[ \text{[8, 27, 33]}; \] finally, the ASRCs and the NALRCs are added back together at $(\mathbf{k}, \mathbf{q})$ to obtain the correctly interpolated quantities \[ \text{[39]} \].

Although the above procedure is usually applied to the undoped setup, Eqs. \[ \text{1} \text{ and } \text{2} \] and the argument for the analyticity of $Z_{s,\alpha}(q)$ are still formally valid even in presence of a carrier concentration $n$ at finite temperature $T$, with the difference that the effective charge tensors and the screening now formally depend on $n$ and $T$. With this dependence, Eqs. \[ \text{1} \text{ and } \text{2} \] become analytical around $\Gamma$ due to the regularization of the inverse dielectric matrix for small $q$, induced by the presence of free carriers \[ \text{[11]} \]. Nonetheless in the limit of small doping, which is the relevant one when studying, e.g., the intrinsic transport coefficients, the region of regularization in reciprocal space is vanishingly small and does not practically guarantee that the dynamical matrix and the EPI can be interpolated without any precaution. As a consequence, if we want to interpolate the phonon frequencies and the EPI on a generic $(\mathbf{k}, \mathbf{q})$ point we still have to resort to techniques that rely on the subtraction and addition of the asymptotic formula for the NALRCs, which in principle have to be calculated for each $n$ and $T$. In practice, one can compute the EPI and dynamical matrices from \textit{ab-initio} and extract the ASRCs on a coarse grid of points exactly as for the undoped case at $0K$, interpolate the ASRCs in $(\mathbf{k}, \mathbf{q})$, and then add the NALRCs of Eqs. \[ \text{1} \text{ and } \text{2} \] on the same point but now including the correct $n$ and $T$ dependences. One main simplification to

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FIG. 1. Real part of the screened effective charge $Z_{s,\alpha}(q)$ as a function of $q$ along the line $q = (q/\sqrt{2}, q/\sqrt{2}, 0)$ (displayed in the BZ as a red line) for both the atoms in 3C-SiC in the undoped case (filled cicles) and in the doped case with a carrier concentration of $n = 4.827 \times 10^5$ cm$^{-3}$ (filled squares). The continuous lines are drawn using Eq. \[ \text{7} \] in the undoped case, while the dashed ones are drawn using Eq. \[ \text{4} \] for the doped case.
The practical strength of this method is that if we are interested in interpolating the phonon frequencies and the EPI on a fine grid (and not only in a point) we do not need to compute the inverse dielectric tensor for all the points of the grid, but we actually need to compute the leading order of $\epsilon^{-1}(\mathbf{q}, n, T)$ only on a number of lines which is equivalent to the number of independent components of the $\mathbf{\epsilon}^{\infty}$ tensor, for symmetry reasons [35]. Also, the computation on each line can be performed on a coarse set of points, while the evaluation on a fine mesh can be obtained by a simple polynomial interpolation, without any phase problem since the dielectric function is purely real in the static case.

In order to validate the above described theoretical approach, we numerically study 3C-SiC—one of the promising wide-gap semiconductors for power electronics [42]—as a prototypical example of polar material where phonon frequencies and EPI are heavily influenced by doping.

To perform the first-principles calculations we use a private version of the QUANTUM ESPRESSO code [22], developed in Ref. [31] to compute the charge density entering the electron energy loss spectroscopy (EELS) cross section. We use PBE-GGA functionals [43] and norm-conserving pseudopotentials, and we sample the BZ with telescopic grids that in the densest region are equivalent to Monkhorst-Pack grids of dimension $729^3$ in order to well describe the region around the Fermi level [35]. We use an hole doping concentration of $10^{-7}$ holes per cell, corresponding to a carrier density of $n = 4.827 \times 10^{15} \text{cm}^{-3}$—which is roughly the lower limit of carrier concentrations for p-type 3C-SiC transport experiments [44]—by reducing the total number of electrons in the system while introducing a negative uniform jellium background to preserve the neutrality of the system. The Wannier interpolation is performed using a private version of EPW [24], adapted for the doped case, using meshes of $16^3$ for the electrons and $8^3$ for the phonons. More information are presented in the Supplementary Information.

We start by studying the macroscopic screened effective charge $Z_{s,\alpha}(\mathbf{q})$ for the undoped and the doped setups at zero temperature, simulated in the latter case with a Gaussian smearing of $10^{-5}$ Ry. We present in Fig. 1 the ab-initio calculation of $\text{Re}Z_{s,y}(\mathbf{q})$ along a specific line in reciprocal space; the particular choice of the line is dictated by symmetry considerations on the effective charge tensors for the cubic case [35], even though the procedure may be generalized to any symmetry. We compare it with the leading order expansions

$$Z_{s,y}(\mathbf{q}) = Z_s^* / \sqrt{2} / \epsilon^\infty,$$

$$Z_{s,y}(\mathbf{q}, n, T) = \epsilon^{-1}(\mathbf{q}, n, T) Z_s^* / \sqrt{2}$$

where $Z_{s,\alpha}\delta_{\alpha\beta} = Z_s^* \delta_{\alpha\beta}$ and $\mathbf{\epsilon}^{\infty}\delta_{\alpha\beta}$ are computed with DFPT at $\mathbf{q} = \mathbf{0}$ in the undoped setup, while $\epsilon^{-1}(\mathbf{q}, n, T)$ is computed using DFPT for each $\mathbf{q}$ point in the doped case. We notice that the agreement between

FIG. 2. Upper) comparison between the phonon frequencies obtained from first-principles in the undoped and doped setup (circles and squares), and their Fourier interpolation obtained as explained in the text (continuous and dashed lines). The TO mode for the doped case numerically coincides, on this scale, with the undoped one. Lower) same comparison for the EPI, with interpolation obtained by Fourier and Wannier transforms. The $\mathbf{k}$-point is fixed at the $\Gamma$ value, while the indexes of the coupling correspond to the highest valence band (both for in and out states) and the highest optical branch (LO).
calculations and Eqs. \[3\] and \[4\] is excellent at small $q$ with small differences arising at larger wavevectors that are ascribable to higher order terms in $Z_{s,y}(q)$. Notably, in Eq. \[4\] we have used the Born effective charge tensor computed in the undoped setup, signalling that the effect of doping enters predominantly in the head of the inverse dielectric screening while weakly affecting the wings at the leading order, as already anticipated.

The computation of the second order expansion of $Z_{s,a}(q)$, related to the dynamical effective quadrupole tensor, can be performed similarly on the same line but looking at the expansion of the imaginary part of $\bar{\chi}(0)$ at $0 \text{K}$; red and blue arrows correspond to the intraband and interband contributions that are correctly included in the calculation, while the violet arrow represents the contributions that shouldn’t be present but are included in $\chi^0$.

It is evident that doping effects are very relevant for the asymptotic behaviour of both the phonon frequencies and the EPI in the $q \to 0$ limit. In the first case, the LO frequency is strongly softened and the LO-TO splitting is substantially suppressed; in the second case, the Fröhlich coupling is strongly reduced.

A diriment question regarding the methodology here presented is whether we can obtain the macroscopic dielectric function $\epsilon^{-1}(q,n,T)$ without resorting to first-principles calculations, while preserving the qualitative trends presented in Fig. 2. This would provide a fast method to evaluate the dielectric screening for a whole range of carrier concentrations and temperatures without resorting to first principles calculations. To reach our goal, we use the RPA relation $\epsilon(q,n,T) = 1 - 4\pi e^2/q^2\chi^0(q,n,T)$, and $\epsilon^{-1}(q,n,T) \approx 1/\epsilon(q,n,T)$—the latter relation is an approximation implying that local fields are neglected (LFs \[35\]), but this is not stringent since LFs corrections typically give corrections of order 10% on the value of $\epsilon^{-1}$ \([37,39]\). For the evaluation of $\chi^0(q,n,T)$ the strategy used here, and similar to what done in Ref. \[50\], is to split the independent particle polarizability into two contributions, as follows:

$$\chi^0(q,n,T) \approx \chi^0_{\text{undop.}}(q) + \delta\chi^0(q,n,T),$$

$$\delta\chi^0(q,n,T) = \frac{2}{V} \sum_{m,n,T} \frac{\delta f_{mk} - \delta f_{m'k'+q}}{\epsilon_{mk} - \epsilon_{m'k'+q}} |\langle u_{mk}|u_{m'k+q}\rangle|^2,$$

where $\delta f = f_{\text{dop.}} - f_{\text{undop.}}$, $f$ being the electronic statistical distribution. We have supposed that we are in a regime where DFT energies and overlaps between the periodic parts of the Bloch functions can be approximated with the ones of the undoped setup and that $\chi^0_{\text{undop.}}(q,T) \approx \chi^0_{\text{undop.}}(q)$; we also further neglect the change of the interband contributions between the doped and undoped setups (see the inset of Fig. 3 for a schematic representation of the approximation). Within these approximations, we compute $\chi^0_{\text{undop.}}(q)$ just once directly \textit{ab-initio}, while for each doping and temperature values we evaluate $\delta\chi^0(q,n,T)$ using Wannier interpolation, that can be operatively restricted to the bands near the Fermi level. A further practical simplification comes from neglecting the $q$ dependence of the undoped polarizability using $1 - 4\pi e^2/\chi^0_{\text{undop.}}(q) \approx \epsilon^{-1}$.

The result of this procedure is compared in Fig. 3 with the first-principles $\epsilon(q,n,T)$. Despite the number of approximations involved, we notice that the agreement is qualitatively good, and it comes along with a great speed-up in computational cost since Eq. \[4\] can be evaluated very quickly on a large set of points. In passing by, we also notice from Fig. 3 that the \textit{ab-initio} head of the inverse of dielectric matrix is prone to polynomial interpolation so that, as already anticipated, when simple modelling is not accurate enough, calculations may be performed \textit{ab-initio} on a relatively coarse set of points and then interpolated inexpensively on fine meshes.

![FIG. 3. Comparison of the head of the inverse dielectric matrix computed from first principles in the undoped setup (black filled circles) and in the doped case (red filled circles) and compared with the model that uses Eq. 7 (ochre filled circles); dashed lines are drawn for the doped cases as a guide to the eye. Also, $\epsilon^\infty$ is drawn as reference. Inset: pictorial sketch of the approximation for $\chi^0$ at 0 K; red and blue arrows correspond to the intraband and interband contributions that are correctly included in the calculation, while the violet arrow represents the contributions that shouldn’t be present but are included in $\chi^0$.](image-url)
Finally, to quantify the impact that the correct descriptions of the EPI and phonon frequencies have on physical observables, we evaluate the electronic inverse lifetimes in the Self Energy Relaxation Time Approximation (SERTA) as

\[
\tau_{mk}^{-1} = \frac{2\pi}{\hbar} \sum_{m'\nu} \int \frac{d\mathbf{q}}{V_{BZ}} g_{\nu,m'\nu}(\mathbf{k}, \mathbf{q})^2 \times \\
\left[ (1 - f_{m'k+\mathbf{q}} + n_{\nu\mathbf{q}}) \delta(\epsilon_{mk} - \epsilon_{m'k+\mathbf{q}} - \hbar\omega_{\nu\mathbf{q}}) + \\
(f_{m'k+\mathbf{q}} + n_{\nu\mathbf{q}}) \delta(\epsilon_{mk} - \epsilon_{m'k+\mathbf{q}} + \hbar\omega_{\nu\mathbf{q}}) \right],
\]

where \(V_{BZ}\) is the volume of the BZ, and \(n_{\nu\mathbf{q}}\) is the Bose-Einstein occupation factor of the phonon of frequency \(\omega_{\nu\mathbf{q}}\). We compare the cases where the NLARCs are interpolated 1) using Eqs. (1, 2) with the undoped dielectric screening (Eq. [5]), and 2) using Eqs. (1, 2) with the correct screening dependence on \(n\) and \(T\) (Eq. [6]); in the second case, the dielectric function is evaluated at the RPA level. Since the doping-temperature regimes where significant changes of screening properties occur depend critically on the material electronic properties, to perform an exemplary comparison we choose a setup tailored to highlight the differences between the two approaches. For sake of clarity, we fix the Fermi level at the top of valence band and set \(T = 300\) K, so that the numerical difference in the approaches can be ascribed only to differences in the screening; the carrier concentration of this setup corresponds approximately to \(10^{20}\) cm\(^{-3}\). As shown in Fig. 4, discarding the doping dependence of the screening, as currently done in most state-of-the-art first-principles calculations within the rigid-band approximation, implies an overestimation of inverse scattering times up to a factor \(\sim 3\) as soon as the onset of optical phonon scattering is reached (\(w_{LO}\) in figure); on the other hand, an inaccurate description of screening properties leads to underestimate \(\tau^{-1}\) for energies closer to the Fermi level, as a consequence of the aforementioned suppression with doping of the LO frequency, which implies different scattering selection rules for the Fröhlich coupling.

In conclusion, we have proposed a fast and accurate method based on Fourier and Wannier interpolation that is able to correctly reproduce the EPI and phonon frequencies in doped 3-dimensional semiconductors on very fine reciprocal space grids. We have also shown that an approximated approach based on the separation of the independent particle polarizability between intraband and interband contributions may yield, in dependence of the material, good qualitative results, with the benefits of an important reduction of computational time. The comparison with state-of-the-art methodologies shows that these incorrectly estimate the scattering rates due to the Fröhlich interaction in specific regimes, and this may lead to inaccurate conclusions on transport properties in proper doping-temperature regimes. Lastly, we mention that the extension to finite frequencies of our approach for computing the screened charge change may be of particular importance for the interpretation of EELS and other inelastic scattering experiments probing vibrational properties.
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[39] Strictly speaking, the correct procedure would consist in readding the NALRCs using again Ewald summation techniques sending $\epsilon^\infty \rightarrow 1/\epsilon^{-1}(q, n, T)$, which has the correct limiting values and is consistent with the previous steps on the points of the BZ far away from $\Gamma$. Practically, we find that for the region of points studied in this work the difference between the procedures is negligible.

[40] From a qualitative point of view, this can be explained treating the doping within a Thomas-Fermi approach, with $\epsilon(q) \approx \epsilon^\infty + kF_T^2(T)/q^2$. One can then replace $q^2/\epsilon^\infty \cdot q \rightarrow q^2/(kF_T^2(T) + q \cdot \epsilon^\infty \cdot q)$ in the formula for the NALRCs, thus expressing them in a manifestly analytic form. This argument is purely qualitative since the Thomas-Fermi model is not able to describe efficiently the screening in the regimes studied in this work.

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