Electron–phonon interaction without overscreening: a strategy for first–principles modelling

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State-of-the-art approaches to calculate the electron-phonon and the phonon-electron self-energy are based on a mean-field approximation for the interacting electronic system. This approach introduces an overscreening error which results in an underestimation of the electron-phonon coupling strength. We introduce a theoretical and numerical approach for the calculation of the phonon-electron self-energy without the overscreening error. Starting from the out-of-equilibrium Kadanoff-Baym equations for the phonon propagator, we discuss and compare the overscreened (i.e., symmetrically screened) and overscreening–free (i.e., asymmetrically screened) cases. We point out that the difficulty in treating the latter stems from the static approximation to the dielectric function and from the need to obtain a self-energy that preserves the elementary scattering processes. We solve both problems in the equilibrium case by considering a manifestly symmetric form of the correct self-energy which can be easily calculated numerically and yields an overscreening–free coupling strength. Finally, we describe the numerical implementation of this treatment into the first–principles Yambo code for the calculations of phonon linewidths.

I. INTRODUCTION

Electron–phonon interaction is one of the widest topics in all of condensed matter theory because the complex interplay between lattice and electronic motions is responsible for many important crystal properties and physical mechanisms. Among such mechanisms, the finite lifetime of lattice vibrational modes induced by electron-phonon interaction strongly characterises phononic spectral properties and is the foundation of many relevant phenomena including superconductivity coupled ultrafast dynamics and lattice instabilities.

The standard approach to model how phonon-related properties are affected by the interaction with electrons involves perturbation theory, typically at first order in the electron-phonon interaction. It is important to point out that in this approach the electronic perturbation must also incorporate electron–electron interactions in order to yield physically relevant phonon frequencies and spectra. These interactions are generally approximated at a mean–field level by both first–principles – i.e., density functional perturbation theory (DFPT) and lattice instabilities – and model Hamiltonians. No problems arise from this approximation if the goal is to describe how the interacting electron system is perturbed by the lattice vibrations. However, this approach introduces a systematic error in the aforementioned, specular case of the phonon system perturbed by the interacting electrons. The error is caused by the double counting of the electronic screening in the electron-phonon coupling strengths and has been studied in the context of adiabatic phonon frequency corrections, what is lacking is both an assessment of the magnitude of such overscreening error (OE) and a general theoretical scheme able to produce overscreening error–free (OE–free) coupling strengths amenable for calculations of phonon linewidths and lifetimes in realistic materials.

We have addressed the former topic in our companion paper showing giant corrections in the computed phonon linewidths of superconducting MgB2 by using the OE–free approach. In this manuscript, we address instead the latter issue by providing a complete theoretical framework making use of many–body perturbation theory, followed by the numerical implementation of the results in the first–principles code Yambo. We start from a very general out-of-equilibrium treatment involving the Kadanoff-Baym equations of motion for the phonon propagator and emphasise the correct way to obtain an OE–free phonon-electron self-energy. The generality of the approach is important, because it is not tied to a static screening approximation and thus may constitute the starting point for the wide application of an OE–free scheme in various physical scenarios: from out-of-equilibrium lattice dynamics and relaxation rates, to nonadiabatic phonon spectra. At equilibrium and in the static screening approximation, this approach reduces to an easily implementable expression for the OE–free electron-phonon coupling strengths, which may be used to estimate, e.g., the critical temperatures and energy gaps of BCS superconductors following a computation of the relative phonon linewidths.

The paper is organised as follows. In Sec. I we derive and discuss the phonon-electron self-energy in the OE and OE–free cases after introducing the equations of motion for the phonon propagator. In Sec. II we address the equilibrium case and discuss the OE–free electron-phonon coupling strength. We also obtain the expressions for the phonon linewidths starting from the self-energy. In Sec. III we describe the new code implementation we developed in this work using the Quantum Espresso and Yambo codes, and we also provide a scheme.
of the calculation flow.

II. PHONON-ELECTRON SELF-ENERGY

In this Section we derive and compare the phonon-electron self-energy in the overscreened (OE) and OEl-free cases. The notation and Eqs. from (1) to (10) are adapted from Marini & Pavlyuk\cite{12} which will be our starting point. In order to follow this Section with the free cases. The notation and Eqs. from (1) to (10) are steps of Ref.\cite{12}. We write the Hamiltonian of the electron-phonon (e–ph) coupling as

\[ H = H_e + H_{ph} + H_{e-ph}, \]

with

\[ H_e = \sum_i \epsilon_i \hat{a}_i^\dagger \hat{a}_i, \]

\[ H_{ph} = \frac{1}{2} \sum_\nu \Omega_\nu (\hat{P}_\nu^2 + \hat{Q}_\nu^2), \]

\[ H_{e-ph} = \sum_{ij} g_{\nu ij} \hat{a}_i^\dagger \hat{a}_i \hat{Q}_\nu. \]

Here, \( \hat{a}_i \) and \( \hat{a}_i^\dagger \) are the fermionic destruction and creation operators of an electronic state with generalised index \( i \) and energy \( \epsilon_i \). The bosonic coordinates \( (\hat{Q}_\nu) \) and momenta \( (\hat{P}_\nu) \) for a harmonic lattice vibration (phonon) of generalised index \( \nu \) and energy \( \Omega_\nu \) can be expressed in terms of bosonic destruction and creation operators \( \hat{b}_\nu \) and \( \hat{b}_\nu^\dagger \) as \( \hat{Q}_\nu = 1/\sqrt{2} (\hat{b}_\nu^\dagger + \hat{b}_\nu) \) and \( \hat{P}_\nu = i/\sqrt{2} (\hat{b}_\nu^\dagger - \hat{b}_\nu) \). Finally, \( g_{\nu ij} \) is the electron-phonon (e–ph) coupling matrix element.

Now we have to decide how to treat the electron-electron interaction. If a mean-field approach is chosen, then the energies \( \epsilon_i \) result from the solution of the mean-field problem, e.g., the Kohn-Sham eigenvalues from density functional theory. This implies that the phonon quantities \( \Omega_\nu \) and \( g_{\nu ij} \) are statically screened (this renormalisation is sometimes called adiabatic correction\cite{14}). For now, however, we do not specify if \( \Omega_\nu \), \( \epsilon_i \) and \( g_{\nu ij} \) are bare quantities or mean-field corrected, statically screened ones, since at this point it does not affect the derivation.

If we use the position-dependent electronic field operators, i.e., \( \hat{\psi}(x) = \sum_i \phi_i(x) \hat{a}_i \) with the \( \phi_i(x) \) being eigenfunctions of \( H_e \), we can rewrite the electron-phonon Hamiltonian as

\[ \hat{H}_{e-ph} = \sum_\nu \int dx \hat{\psi}(x) \partial_\nu |_{eq} W_{e-ph}(x) \hat{\psi}(x), \]

where \( W_{e-ph}(x) \) is the static electron-lattice interaction (screened or otherwise). The corresponding e–ph matrix element in the position basis is

\[ g_{\nu}(x) \equiv \partial_\nu |_{eq} W_{e-ph}(x). \]

At this point we can apply the standard procedure to obtain the Kadanoff-Baym equation of motions (EOM) for the Green’s function, in the phonon case. First we switch to the time-dependent Heisenberg operator representation with time arguments \( z \) on the Keldysh contour\cite{22} by employing the Keldysh formalism we ensure that our treatment is not limited to the equilibrium and/or zero-temperature cases. We now apply Heisenberg’s EOM for operators, \( \frac{d}{dt} \hat{O}(z) = i[\hat{O}(z), \hat{H}(z)] \), with \( \hat{O}(z) = \hat{Q}_\nu(z) \) and \( \hat{O}(z) = \hat{P}_\nu(z) \). Having then obtained the coupled EOMs for the bosonic operators, we combine them to arrive at the EOM for the phonon Green’s function \( D_{\nu\rho}(z_1, z_2) \). In fact, the definition of \( D_{\mu\nu}(z_1, z_2) \) is

\[ D_{\mu\nu}(z_1, z_2) = -2i \left\langle T \left( \hat{Q}_\mu(z_1) \hat{Q}_\nu(z_2) \right) \right\rangle, \]

where \( \langle \cdots \rangle \) is the trace over the exact density matrix, \( T \) the contour-ordering operator, and we made the additional assumption (which is true at equilibrium) that \( \langle \hat{Q}_\nu(z) \rangle = 0 \). For future reference, we also give the definition of the electron Green’s function \( G(x_1, z_1; x_2, z_2) \equiv G(1, 2): \)

\[ G(1, 2) = -i \left\langle T \left( \hat{\psi}(1) \hat{\psi}^\dagger (2) \right) \right\rangle. \]

The EOM for \( D_{\mu\nu}(z_1, z_2) \) obtained in this way can be closed if we express the electron-phonon interaction in terms of a self-energy \( \Pi_{\mu\nu}(z_1, z_2) \) which is then truncated at first order in the interaction. With such a self-energy, the second-order differential EOMs for \( D \) are:

\[
\left[ \frac{d^2}{dz_1^2} + \Omega_\mu \right] D_{\mu\nu}(z_1, z_2) = -2\Omega_\mu \delta_{\mu\nu} \delta(z_1 - z_2) - 2\Omega_\mu \sum_\alpha \int dz_3 \Pi_{\mu\alpha}^L(z_1, z_3) D_{\alpha\nu}(z_3, z_2),
\]
We refer to Eq. \((7)\) as the left EOM and to Eq. \((8)\), which is its adjoint, as the right EOM. As can be seen by the superscripts, the left and right equations depend on left \((L)\) and right \((R)\) self-energies. For a detailed examination on how to consistently derive and approximate left and right self-energies, see Ref.\(^{23}\) or Chap. 9 of Ref.\(^{22}\). Here it is sufficient to remark that \(\Pi^L = \Pi^R\) for the full self-energy, i.e., the one containing all orders of the interaction, but we explicitly need to ensure that this will be the case also for any truncated self-energy.

\[
\Pi_{\alpha\beta}(z_1, z_2) = \int d34 \int dx_1 g_\alpha(x_1)G(1,3)G(4,1)\Gamma_{\alpha\beta}^{e-ph}(3,4; z_2). \tag{9}
\]

This expression contains the electron-phonon three-points vertex function \(\Gamma_{\alpha\beta}^{e-ph}\), which in turn is given by the following self-consistent equation:

\[
\Gamma_{\alpha\beta}^{e-ph}(1,2; z_3) = \delta(1,3)\delta(1,2)g_\alpha(x_1) + \int d4567 \frac{\delta M(1,2)}{\delta G(4,5)}G(4,6)\Gamma_{\alpha\beta}^{e-ph}(6,7; z_3)G(7,5), \tag{10}
\]

where \(M(1,2)\) is the mass operator containing electronic correlations other than a mean-field potential (recall that at this point we did not yet specify whether the mean-field potential is actually present or not). The kernel \(\delta M/\delta G\) then has the form of an effective potential.

In summary, so far we have obtained the form of the many-body phonon-electron self-energy. The task now is to approximate it to a form useful for actual calculations, e.g., of the phonon linewidths. In particular, we have to consider how to perform first-order truncations of Eq. \((9)\) by taking into account the screening of the electron-phonon interaction \(H_{e-ph}\).

**C. Electronic screening**

The inverse electronic dielectric function \(\epsilon^{-1}\) is expressed as

\[
\epsilon^{-1}(1,2) = \delta(1,2) + \int d3 v(1,3)\chi(3,2). \tag{11}
\]

Here, \(v(1,3) = v(x_1, x_3)\delta(z_1, z_3)\) is the Coulomb interaction, while \(\chi(3,2)\) is a linear response function to a perturbing external electric field. The most common approximation for \(\chi\) is the Hartree or RPA approximation which includes the contributions of noninteracting electron-hole pair excitations (“bubbles”) at all orders.

If this condition is ensured, then the self-energy is said to be conserving and it guarantees conservation of the elementary scattering processes.

**B. Phonon-electron self-energy**

The last result we need from Ref.\(^{23}\) is the form of the full phonon-electron self-energy II, which is

Within the Hartree approximation, \(\chi(1,2)\) is then written as the following self-consistent equation,

\[
\chi(1,2) = P^0(1,2) + \int d34 P^0(1,3)v(3,4)\chi(4,2) \tag{12}
\]

where \(P^0(1,2)\) represents a polarization “bubble” and is thus

\[
P^0(1,2) = -iG^0(1,2)G^0(2,1). \tag{13}
\]

Here notice that \(G^0\) refers to the noninteracting electronic Green’s function, i.e. Eq. \((6)\) where the trace is evaluated on the noninteracting density matrix from \(H_e\). From Eqs. \((11)\) and \((12)\) it also follows that \(\epsilon\) is equal to

\[
\epsilon(1,2) = \delta(1,2) - \int d3 v(1,3)P^0(3,2). \tag{14}
\]

This expression will become useful in the following.

**D. Overscreened self-energy**

The bare electron-phonon matrix element is given by just the first-order variation of the Coulomb interaction \(V_{e-ph}\) between electrons and ions due to lattice displacements:

\[
g^B_\nu(x) = \partial_\nu |_{eq} V_{e-ph}(x). \tag{15}
\]
In a many-body formulation and with harmonic phonons, the electronically screened electron-phonon matrix elements are instead

$$g_S^S(1, z_2) = \int dx_2 \epsilon^{-1}(1, x_2 z_2) g^B(x_2). \tag{16}$$

If our starting Hamiltonian, Eq. (1), contains a mean-field potential, then the e–ph matrix elements included in $H_{e-ph}$ are statically screened, therefore we have an approximated mean-field screening function $\epsilon^{-1}(1, 2) \simeq \epsilon^{-1}(x_1, x_2)$ and thus

$$g_S^S(x) = \int dx_2 \epsilon^{-1}(1, x_2) g^B(x_2). \tag{17}$$

For example, the most common mean-field approximation is arguably the one used in DFT (i.e., static Hartree and exchange-correlation, $H_{xc}$). In this case Eq. (17) coincides with the DFPT case:

$$\int dx_2 \epsilon^{-1}_{H_{xc}}(x, x_2) g^B(x_2) = \delta_p |_{eq} V_{self}(x) \tag{18}$$

where $V_{self}$ is the self-consistent DFPT electron-ions potential.

In this screened case, we then have $g \rightarrow g^S$ in the self-energy expression Eq. (17). Then, its first-order truncation is obtained by letting $G \rightarrow G^0$ and taking only the zeroth order of the vertex function in Eq. (10), i.e.,

$$\Gamma^{e-ph}_\alpha(12; z_3) |_{0} = \delta(1, 3) \delta(1, 2) g^S_\alpha(1). \tag{19}$$

Therefore, we get an expression for the first-order $\Pi$ which is symmetrically screened (SS) on both sides:

$$\Pi^{SS}_\beta(z_1, z_2) = \int dx_1 x_2 g_\alpha^S(x_1) P^0(1, 2) g_\alpha^S(x_2). \tag{20}$$

notice that this also means that the $R$ and $L$ versions of the SS self-energy in the EOMs [7 and 8] coincide.

Although this self-energy might appear very reasonable, we immediately notice from its diagrammatic representation in Fig. (1(b) that it contains a double counting of the screening, as each order of the expansion of $\epsilon^{-1}$ in polarization bubbles – recall Eq. (12) – appears multiple times: this makes it clear that $\Pi^{SS}$ in Eq. (20) suffers from the overscreening error. Note that in this case the double counting is embedded into the static $g^S$ from the start, while also forcing a static approximation to the screening function despite being out of equilibrium.

E. Derivation of the OE-free self-energy

In order to avoid the double counting, it is necessary to treat the screening consistently on a many-body level. Therefore, we start with the bare e–ph coupling $g \rightarrow g^S$ and look for how the electronic screening enters the self-energy in Eq. (9). It does so via the vertex function $\Gamma^{e-ph}_\alpha$ in Eq. (10). In particular, we know that the RPA screening in Eq. (12) is obtained from the Hartree term $V_H$ in the electron-electron interaction,

$$V_H(1) = \int d2 \left< n_e(1) \right> \tag{21}$$

where $\left< n_e(1) \right> = -iG(1, 1^+) is the electronic particle density.

Therefore, we approximate the mass operator as just the Hartree term, $M(1, 2) \sim V_H(1) \delta(1, 2)$, so that the kernel in Eq. (10) becomes:

$$\frac{\delta M(1, 2)}{\delta G(4, 5)} \sim \delta(1, 2) \delta(4, 5) \frac{\delta V_H(1)}{\delta G(4, 4^+)} = -i \nu(1, 4). \tag{22}$$

Consequently, the vertex

$$\Gamma^{e-ph}_\alpha(12; z_3) = \delta(1, 2) \Gamma^{e-ph}_\alpha(1; z_3)$$

is itself reduced to a two-points function, and Eq. (10) takes the following form:

$$\Gamma^{e-ph}_\alpha(1; z_2) = \delta(1, 2) g^B_\alpha(x_1) - i \int d34 v(1, 3) G(3, 4) G(4, 3) \Gamma^{e-ph}_\alpha(4; z_2) \tag{23}$$

In the last line of Eq. (23) we have replaced $G$ with the noninteracting $G^0$ and thus obtained the polarization bubble $P^0$. We now invert this equation and find

$$\Gamma^{e-ph}_\alpha(1; z_2) = \left[ \delta(1, 4) - \int d3 v(1, 3) P^0(3, 4) \right]^{-1} \delta(1, 2) g^R_\alpha(x_1)$$

$$= \int dx_2 \epsilon^{-1}(1, 2) g^R_\alpha(x_2) = g_S(1, z_2), \tag{24}$$

where we used Eq. (14) to get to the second line.

We have found that in the Hartree approximation, the
electron-phonon vertex function reduces to the RPA, dynamically screened e–ph coupling. By substituting Eq. (23) in Eq. (10), we then get an expression for the first-order self-energy \( \Pi \) which is \textit{asymmetrically} screened (BS) on its sides:

\[
\Pi_{\alpha\beta}^{BS}(z_1, z_2) = i \int d3 \int dx_1 x_2 \, g_{\alpha}(x_1) \, P^0(1, 3) g_{\beta}^{S}(3, 2)
\]

(25)

**F. Analysis of the OE-free self-energy**

The self-energy \( \Pi^{BS} \), Eq. (25), is a \textit{left} self-energy, i.e., it is derived as the first-order truncation of \( \Pi^S \) appearing in the \textit{left} EOM, Eq. (7). It is clear from the graphical representation of \( \Pi^{BS} \) in Fig. 1(c) that this self-energy does not suffer from the double counting problem. In fact, the diagram is symmetric in the “bubble” expansion. The \textit{right} self-energy \( \Pi^R \), appearing in the \textit{adjoint} EOM Eq. (8), is instead screened on the opposite side, i.e., \( \Pi^{SB} \). It is also clear that it does not matter which side of the self-energy is screened, as long as only one side is. Indeed, Eq. (25) can also be made manifestly symmetric with the following steps, which use the fact that \( P^0 \) and \( \epsilon^{-1} \) are symmetric with respect to the interchange of their arguments:

\[
\Pi_{\alpha\beta}^{BS}(z_1, z_2) = 
\begin{align*}
&= i \int d3 \int dx_1 x_2 \, g_{\alpha}^{B}(x_1) \, P^0(1, 3) \epsilon^{-1}(3, 2) g_{\beta}^{B}(x_2) \\
&= i \int d3 \int dx_1 x_2 \, g_{\alpha}^{B}(x_1) \epsilon^{-1}(2, 3) P^0(3, 1) g_{\beta}^{B}(x_2) \\
&= i \int d3 \int dx_1 x_2 \, \chi(x_1) \epsilon^{-1}(1, 2) g_{\beta}^{B}(x_2).
\end{align*}
\]

(26)

In the last line we have used the relation

\[
\chi(1, 2) = \int d3 \, \epsilon^{-1}(1, 3) P^0(3, 2),
\]

(27)

which follows from Eqs. (12) and (14), in order to obtain the RPA response function \( \chi \). These steps are shown graphically in Fig. 1(c).

Obviously, the same result can be obtained in the case of \( \Pi^{SB} \), as can be seen diagrammatically and from the second line of Eq. (26). In fact, in order to obtain \( \Pi^{SB} \) it is sufficient to move the dynamical screening \( \epsilon^{-1} \) (represented by the diagonally barred ellipse) to the left side of \( P^0 \) in Fig. 1(c). Then the same perturbative “bubble” expansion as shown can be repeated. This means that a symmetric form involving \( g^S \) and \( P^0 \) instead of \( \epsilon^{-1} \) is also valid in the statically screened case – is obtained by defining \( \Pi \equiv (\Pi^{SB} + \Pi^{BS})/2 \) in this way:

\[
\Pi_{\alpha\beta}(z_1, z_2) = \frac{1}{2} \left( \Pi_{\alpha\beta}^{BS}(z_1, z_2) + \Pi_{\alpha\beta}^{SB}(z_1, z_2) \right) \\
= i \int d3 \int dx_1 x_2 \, g_{\alpha}^{B}(x_1) \chi(x_1, 2) g_{\beta}^{B}(x_2)
\]

(28)

This last expression for the self-energy \( \Pi \) is relevant because in practical equilibrium calculations, such as in DFPT, we may only have access to the statically screened \( g^S \) matrix elements, not to their time or frequency dependence. In particular, if we take the DFPT/static screening approximation then Eq. (25) reduces to

\[
\Pi_{\alpha\beta}^{BS}(z_1, z_2) = i \int dx_1 x_2 \, g_{\alpha}^{B}(x_1) \, P^0(1, 2) g_{\beta}^{S}(x_2).
\]

(29)

This means that the electrons instantaneously adjust to the variation of the Coulombian electron-ion interaction induced by the lattice vibrations, with no retardation effects. The \( g^S \) are then to be used as “ingredients” of larger simulations, and it is not straightforward to extract a dielectric function from them. Therefore, it is much easier to employ Eq. (28) to obtain a manifestly symmetric form of the coupling also in the practical case, with the advantage of ensuring that the coupling strength remains real, as we shall see below. Let us further remark that while the DFPT/static screening approximation is at the basis of an equilibrium treatment, further analysis of Eq. (25) in the context of the Kadanoff-Baym EOMs, i.e., Eqs. (7)–(8), is required before taking this approximation out of equilibrium.

**III. COUPLING STRENGTH AND PHONON LINEWIDTHS AT EQUILIBRIUM**

We now specialise our treatment to the equilibrium case. To follow this Section, we first direct the reader to Chap. 3 of Mahan’s book for a discussion of the finite-temperature, equilibrium Matsubara formalism in which the retarded self-energy may be calculated. This reference is enough to translate the concise list of steps given below into actual calculations.

**A. Dyson’s equation at equilibrium**

We consider the overscreened self-energy \( \Pi_{\alpha\beta}^{SBS}(z_1, z_2) \) – Eq. (20) – and the symmetrised, correctly screened self-energy \( \Pi_{\alpha\beta}(z_1, z_2) \) – Eq. (28). Both equations are taken in the statically screened case. First, we write these expressions in the electron single-particle basis rather than in the position basis. Second, we take the “Matsubara” component of the Keldysh expressions with \( \tau \)-time arguments lying on the imaginary axis, i.e., \( \Pi_{\alpha\beta}^{SBS}(\tau_1, \tau_2) \) and \( \Pi_{\alpha\beta}(\tau_1, \tau_2) \). This Keldysh component describes the properties at thermodynamic equilibrium. Third, we consider only the diagonal elements of the self-energies: \( \Pi_{\alpha\alpha}(\tau_1, \tau_2) = \delta_{\alpha\alpha} \Pi_{\tau\tau}(\tau_1, \tau_2) \). Fourth, we separate the generalised phonon and electron indices into branch/band index and momentum index: \( \alpha \to \lambda q, i \to nk, j \to mk \) with \( k' = k - q \) because of momentum conservation. Fifth, being at equilibrium, we switch to frequency space via Fourier transform, obtaining \( \Pi_{\lambda\lambda}^{SBS}(\omega_n) \)
(a) Legend

- $g^B$ Eq. (15)
- $g^S$ (static) Eq. (17)
- $G^0$
- $\epsilon^{-1}$ Eq. (11)
- $G^0 G^0, P^0$ Eq. (13)
- $\Gamma$
- $\chi$ Eq. (12)

(b) $\Pi^{SS}(t_1, t_2)$ (overscreened case)

![Graphical representation of the double-counting problem in the overscreened $\Pi^{SS}$ self-energy and its embedding in the static screening approximation.](image)

Used in state-of-the-art calculations

(c) $\Pi^{BS}(t_1, t_2)$

![Graphical representation of the correct treatment of double counting in the self-energy $\Pi^{BS}$.](image)

Correct treatment

(Symmetrised version, Eq. (28), used in this work.)

Figure 1. Graphical representation of the main equations relevant to the phonon self-energy discussion in Sec. II (a) Legend of the symbols corresponding to the many-body quantities that appear below. The equations in which they are defined are marked. (b) Double-counting problem in the overscreened $\Pi^{SS}$ self-energy and its embedding in the static screening approximation. (c) Double counting is removed in self-energy $\Pi^{BS}$. The graphs are only labelled by the time arguments for simplicity, but they may be transformed in proper Feynman diagrams with the appropriate additional labeling, prefactors, and diagrammatic rules.)
and $\Pi_{\lambda q}(i\omega_n)$; here $\omega_n = (2n+1)\pi/\beta$, with $n$ integer and $\beta$ the inverse temperature, is the Matsubara imaginary frequency. Sixth, we perform the Matsubara summation of the internal frequency, so that the only integration left is the one over momenta $k$. Furthermore, this latter integration is discretized as $\int d^3k/\Omega_{BZ} \to \sum_k/N_k$. Here, $\Omega_{BZ}$ is the reciprocal-space volume of the Brillouin zone (BZ), while $N_k$ is the number of $k$-points in a discrete mesh spanning the BZ itself. After this step, the EOMs in Eq. (7) and (8) reduce to the integral Dyson’s equation in frequency space, i.e.,

$$D_{\lambda q}^{QP}(i\omega_n) = D_{\lambda q}(i\omega_n) + D_{\lambda q}(i\omega_n)\Pi_{\lambda q}(i\omega_n)D_{\lambda q}^{QP}(i\omega_n),$$

(30)

where $D_{\lambda q}^{QP}$ is a quasiparticle phonon propagator including the nonadiabatic corrections to the phonon mode $\lambda q$ induced by $\Pi_{\lambda q}$. Its spectral function $\propto \text{Im} D_{\lambda q}^{QP}(\omega)$ can be obtained by inverting Eq. (30) and will include renormalised, complex phonon energies and electron-phonon satellites. The static phonon propagator in this form is

$$D_{\lambda q}(\omega) = -2\Omega_{\lambda q}/(\omega^2 + \Omega_{\lambda q}^2),$$

while the explicit form of the self-energies from Eqs. (20), (23) and (28) is, respectively,

$$\Pi_{\lambda q}^S(i\omega_n) = \frac{2}{N_k} \sum_{nmk} \frac{\Delta_{\lambda q,mnk}(\omega_n)}{\omega_n + \epsilon_{mk-q} - \epsilon_{nk}},$$

(31)

$$\Pi_{\lambda q}^{BS}(i\omega_n) = \frac{2}{N_k} \sum_{nmk} \frac{\Delta_{\lambda q,mnk}(\omega_n)}{\omega_n + \epsilon_{mk-q} - \epsilon_{nk}},$$

$$\Pi_{\lambda q}(i\omega_n) = \frac{2}{N_k} \sum_{nmk} \frac{\Delta_{\lambda q,mnk}(\omega_n)}{\omega_n + \epsilon_{mk-q} - \epsilon_{nk}}.$$  

Here $\epsilon_{nk}$ and $\epsilon_{mk-q}$ are electronic energies, the functions $f_{mk-q}$ and $f_{nk}$ are the temperature-dependent electronic Fermi-Dirac occupation factors and the prefactor of 2 comes from the spin summation. We see from these equations that the overscreened and overscreening–free self-energies only differ by the coupling strengths $\Delta_{\lambda q,mnk}$.

We also recall that because of their analytic structure – when taken as functions of real frequencies – the real and imaginary parts of the self-energies in Eq. (31) are not independent, but connected via Hilbert transformation:

$$\text{Re} \Pi_{\lambda q}(\omega) = \frac{1}{\pi} \int_{-\infty}^{+\infty} d\omega' \frac{\text{Im} \Pi_{\lambda q}(\omega')}{\omega' - \omega},$$

(32)

where $\mathcal{P}$ denotes the principal value. This means that in order to treat in a coherent way both the correction to the phonon energies and their linewidths – as they are related to the real and imaginary part of the self-energy, respectively – the same definition for the self-energy in Eq. (31) has to be used for both types of calculations.

### B. Coupling strengths

In the overscreened case, Eqs. (20) and top line of of Eq. (31), the coupling strength is simply

$$\Delta_{\lambda q,mnk}^S = |g_{\lambda q,mnk}^S|^2.$$  

(33)

In the case of $\Pi^{BS}$, Eq. (29) and middle line of Eq. (31), the question arises about how to treat the coupling strength, since the interaction is proportional to $g_{\lambda q,mnk}^B (g_{\lambda q,mnk}^S)^*$ which is a complex quantity, and the self-energy only has the left term. This is the expression that appears in Refs. [12] here it is not clear if $\text{Im} \Pi^{BS}$, which is responsible for the delta function structure in the linewidth, also includes the contribution from $\text{Im} g^B g^S$, or if $[g^B g^S]$ was taken instead, neglecting the complex phase. In both instances, it is not clear what the consequences are and how large an error is possibly made.

On the contrary, in our case we use the symmetrised expression Eq. (28), which contains both left and right components while yielding a real coupling strength. In fact, if we write the statically screened Eq. (28) at equilibrium and in the single-particle basis, we immediately see that

$$\gamma_{\lambda q,mnk}^{BS+SB} = \frac{1}{2} \left\{ g_{\lambda q,mnk}^S (g_{\lambda q,mnk}^B)^* + g_{\lambda q,mnk}^B (g_{\lambda q,mnk}^S)^* \right\} = \text{Re} g_{\lambda q,mnk}^S \text{Re} g_{\lambda q,mnk}^B + \text{Im} g_{\lambda q,mnk}^S \text{Im} g_{\lambda q,mnk}^B.$$  

(34)

We implemented Eq. (34) the Yambo code to evaluate phonon–electron coupling without the double counting error. We also note that this expression was used in Ref. [20] to study the effects of phonon frequency renormalisations in the “constrained DFPT” approach.

### C. From the the self-energies to the linewidths

The imaginary part of the retarded component of a quasiparticle self-energy, when evaluated at the energy of the quasiparticle itself, yields the spectral broadening of the corresponding quasiparticle peak. In particular, the full-width half-maximum (FWHM) of a peak with lorentzian lineshape is given by twice the imaginary part. Therefore, we take the self-energy and operate its analytic continuation to real frequencies, i.e., $\omega_n = \omega + i\eta$ with $\eta \to 0^+$. This procedure yields its “retarded” (i.e., physically meaningful at equilibrium) component. Thus, we obtain the linewidths according to

$$\gamma_{\lambda q}^S = 2 \text{Im} \Pi_{\lambda q}^S(\omega = \Omega_{\lambda q})$$

$$\gamma_{\lambda q}^{BS+SB} = 2 \text{Im} \Pi_{\lambda q}(\omega = \Omega_{\lambda q}).$$

(35)

The explicit expression for both cases is

$$\gamma_{\lambda q}^S = \frac{4\pi}{N_k} \sum_{nmk} \mathcal{S}(f_{mk-q} - f_{nk}) \delta(\Omega_{\lambda q} - [\epsilon_{nk} - \epsilon_{mk-q}]),$$

(36)

with $\mathcal{S} = SS$ or $BS + SB$. This equation describes the probabilities of energy-conserving scatterings between electron-hole pairs with single-particle energies $\epsilon_{nk}$ and $\epsilon_{mk-q}$ and phonons with energies $\Omega_{\lambda q}$. The coupling strength $g_{\lambda q,mnk}^S$ controls the probability of the scattering event and broadens the linewidth.
We may further employ the Allen formula, which permits to simplify Eq. (36) in such a way that its numerical evaluation is more feasible and converges faster. It was originally developed by Allen \cite{Allen} in order to express the electron-phonon Eliashberg spectral function – needed in the Migdal-Eliashberg theory for superconductivity – in terms of phonon linewidths, since at the time they were considered to be readily available quantities from experiments. It is strictly valid in the assumption of temperature-independent linewidths, and it consists in the following replacement for the electronic occupation function \cite{Allen}:

$$f_{nk-q} - f_{nk} \simeq \Omega_{\lambda q} \frac{\partial f}{\partial \varepsilon} \biggr|_{\varepsilon = \varepsilon_{nk}} \simeq \Omega_{\lambda q} \delta(\varepsilon_{nk})$$ (37)

(where the energy-conserving delta function is enforced and $f(\varepsilon) \sim \theta(\varepsilon)$).

Thus, we arrive at the final expressions:

$$\gamma_{\lambda q} = \frac{4\pi \Omega_{\lambda q}}{N_k} \sum_{nmk} g_{nm,kq,\lambda} \delta(\varepsilon_{nk}) \delta(\Omega_{\lambda q} - [\varepsilon_{nk} - \varepsilon_{mk-q}]).$$ (38)

These are the same equations presented in our companion paper \cite{Yambo}.

IV. CODE DEVELOPMENT

The codes we used for the implementation of this work and the subsequent numerical calculations were Quantum Espresso (QE) \cite{QE} for the density functional theory (DFT) and density functional perturbation theory (DFPT) steps, and Yambo \cite{Yambo} for the calculation of the phonon linewidths. Below we discuss the code implementation that was necessary to compute Eq. (38), and next we describe the general scheme of a linewidth calculation.

A. Code implementation

Quantum Espresso. The bare electron-phonon matrix elements, $g_{nm,kq}^B$, were extracted from a QE-DFPT calculation by modifying the part relative to the ph.x executable and in particular the subroutines contained in /PHonon/PH/elphon.f90 so that the bare matrix elements could be stored and printed in a format readable by Yambo. The modifications were done on version 6.6 of the QE distribution \cite{QE}. Note that we directly extract the (complex) spatially integrated matrix elements of the (real) variation of the bare electron-phonon interaction.

Yambo. Equations (36), (34) and (38) were implemented in Yambo – we used version 5.101 as part of the “phonon” project relative to the yambo_ph executable. The implementation is fully parallel. Since Eqs. (36) and (38) require a very large $k$-point mesh in reciprocal space to be accurately converged, a double grid support was added in order to compute electronic eigenvalues on a finer grid with respect to the one used for the electron-phonon calculations. We denote the fine grid as FG and the original, coarse grid as CG. From now on, reciprocal-space points belonging to the FG (CG) are written as lowercase $k$ (calligraphic $K$). The FG can be both a regular or random grid (we used the latter). Using Eq. (38) as an example, this means that the corresponding expression computed by Yambo when the double grid support is turned on is

$$\gamma_{\lambda q} = \frac{4\pi \Omega_{\lambda q}}{N_K} \sum_{K \in CG} \sum_{nm} g_{nm,kq,K} \frac{1}{n_k} \sum_{k \in FG} \frac{1}{n_k} \sum_{k' \in FG} \frac{1}{n_{k'}} \frac{1}{\pi} \frac{1}{\varepsilon_{nk} - i\eta} \frac{1}{\pi} \frac{1}{\Omega_{\lambda q} - \Delta_{nk,mk'} - i\eta},$$ (39)

with $\Delta_{nk,mk'} = \varepsilon_{nk} - \varepsilon_{mk'}$ and $k' = k - q$. Note here that the FG depends on the CG it was generated from. In particular, $N_K$ is the number of CG points in the Brillouin Zone (BZ), while $\sum_{k \in FG}$ represents a sum over the subset of the FG random $k$-points which are closest to each $K$-point of the original CG. The number of $k$-points contained in each $K$ subset (which may vary because of randomness and when close to the BZ edge) is $n_K$. For each $K$-point, the FG subsets around $K$ and $K' = K - q$ are both needed. Crucially, both the CG and the FG must undergo convergence tests: a python workflow using the yambopy package was created to automatically generate CG-FG pairs. The numerical evaluation of the delta functions involves the broadening parameter $\eta$, which has to be chosen – naturally, as small as possible – according to the densities of the CG and FG grids. In addition, in order to avoid the unnecessary, time- and memory-expensive counting of transitions contributing negligibly, Yambo automatically selects only transitions satisfying $\varepsilon_{nk} - \varepsilon_{mk-q} \leq \Omega_{\lambda q} \pm 3\eta$. Note finally that here the e–ph matrix elements are properly normalized by the phonon energies, i.e., $g_{nm,kq,B} \rightarrow g_{nm,kq,B}/\sqrt{2\Omega_{\lambda q}}$.

The implementation described here is not released yet, but is readily available upon request to the corresponding author.

B. Calculation flow

The ab initio phonon linewidths calculation comprises the following six interdependent steps, which are de-
scribed in detail in Fig. 2 and we briefly summarize here. In order to give an assessment of the numerical load, the scheme also lists the converged values of the various reciprocal-space grids used to calculate the linewidths in MgB$_2$ as reported in our companion paper$^{22}$, with the input files available in the Supplemental Material. We will cite these input files here as practical examples.

(i) Self-consistent-field (scf) ground-state calculation using a regular $\{k\}_{scf}$ grid. (ii) Derivatives of the scf potential (dV$_{scf}$) and interatomic force constants calculation using a regular $\{k\}_{dVscf}$ grid. This fixes the list of phonon momenta $q$, which may be automatically generated (regular grid) or customly chosen. (iii) Non-self-consistent-field (nscf) calculation. The CG $\{k\}_{CG}$ grid used in this calculation defines the $K$-points and has to be carefully converged together with the $FG$ $\{k\}_{FG}$ from Step (v) and the broadening parameter $\eta$. The grids convergence can be tested on the phonon linewidths calculations $\gamma_{\lambda q}$ – step (vi) – looking both at $q$-averages along the BZ and at the values at high-symmetry $q$-points. This step also computes the electron energies $\varepsilon_{nk}$. (iv) Electron-phonon matrix elements (elph) calculation. In this step – to be run on top of Steps (ii) and (iii) – the $g_{B/S}^{\lambda_q\lambda_{qn}nk}$ are computed. (v) Second non-self-consistent-field (nscf) calculation. This fixes the $FG$, which defines the $k$-points. Random $k$-points are used since they yield faster convergence, and the randomly-distributed $FG$ $\{k\}_{FG}$ points can be generated with Yambo. The $FG$ has to be carefully converged together with the CG $\{k\}_{CG}$ from Step (iii) and the broadening parameter $\eta$ (see Step (iii)). It is in this step that the fine-grid electron energies $\varepsilon_{nk}$ are computed. (vi) Phonon linewidths calculation. This is the final calculation that yields $\gamma_{\lambda q}$. The results of steps (iv) and (v) are collected by Yambo, and then used to compute Eq. (39).

V. CONCLUSION

In this work we developed a general framework for the treatment of electron-phonon interaction avoiding the overscreening error. This amounts to the removal of a systematic error previously affecting the theoretical prediction a wide range of physical phenomena which depend on the phonon lifetimes$^{5,23,24,36–42}$. In addition to removing the systematic overscreening, this approach does not rely on a static screening assumption imposed from the start, thus neglecting the dynamical dependence of the electron-phonon coupling strengths. We believe that our treatment makes it clear that this is not a sound way to proceed in the case of out-of-equilibrium systems. Therefore, we believe this general approach to be mandatory for any description of coupled electron-lattice dynamics, including ultrafast relaxation processes, i.e., those processes where the phononic EOMs from Eqs. (7) and (8) are coupled to the corresponding EOMs for the electronic propagator$^{26,40,43,44}$.

We also provided a first-principles numerical scheme for the calculation of overscreening error–free phonon linewidths in the equilibrium case, at no additional cost with respect to the state-of-the-art, systematically overscreened approach. This can be applied to any system whose mean-field description is accessible via DFPT. We think it will be interesting to carry out extensive OE–free calculations on various relevant superconducting systems (such as 2D superconductors$^{45,46}$) in order to investigate whether giant corrections such as those observed in MgB$_2$ occur frequently. Besides, our equilibrium implementation can be easily generalised to obtain the full frequency-dependent phonon spectral function.

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Figure 2. Scheme of the linewidths calculation flow discussed in Sec. IV. The calculations corresponding to the various steps are shown in boxes. If two boxes are connected by an arrow, it means that the calculation at the ending point depends on the calculation at the starting point. The various calculations may depend on different meshes of reciprocal-space points (\{k\} for electrons, \{q\} for phonons): these meshes are shown in the boxes along with the values we used in Ref. 22 to obtain our results. On the side of the boxes, additional descriptions of the type of calculations are provided, together with the software package needed (QE or Yambo) and the specific executables (\texttt{pw.x}, \texttt{ph.x}, \texttt{yambo}, \texttt{ypp}, \texttt{yambo_ph}, \texttt{ypp_ph}) in brackets. The green (red) frame denotes the calculations which depend on the coarse grid \textit{CG} of \textit{K}-points (fine grid \textit{FG} of \textit{k}-points). See text for more information.
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The relation between the names of the input files provided in the Supplemental Material of Ref. 22 and the calculation steps described in the text is as follows. mgb2.scf for the scf step, mgb2.dvscf for the dvscf step, mgb2.nscf for the first nscf / coarse grid step, mgb2.elph for the elph step, mgb2.nscf_FG for the second nscf / fine grid step, mgb2_phel_BS-SS_broad_5_meV_NkCG_13824_NkDG_55296.in and mgb2_phel_SS_broad_5_meV_NkCG_13824_NkDG_55296.in for the linewidths step in the OE–free and OE cases, respectively.

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