

***Ab initio* theory of coherent phonon damping in semimetals**Yiming Pan¹, Christoph Emeis¹, Stephan Jauernik², Michael Bauer^{2,3} and Fabio Caruso^{1,3}¹*Institute of Theoretical Physics and Astrophysics, Kiel University, 24118 Kiel, Germany*²*Institute of Experimental and Applied Physics, Kiel University, 24118 Kiel, Germany*³*Kiel Nano, Surface and Interface Science KiNSIS, Kiel University, 24118 Kiel, Germany*

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Phonon decoherence determines the characteristic timescales over which coherent lattice vibrations decay, making it a crucial process for understanding the nonequilibrium dynamics of crystal lattices after excitation by a pump pulse. Here, we report a theoretical and computational investigation of the origin of phonon decoherence within a first-principles many-body framework. We derive quantum kinetic equations for the dynamics of coherent phonons by explicitly accounting for dissipation processes induced by electron-phonon and phonon-phonon interactions. The decoherence rate and frequency renormalization are formulated in terms of the nonequilibrium phonon self-energy, providing a framework amenable for *ab initio* calculations. To validate this approach, we conduct a first-principles study of phonon decoherence for the elemental semimetals antimony and bismuth. The robust agreement with available temperature- and fluence-dependent experimental data confirms the accuracy of our theoretical and computational framework. More generally, our findings reveal that either electron-phonon or phonon-phonon coupling can prevail in determining the decoherence time, depending on the temperature and driving conditions. Overall, this work fills a critical gap in the theoretical understanding of phonon decoherence, providing a predictive framework for determining the timescales of light-induced structural dynamics in driven solids.

DOI: [10.1103/5291-19g4](https://doi.org/10.1103/5291-19g4)**I. INTRODUCTION**

Recent advances in laser technology have expanded the opportunities to optically control the coherent motion of crystal lattices. Coherent phonons—oscillatory displacements of atoms from equilibrium—enable a wide range of nonequilibrium phenomena, including the initiation of nonthermal phase transitions [1–4], access to metastable phases [5–7], control of material order parameters [8–10], and the activation of optically inactive modes via nonlinear phononic principles [11]. Coherent phonons can undergo a variety of excitation pathways, which depend on the electronic and crystal structures as well as driving conditions such as light frequency, fluence, and polarization. Theoretical investigations of coherent phonons have primarily addressed the excitation principles at the origin of coherent lattice motion [9,12–19]. Conversely, with the exception of a few numerical studies based on molecular dynamics simulations [20,21], a systematic description of phonon decoherence, namely, the decay of a coherent state resulting in the progressive disappearance of coherent phonon oscillations, has thus far remained uncharted.

Phonon damping has been extensively discussed under equilibrium conditions, that is, in the absence of coherent motion [22–32]. There is *a priori* no formal justification for

directly employing the equilibrium theory of phonon damping to model phonon decoherence out of equilibrium. Based on the observation that decoherence occurs over timescales of the order of a few picoseconds, its origin is typically attributed to anharmonic phonon-phonon scattering processes. The timescales of decoherence in semimetals, however, further exhibit a striking dependence of the driving fluence, namely, the energy per unit area of the pump, suggesting that electron-phonon interactions mediated by photoexcited carriers can also prevail under suitable conditions [18,33]. This behavior is schematically illustrated in Fig. 1: An increase in driving fluence simultaneously enhances the oscillation amplitudes arising from coherent phonons while accelerating decoherence, resulting in an inverse proportionality between lifetimes and driving fluence (inset).

The description of phonon decoherence has thus far been limited to the inclusion of a phenomenological damping term in the equation of motion [9,12,16]; however, this approach is not suitable for clarifying the origin of decoherence and its dependence on materials' properties and excitation conditions. Overall, a rigorous and predictive theory capable of accounting for the energy dissipation of coherent phonons induced by an interaction with other degrees of freedom of the crystal remains elusive. As decoherence largely influences the characteristic timescales of coherent lattice motion, this constitutes a critical gap in the understanding of light-induced structural motion in materials, raising the question of how to systematically account for dissipation within *ab initio* theories and computational frameworks addressing light-induced structural motion.

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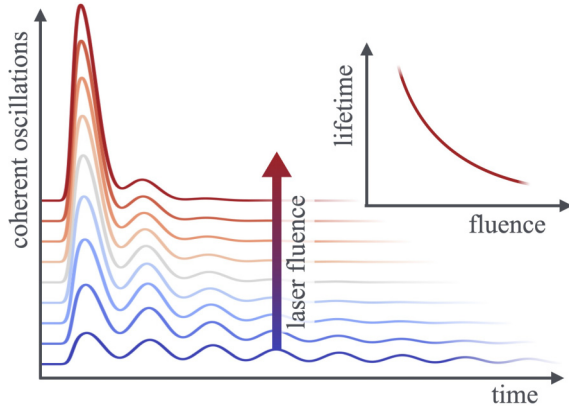


FIG. 1. Schematic illustration of the characteristic fluence dependence of the coherent phonon amplitude and decoherence. Inset: dependence of the decoherence time on the fluence.

In this paper, we investigate the origin of dissipation in the dynamics of coherent phonons on the basis of an *ab initio* many-body approach. We derive quantum kinetic equations for the dissipative coherent phonons dynamics in the presence of electron-phonon and phonon-phonon interactions. Specifically, the decoherence rates and frequency renormalization are formulated in terms of the nonequilibrium phonon self-energy due to electron-phonon and phonon-phonon coupling. We apply this approach to the elemental semimetals antimony and bismuth, for which a thorough experimental characterization of phonon decoherence has been reported. Overall, our results are in good quantitative agreement with experiments, and they capture the temperature and fluence dependence of the decoherence. These findings provide the foundation for future computational studies of phonon decoherence in materials.

II. THEORY OF PHONON DECOHERENCE

In this section, we define the theoretical framework for the description of phonon decoherence and frequency renormalization in anharmonic solids. The harmonic nonadiabatic Hamiltonian introduced in Ref. [34] and its extension to account for three-phonon scattering processes constitute the starting point for the description of phonon decoherence in this paper. Since this is a nonstandard Hamiltonian, we discuss its definition in Sec. II A. In Secs. II B and II C, we derive explicit expressions suitable for first-principles calculations of the decoherence rate and frequency renormalization due to electron-phonon and phonon-phonon coupling, respectively.

A. The phonon Hamiltonian

We consider the *ab initio* Hamiltonian in the presence of electron-nucleus interactions:

$$\hat{H} = \hat{H}_e + \hat{H}_{0,p} + \hat{H}_{ep} + \hat{H}_{ext}, \quad (1)$$

where the total Hamiltonian is split into electronic Hamiltonian $\hat{H}_e$, free phonon Hamiltonian $\hat{H}_{0,p}$, and electron-phonon interaction Hamiltonian $\hat{H}_{ep}$ and $\hat{H}_{ext}$ is the coupling of electrons to the external field. This choice for the Hamiltonian relies on the assumption that nuclear displacements from the

equilibrium coordinates remain small throughout the nonequilibrium dynamics of the system. This condition assumes that the potential energy surface is well approximated by its third-order Taylor expansion. It is satisfied by coherent phonons in semimetals, where the light-induced nuclear displacements are of the order of 10^{-3} Å [35]. Conversely, it is violated for highly anharmonic crystals, such as materials described by double-well potentials and in the proximity of second-order structural phase transitions. The electronic Hamiltonian $\hat{H}_e$ is the sum of free electrons and electron-electron interaction:

$$\hat{H}_e = \int d\mathbf{r} \hat{\psi}^\dagger(\mathbf{r}) \left[-\frac{\nabla^2}{2} + V^{\text{ion}}(\mathbf{r}; \mathbf{R}_0) \right] \hat{\psi}(\mathbf{r}) + \frac{1}{2} \int d\mathbf{r} d\mathbf{r}' \hat{\psi}^\dagger(\mathbf{r}) \hat{\psi}^\dagger(\mathbf{r}') v(\mathbf{r} - \mathbf{r}') \hat{\psi}(\mathbf{r}') \hat{\psi}(\mathbf{r}), \quad (2)$$

where the bare ionic potential $V^{\text{ion}}(\mathbf{r}; \mathbf{R}_0)$ is evaluated at equilibrium ionic configuration $\mathbf{R}_0$ and $v(\mathbf{r} - \mathbf{r}')$ represents the Coulomb interactions between the electrons.

The free phonon Hamiltonian $\hat{H}_{0,p}$ and electron-phonon interaction $\hat{H}_{ep}$ can be expanded up to second order in the displacement amplitude $\hat{U}_{q\nu}$ [34]:

$$\hat{H}_{0,p} = \frac{1}{2} \sum_{\mathbf{q}\nu} \hat{P}_{-\mathbf{q}\nu} \hat{P}_{\mathbf{q}\nu} + \frac{1}{2} \sum_{\mathbf{q}\nu\nu'} \hat{U}_{-\mathbf{q}\nu} K_{\mathbf{q}\nu\nu'} \hat{U}_{\mathbf{q}\nu}, \quad (3)$$

$$\hat{H}_{ep} = \int d\mathbf{r} [\hat{n}(\mathbf{r}) - n_0(\mathbf{r})] \sum_{\mathbf{q}\nu} g_{\mathbf{q}\nu}(\mathbf{r}) \hat{U}_{\mathbf{q}\nu}. \quad (4)$$

The density operator $\hat{n}(\mathbf{r}) = \hat{\psi}^\dagger(\mathbf{r}) \hat{\psi}(\mathbf{r})$ is given in terms of fermionic field operators $\hat{\psi}(\mathbf{r})$ and $\hat{\psi}^\dagger(\mathbf{r})$. The electron-phonon coupling function is defined as

$$g_{\mathbf{q}\nu}(\mathbf{r}) = \sum_{p\kappa} \frac{1}{\sqrt{NM_\kappa}} e^{i\mathbf{q}\cdot\mathbf{R}_p} \mathbf{e}_{\mathbf{q}\nu}^\kappa \cdot \left. \frac{\partial V^{\text{ion}}(\mathbf{r})}{\partial \Delta \mathbf{\tau}_{\kappa p}} \right|_{\{\mathbf{\tau}_{\kappa p}\}=\mathbf{R}_0}. \quad (5)$$

Here, $\mathbf{R}_p$ is a crystal lattice vector, M_κ is the mass of the κ th nucleus, $\mathbf{e}_{\mathbf{q}\nu}^\kappa$ denotes a normal-mode basis vector, $\Delta \mathbf{\tau}_{\kappa p}$ is the displacement vector of the κ th atom in the p th unit cell from its equilibrium coordinate, and $V^{\text{ion}}(\mathbf{r}) = -\sum_{\kappa p} Z_\kappa |\mathbf{r} - \mathbf{\tau}_{\kappa p}|^{-1}$ is the electron-nucleus potential. The sum over p and κ runs over all unit cells in the Born-von Kármán supercell and over all atoms in the unit cell, respectively.

The operator $\hat{U}_{\mathbf{q}\nu}$ characterizes the coherent displacement of the lattice along the mode $(\mathbf{q}, \nu)$. It is related to the Cartesian representation of the displacement operator $\Delta \hat{\mathbf{\tau}}_{\kappa p}$ via the normal coordinate transformation:

$$\Delta \hat{\mathbf{\tau}}_{\kappa p} = (NM_\kappa)^{-\frac{1}{2}} \sum_{\mathbf{q}\nu} e^{i\mathbf{q}\cdot\mathbf{R}_p} \mathbf{e}_{\mathbf{q}\nu}^\kappa \hat{U}_{\mathbf{q}\nu}. \quad (6)$$

$\hat{P}_{\mathbf{q}\nu}$ is the canonical momentum defined by the commutation relation $[\hat{U}_{\mathbf{q}\nu}, \hat{P}_{\mathbf{q}'\nu'}] = i\delta_{-\mathbf{q},\mathbf{q}'}\delta_{\nu,\nu'}$. In Eq. (3), $K_{\mathbf{q}\nu\nu'}$ is the normal coordinate representation of the elastic tensor, defined as

$$K_{\mathbf{q}\nu\nu'} = \sum_{\kappa\kappa'p} e^{i\mathbf{q}\cdot\mathbf{R}_p} \mathbf{e}_{\mathbf{q}\nu}^\kappa \cdot \frac{\mathbf{K}_{\kappa p, \kappa'0}}{\sqrt{M_\kappa M_{\kappa'}}} \cdot \mathbf{e}_{\mathbf{q}\nu'}^{\kappa'}, \quad (7)$$

where $\mathbf{K}_{\kappa p, \kappa'p'} = \Delta_{\kappa p} E_{n-n} + \int d\mathbf{r} n_0(\mathbf{r}) \Delta_{\kappa p} \frac{V^{\text{ion}}}{\kappa' p'}$, we introduced the abbreviation $\Delta_{\kappa p} = \frac{\partial^2}{\partial \Delta \mathbf{\tau}_{\kappa p} \partial \Delta \mathbf{\tau}_{\kappa' p'}}$, and $E_{n-n} = \frac{1}{2} \sum_{\kappa p \neq \kappa' p'} Z_\kappa Z_{\kappa'} |\mathbf{\tau}_{\kappa p} - \mathbf{\tau}_{\kappa' p'}|^{-1}$ is the nucleus-nucleus Coulomb energy.

Finally, in order to launch the coherent phonon, one needs to drive the system into the nonequilibrium state via an external field. This is accounted for by the time-dependent Hamiltonian $\hat{H}_{\text{ext}}(t)$. In the case where the electronic system is driven by an ultrafast pump field, this Hamiltonian is represented by $\hat{H}_{\text{ext}} = \int d\mathbf{r} \hat{\psi}^\dagger(\mathbf{r})[\mathbf{r} \cdot \mathbf{E}(t)]\hat{\psi}(\mathbf{r})$ in the length gauge. Here, we assume that the external field acts on timescales much faster than those of ionic motions. We replace $\hat{H}_{\text{ext}}$ with a light-induced electronic density fluctuation at the beginning $\delta n_0(\mathbf{r})$. This choice is consistent with the assumptions used in the mechanism of displacement excitation of a coherent phonon [35]. However, the formalism discussed here is general, and it can also be applied to models with explicit time-dependent driving fields.

The relation between the Hamiltonian defined by Eq. (3) and the adiabatic (Born-Oppenheimer) harmonic Hamiltonian—the usual starting point for the definition of phonons in solids—was discussed in detail in Ref. [34]. The key differences of relevance for this work are briefly summarized below: (1) Eq. (3) does not rely on the Born-Oppenheimer approximation; (2) the elastic tensor $K_{\mathbf{q}\nu\nu'}$ includes only the contribution of the bare nuclei, and it is, in general, not positive definite [34,36]. As a consequence, the eigenvalues of $\mathbf{K}$ are not positive definite and cannot be interpreted as normal vibrational frequencies of the lattice. (3) Adiabatic-harmonic phonon frequencies are defined only in terms of the adiabatic phonon self-energy; (4) the lattice dynamics depends on the time-dependent electron density $\hat{n}(\mathbf{r}, t)$ via Eq. (4).

B. Decoherence and frequency renormalization mediated by electron-phonon coupling

In the following, we proceed to derive the damped equation of motion of the nuclear displacements $U_{\mathbf{q}\nu}$. The Heisenberg picture is implied throughout. We begin by considering the Heisenberg equation of motion for the displacement operator:

$$\frac{\partial^2 U_{\mathbf{q}\nu}}{\partial t^2} = -\langle [\hat{U}_{\mathbf{q}\nu}, [\hat{U}_{\mathbf{q}\nu}, \hat{H}]] \rangle. \quad (8)$$

Making use of Eqs. (1)–(4) along with the commutation relations for $U_{\mathbf{q}\nu}$ and $P_{\mathbf{q}\nu}$, one readily obtains

$$\frac{\partial^2 U_{\mathbf{q}\nu}}{\partial t^2} + \sum_{\nu'} K_{\mathbf{q}\nu\nu'} U_{\mathbf{q}\nu'} = - \int d\mathbf{r} g_{-\mathbf{q}\nu}(\mathbf{r}) \delta n(\mathbf{r}, t). \quad (9)$$

The right-hand side represents the force exerted on the lattice by a time-dependent density fluctuation, $\delta n(\mathbf{r}, t) = n(\mathbf{r}, t) - n_0(\mathbf{r})$, where $n_0(\mathbf{r})$ denotes the equilibrium density. Here, we consider the case in which finite values for $\delta n(\mathbf{r}, t)$ result from time-dependent perturbations, such as an external driving field coupled to the electron density, resulting in the excitation of coherent phonons.

Equation (9) resembles the equation of motion of an undamped driven harmonic oscillator, which is reflected by the absence of a term proportional to $\partial U_{\mathbf{q}\nu}/\partial t$. However, since Eq. (9) is an exact result for the Hamiltonian specified by Eqs. (1)–(4), we expect the effects of decoherence to be encoded in the density fluctuation $\delta n(\mathbf{r}, t)$. To retrieve an explicit expression for the decoherence rate mediated by the electron-

phonon coupling, we express $\delta n(\mathbf{r}, t)$ by making use of linear response theory:

$$\delta n(\mathbf{r}, t) = \delta n(\mathbf{r}, t_0) + \int d\mathbf{r}' \int_{t_0}^t dt' \chi^r(\mathbf{r}, t; \mathbf{r}', t') \delta V^{\text{ion}}(\mathbf{r}', t'), \quad (10)$$

where $\delta n(\mathbf{r}, t_0)$ is the variation of the electronic density induced by the time-dependent perturbation (i.e., a pump pulse) responsible for triggering the coherent dynamics of the lattice and the second term accounts for density fluctuations induced by the electron-phonon interactions. $\chi^r(\mathbf{r}, t; \mathbf{r}', t')$ is the retarded density-density response function. The response function incorporates the many-body interaction and is reducible in v , the electron-electron interaction. However, since the variation of the potential is induced by ionic displacement, i.e., $\delta V^{\text{ion}}(\mathbf{r}, t) = \sum_{\mathbf{q}\nu} g_{\mathbf{q}\nu}(\mathbf{r}) U_{\mathbf{q}\nu}(t)$, it is irreducible in phonons [37].

By introducing the adiabatic and nonadiabatic density responses to the electron-phonon interactions, δn^A and δn^{NA} , respectively, we can rewrite Eq. (10) as

$$\delta n(\mathbf{r}, t) = \delta n(\mathbf{r}, t_0) + \delta n^A(\mathbf{r}, t) + \delta n^{\text{NA}}(\mathbf{r}, t). \quad (11)$$

The adiabatic density response arises from the instantaneous response of the density [38,39] to the nuclear displacement $U_{\mathbf{q}\nu}$, and it can be derived by assuming a parametric dependence in the form $\delta n^A(\mathbf{r}, t) = \delta n^A(\mathbf{r}; U_{\mathbf{q}\nu}(t))$, leading to (see Appendix B of Ref. [34])

$$\delta n^A(\mathbf{r}, t) = \int d\mathbf{r}' \chi^r(\mathbf{r}, \mathbf{r}'; 0) \sum_{\nu'} g_{\mathbf{q}\nu'}(\mathbf{r}') U_{\mathbf{q}\nu'}(t), \quad (12)$$

where $\chi^r(\mathbf{r}, \mathbf{r}'; 0)$ is the Fourier transform of $\chi^r(\mathbf{r}, t; \mathbf{r}', t')$ at zero frequency.

Substitution of Eqs. (10)–(12) into (9) enables us, after a few algebraic manipulations, to rewrite the equation of motion for coherent phonons in the presence of electron-phonon interactions in the form [40]

$$\frac{\partial^2 U_{\mathbf{q}\nu}}{\partial t^2} + 2\Gamma_{\mathbf{q}\nu}^{\text{ep}} \frac{\partial U_{\mathbf{q}\nu}}{\partial t} + \Omega_{\mathbf{q}\nu}^2 U_{\mathbf{q}\nu} = - \int d\mathbf{r} g_{-\mathbf{q}\nu}(\mathbf{r}) \delta n(\mathbf{r}, t_0). \quad (13)$$

This expression resembles the equation of motion for a driven-damped harmonic oscillator, with the general solution $U_{\mathbf{q}\nu}(t) = U_{\mathbf{q}\nu}^0 e^{-\Gamma_{\mathbf{q}\nu}^{\text{ep}} t} \sin(\Omega_{\mathbf{q}\nu} t + \varphi)$. The quantities $\Gamma_{\mathbf{q}\nu}^{\text{ep}}$ and $\Omega_{\mathbf{q}\nu}$ can be identified with the decoherence rate and with the renormalized vibrational frequency, respectively, and are defined as

$$\Gamma_{\mathbf{q}\nu}^{\text{ep}} = -\text{Im} \Pi_{\mathbf{q}\nu}^{\text{ep,NA}}, \quad (14)$$

$$\Omega_{\mathbf{q}\nu}^2 = \omega_{\mathbf{q}\nu}^2 + 2\omega_{\mathbf{q}\nu} \text{Re} \Pi_{\mathbf{q}\nu}^{\text{ep,NA}}. \quad (15)$$

Here, $\Pi_{\mathbf{q}\nu}^{\text{ep,NA}}$ is the nonadiabatic contribution to the phonon self-energy due to the electron-phonon interaction, evaluated within the on-the-mass-shell approximation (OMSA):

$$\Pi_{\mathbf{q}\nu}^{\text{ep,NA}} = \frac{1}{2\omega_{\mathbf{q}\nu}} \int d\mathbf{r} d\mathbf{r}' g_{-\mathbf{q}\nu}(\mathbf{r}) \chi^r(\mathbf{r}, \mathbf{r}'; \omega_{\mathbf{q}\nu}) g_{\mathbf{q}\nu}(\mathbf{r}') - \frac{1}{2\omega_{\mathbf{q}\nu}} \int d\mathbf{r} d\mathbf{r}' g_{-\mathbf{q}\nu}(\mathbf{r}) \chi^r(\mathbf{r}, \mathbf{r}'; 0) g_{\mathbf{q}\nu}(\mathbf{r}'). \quad (16)$$

In Eq. (15) we introduced the Born-Oppenheimer (harmonic) phonon frequencies $\omega_{\mathbf{q}\nu}$, which are related to the elastic tensor $K_{\mathbf{q}\nu\nu'}$ via

$$\omega_{\mathbf{q}\nu}^2 = K_{\mathbf{q}\nu}\delta_{\nu\nu'} + 2\omega_{\mathbf{q}\nu}\text{Re}\Pi_{\mathbf{q}\nu}^{\text{ep,A}}, \quad (17)$$

where the adiabatic phonon self-energy $\Pi_{\mathbf{q}\nu}^{\text{ep,A}}$ is defined as

$$\Pi_{\mathbf{q}\nu}^{\text{ep,A}} = \frac{1}{2\omega_{\mathbf{q}\nu}} \int d\mathbf{r}d\mathbf{r}' g_{-\mathbf{q}\nu}(\mathbf{r})\chi'(\mathbf{r}, \mathbf{r}'; 0)g_{\mathbf{q}\nu}(\mathbf{r}'). \quad (18)$$

Equations (13)–(16) are the central result of our work, and their detailed derivation is reported in the Supplemental Material (SM) [40]. These expressions formally relate the decoherence rate of coherent phonons $\Gamma_{\mathbf{q}\nu}$ and their frequency renormalization to the nonadiabatic phonon self-energy, thus justifying the approximate identification of coherent phonon lifetimes with the ordinary phonon lifetimes in the vicinity of equilibrium. The phonon self-energy, Eqs. (17) and (18), here treated within OMSA, can further be expressed within the self-consistent quasiparticle approximation, as in Refs. [34,41,42]. Our formalism is consistent with Ref. [42], where Eqs. (7)–(9) are solved in the thermodynamic limit assuming that phonons are coupled to a continuum of electronic excitations. However, it has also been shown [31] that explicit solutions of Eqs. (7)–(9) can lead to undamped oscillations since in Ref. [31] the spectrum of the electronic system considered is discrete. To avoid ambiguities, we emphasize that our approach is valid for a continuous electronic spectrum. Additionally, the close relation of Eq. (16) to the nonadiabatic phonon self-energy derived within the equilibrium Green's function formalism enables the application of the available implementations for the phonon self-energy to study the decoherence and softening of coherent phonons. In particular, the phonon self-energy introduced above coincides with the expressions reported in nonequilibrium [34] Green's function formulation of the electron-phonon problem, which reduce to the equilibrium Green's function formalism [43] for electronic occupations described by Fermi-Dirac statistics. We note that in Eq. (16) the self-energy is expressed in terms of the reducible function and the bare electron-phonon vertex g . Alternatively, the self-energy can be rewritten in terms of the irreducible response function, involving a bare electron-phonon coupling matrix element $g_{\mathbf{q}\nu}(\mathbf{r})$ and dynamically screened coupling matrix elements [34,37,41,43–45]. Recent investigations showed that it is convenient to recast Eq. (16) into a scheme with two screened electron-phonon vertices for calculations [27,37,44,45]. That discussion is beyond the scope of this paper, and we adopt the double-screened expression to compute the phonon linewidth. A rigorous formal justification of the double-screened expression is not yet fully justified. However, this approach has been widely employed in first-principles calculations of phonon linewidths.

C. Decoherence and frequency renormalization mediated by phonon-phonon coupling

In the following, we extend the formalism developed in Sec. II B to describe the effects of phonon-phonon interactions on the decoherence and renormalization of coherent phonons. Phonon-phonon scattering underpins the temperature dependence of the phonon decoherence, and it constitutes

the leading dissipation mechanisms in crystals with low free-carrier concentrations (semiconductors, insulators, and semimetals) [46–49]. We hereby restrict ourselves to consider third-order anharmonicities in the Born-Oppenheimer approximation. We further neglect fourth- and higher-order terms, which can, however, be considered with an approach similar to that for third-order terms. These approximations can be circumvented within the formalism of the nonequilibrium Green's function theory, which results in a variety of diagrams contributing to the anharmonic self-energy of the phonons [50]. Under the approximations we adopted, the Hamiltonian is the sum of harmonic phonons and the third-order phonon-phonon Hamiltonian:

$$\hat{H} = \frac{1}{2} \sum_{\mathbf{q}\nu} (\hat{P}_{-\mathbf{q}\nu}\hat{P}_{\mathbf{q}\nu} + \omega_{\mathbf{q}\nu}^2 \hat{U}_{-\mathbf{q}\nu}\hat{U}_{\mathbf{q}\nu}) + \frac{1}{3!} \sum_{\mathbf{q}\nu\mathbf{q}'\nu'\mathbf{q}''\nu''} V_{\mathbf{q}\nu,\mathbf{q}'\nu',\mathbf{q}''\nu''}^{(3)} \hat{U}_{\mathbf{q}\nu}\hat{U}_{\mathbf{q}'\nu'}\hat{U}_{\mathbf{q}''\nu''}. \quad (19)$$

$V_{\mathbf{q}\nu,\mathbf{q}'\nu',\mathbf{q}''\nu''}^{(3)}$ denotes the phonon-phonon coupling matrix elements, which involve the third-order derivative of the Born-Oppenheimer potential energy surface relative to the displacements $U_{\mathbf{q}\nu}$. It can be estimated from first-principles via finite-difference calculations based on density-functional theory [51]. The translational invariance of the Hamiltonian requires crystal momentum conservation, $\mathbf{q} + \mathbf{q}' + \mathbf{q}'' = \mathbf{G}$, where $\mathbf{G}$ is a reciprocal lattice vector. To focus on phonon-phonon coupling, we omit terms arising from the electron-phonon interactions, and we note that all corrections are additive at linear order. The coherent-phonon equation of motion of an anharmonic crystal is thus given by

$$\frac{\partial^2 U_{\mathbf{q}\nu}}{\partial t^2} + \omega_{\mathbf{q}\nu}^2 U_{\mathbf{q}\nu} = -\frac{1}{2} \sum_{\mathbf{q}'\nu'\mathbf{q}''\nu''} V_{-\mathbf{q}\nu,\mathbf{q}'\nu',\mathbf{q}''\nu''}^{(3)} \langle \hat{U}_{\mathbf{q}'\nu'}\hat{U}_{\mathbf{q}''\nu''} \rangle. \quad (20)$$

Similar to the case discussed in Sec. II B, Eq. (20) lacks a term proportional to $\partial U_{\mathbf{q}\nu}/\partial t$, which may be promptly identified with the source of decoherence due to anharmonicities. In this case, decoherence is expected to arise from the two-phonon density matrix $\langle \hat{U}_{\mathbf{q}'\nu'}\hat{U}_{\mathbf{q}''\nu''} \rangle$ on the right-hand side of Eq. (20). Besides decoherence, this quantity incorporates the nonlinear phononic driving force [9,16,17], as well as the fluctuation of atomic vibrations (incoherent phonons) [19]. The dynamics of $\langle \hat{U}_{\mathbf{q}'\nu'}\hat{U}_{\mathbf{q}''\nu''} \rangle$ can be determined by generalizing the Heisenberg equation of motion from Eq. (8):

$$\begin{aligned} & \frac{\partial^2 \langle \hat{U}_{\mathbf{q}'\nu'}\hat{U}_{\mathbf{q}''\nu''} \rangle}{\partial t^2} + (\omega_{\mathbf{q}'\nu'}^2 + \omega_{\mathbf{q}''\nu''}^2) \langle \hat{U}_{\mathbf{q}'\nu'}\hat{U}_{\mathbf{q}''\nu''} \rangle \\ &= 2 \langle \hat{P}_{\mathbf{q}'\nu'}\hat{P}_{\mathbf{q}''\nu''} \rangle - \frac{1}{2} \sum_{1,2} V_{-\mathbf{q}'\nu',\mathbf{q}_1\nu_1\mathbf{q}_2\nu_2}^{(3)} \langle \hat{U}_{\mathbf{q}_1\nu_1}\hat{U}_{\mathbf{q}_2\nu_2}\hat{U}_{\mathbf{q}''\nu''} \rangle \\ & - \frac{1}{2} \sum_{1,2} V_{-\mathbf{q}''\nu'',\mathbf{q}_1\nu_1\mathbf{q}_2\nu_2}^{(3)} \langle \hat{U}_{\mathbf{q}_1\nu_1}\hat{U}_{\mathbf{q}_2\nu_2}\hat{U}_{\mathbf{q}'\nu'} \rangle, \end{aligned} \quad (21)$$

where the right-hand side depends on two-momentum $\langle \hat{P}_{\mathbf{q}'\nu'}\hat{P}_{\mathbf{q}''\nu''} \rangle$ and three-phonon correlations. Equations (20) and (21) are the first terms of the Bogoliubov-Born-Green-Kirkwood-Yvon hierarchy, which recast the equation of motion for the n th-order correlator in terms of correlators

of order $n + 1$. In order to close the equation, we can derive the equation of motion for $\langle \hat{P}_{\mathbf{q}'\nu'} \hat{P}_{\mathbf{q}''\nu''} \rangle$ and truncate the three-phonon correlations into the product of a two-phonon correlation and the coherent phonon $\langle \hat{U}_{\mathbf{q}\nu} \rangle$. To close the equation of motion for $\langle \hat{U}_{\mathbf{q}'\nu'} \hat{U}_{\mathbf{q}''\nu''} \rangle$ and $\langle \hat{P}_{\mathbf{q}'\nu'} \hat{P}_{\mathbf{q}''\nu''} \rangle$ we make use of the momentum conservation ($\mathbf{q}_1 + \mathbf{q}_2 = \mathbf{G} + \mathbf{q} - \mathbf{q}''$), and we neglect nondiagonal terms contributing to the correlation function. This is equivalent to considering only diagonal self-energy contributions. After some algebra, the coherent-phonon equation of motion can be recast in a form identical to Eq. (13), with the decoherence rate and frequency renormalization given by

$$\Gamma_{\mathbf{q}\nu}^{\text{pp}} = -\text{Im}\Pi_{\mathbf{q}\nu}^{\text{pp}}, \quad (22)$$

$$\Omega_{\mathbf{q}\nu}^2 = \omega_{\mathbf{q}\nu}^2 + 2\omega_{\mathbf{q}\nu}\text{Re}\Pi_{\mathbf{q}\nu}^{\text{pp}}. \quad (23)$$

$\Pi_{\mathbf{q}\nu}^{\text{pp}}$ is the lowest-order phonon self-energy due to the phonon-phonon interaction, defined according to

$$\begin{aligned} \Pi_{\mathbf{q}\nu}^{\text{pp}} = & - \sum_{\mathbf{q}'\nu', \mathbf{q}''\nu''} |\Psi_{-\mathbf{q}\nu, \mathbf{q}'\nu', \mathbf{q}''\nu''}|^2 \\ & \times \left[\frac{(\omega_{\mathbf{q}'\nu'} + \omega_{\mathbf{q}''\nu''})(n_{\mathbf{q}'\nu'} + n_{\mathbf{q}''\nu''} + 1)}{(\omega_{\mathbf{q}'\nu'} + \omega_{\mathbf{q}''\nu''})^2 - (\omega_{\mathbf{q}\nu} + i\eta)^2} \right. \\ & \left. + \frac{(\omega_{\mathbf{q}'\nu'} - \omega_{\mathbf{q}''\nu''})(n_{\mathbf{q}''\nu''} - n_{\mathbf{q}'\nu'})}{(\omega_{\mathbf{q}'\nu'} - \omega_{\mathbf{q}''\nu''})^2 - (\omega_{\mathbf{q}\nu} + i\eta)^2} \right], \end{aligned} \quad (24)$$

where we defined phonon-phonon coupling matrix elements as $\Psi_{\mathbf{q}\nu, \mathbf{q}'\nu', \mathbf{q}''\nu''} = \frac{V_{\mathbf{q}\nu, \mathbf{q}'\nu', \mathbf{q}''\nu''}^{(3)}}{\sqrt{2\omega_{\mathbf{q}\nu}2\omega_{\mathbf{q}'\nu'}2\omega_{\mathbf{q}''\nu''}}}$ and $n_{\mathbf{q}\nu}$ denotes the phonon distribution function. After $n_{\mathbf{q}\nu}$ is replaced with the equilibrium Bose-Einstein distribution function, Eq. (24) coincides with the phonon self-energy obtained within the equilibrium Green's function formalization [52–56].

A detailed derivation of this result is reported in the SM [40]. Overall, these results formally relate the decoherence rate and frequency renormalization of coherent phonons induced by anharmonic effects to the real and imaginary parts of the phonon self-energy due to phonon-phonon scattering, respectively, thus providing the theoretical foundation for the investigation of phonon decoherence from first principles.

III. PHONON DECOHERENCE IN ELEMENTAL SEMIMETALS

In elemental semimetals, such as bismuth and antimony, coherent phonons have been comprehensively characterized via pump-probe optical and photoemission experiments [18,33,35,57–59]. These works revealed a strong dependence of the decoherence on temperature [57,58] and pump fluence [18,35], making Sb and Bi an ideal platform for assessing the accuracy of the first-principles approach developed in Sec. II. In the following, we thus proceed to investigate the decoherence rate of coherent phonons in Bi and Sb based on *ab initio* simulations of the electron-phonon and phonon-phonon interactions. All calculations are conducted with the *epw* code [28,60], which is part of QUANTUM ESPRESSO [61] and is interfaced with WANNIER90 [62]. Computational details are reported in the SM [40].

In their low-temperature phase, Bi and Sb crystallize in a rhombohedral structure belonging to space group $R\bar{3}m$ [inset of Fig. 2(a)], which arises from a Peierls distortion along the trigonal axis of the high-symmetry cubic phase. The phonon dispersions of Bi and Sb obtained from density-functional perturbation theory are illustrated in Figs. 2(a) and 2(d), respectively. The A_{1g} mode is the dominant coherent vibrational excitation observed in pump-probe experiments [18,35,59], and it coincides with the Γ -point high-energy optical phonon, marked by a red dot in Figs. 2(a) and 2(d). The coherent excitation of the A_{1g} phonon is induced by the displacive mechanism [35,63,64], which results from the emergence of a photoexcited carrier density coupled to the lattice via electron-phonon coupling. An *ab initio* description of the A_{1g} coherent phonon excitation in Sb via the displacive mechanism was recently reported in Ref. [35]. While the E_g mode [marked by blue dots in Figs. 2(a) and 2(d)] can also undergo coherent excitation, its oscillation amplitude is much smaller than that of the A_{1g} mode [13,65], making an experimental assessment of decoherence more challenging. We thus concentrate below on the decoherence rate of the A_{1g} coherent phonon.

We begin by discussing the decoherence rate due to the electron-phonon interaction. In Figs. 2(b) and 2(e), we report the decoherence rates $\Gamma_{\mathbf{q}\nu}^{\text{FM}}$ of the high-energy optical phonons of Bi and Sb, respectively, for momenta along the T - Γ - K high-symmetry path of the Brillouin zone. $\Gamma_{\mathbf{q}\nu}^{\text{FM}}$ is obtained from Eq. (14) by considering the Fan-Migdal (FM) approximation to the phonon self-energy $\Pi_{\mathbf{q}\nu}^{\text{NA}}$ due to electron-phonon coupling. To explicitly account for the energy transferred to the electrons by a pump pulse, we describe electronic occupations via a Fermi-Dirac distribution with an electronic temperature of 3000 K for Bi and 1450 K for Sb. Here and below, the electronic temperatures are chosen to reproduce the experimental conditions of Refs. [18,35], as discussed in the SM [40]. Figures 2(b) and 2(e) indicate that the decoherence rate $\Gamma_{\mathbf{q}\nu}^{\text{FM}}$ peaks around the Γ point ($\mathbf{q} = 0$). However, as Γ is approached, the decoherence rate undergoes a sharp decrease and vanishes exactly at Γ . Since coherent phonons are excited precisely at Γ , one would expect vanishing linewidths, corresponding to undamped coherent oscillations of the lattice.

The vanishing of $\Gamma_{\mathbf{q}\nu}^{\text{FM}}$, however, is an artifact of the Fan-Migdal approximation, and it arises from the omission of intraband transitions, which provide the dominant contribution to the self-energy for $\mathbf{q} = 0$. This result indicates that the Fan-Migdal approximation is not suitable for capturing phonon decoherence. This problem is well-known in the context of (incoherent) phonon damping, and it has been extensively discussed in the context of Raman linewidths of MgB_2 [23,24,30,66]. A solution to this issue requires accounting for the contribution of intraband transitions to the phonon damping. This can be achieved either via the inclusion of additional self-energy diagrams [66] or via the self-consistent solution of the Dyson equation for the phonon self-energy [67]. While a self-consistent solution of the electron-phonon problem is not computationally feasible, recently, a self-consistent phonon-linewidth approach was proposed for both the electron [67] and phonon [68] self-energies. This approach mimics the one-shot solution of the Dyson equation by accounting exclusively for the change in electron linewidths arising from the imaginary part of the electron self-energy.

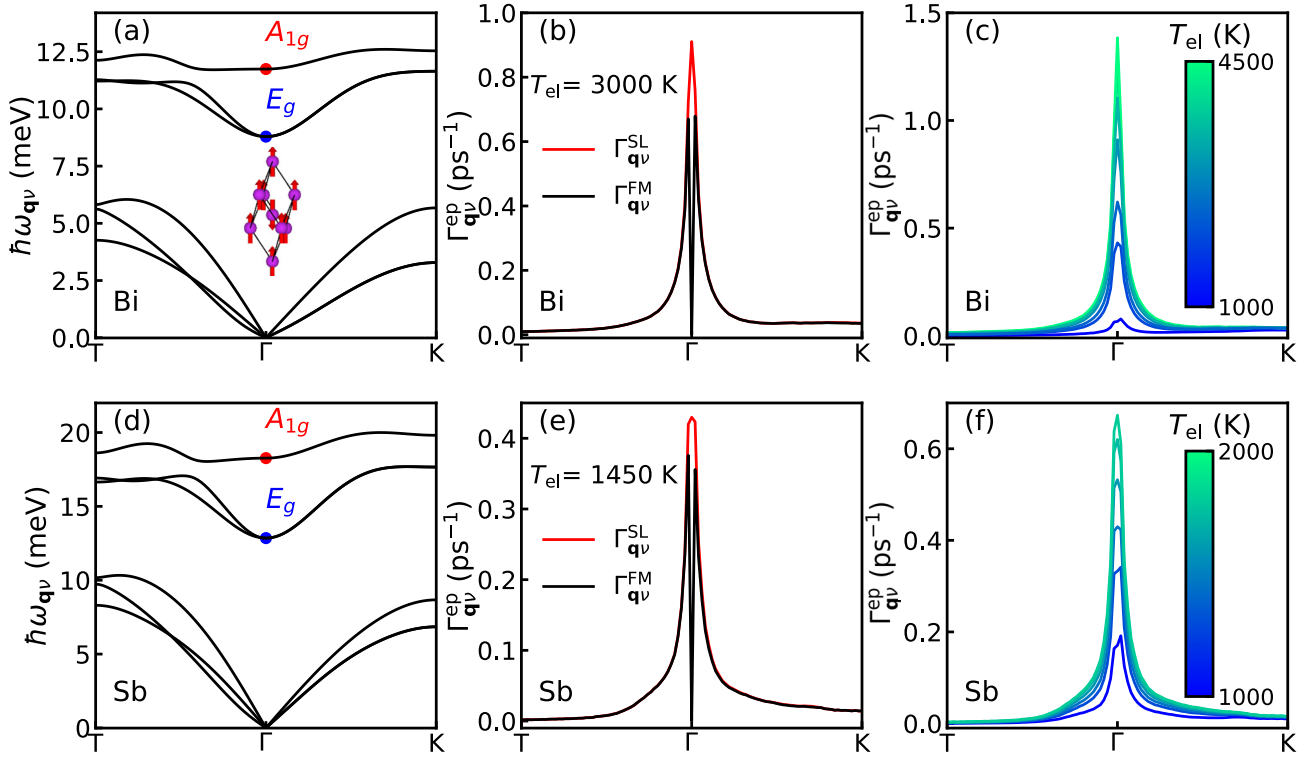


FIG. 2. Phonon dispersion of (a) Bi and (d) Sb along the high-symmetry path T - Γ - K of the BZ. The decoherence rate of the A_{1g} mode at the Γ point along the same high-symmetry path for (b) Bi and (e) Sb with and without the intraband contribution. The electronic temperature is 3000 K for Bi and 1450 K for Sb. The decoherence rate with rising electronic temperature for (c) Bi and (f) Sb, consistent with the fluence of laser pulse employed in the experiments.

It successfully accounts for intraband transitions, and it lifts the unphysical discontinuity of the phonon self-energy as Γ is approached.

In order to extend self-energy calculations to coherent phonon decoherence in semimetals, we implemented the self-consistent linewidth (SL) approximation to the decoherence rate, denoted as $\Gamma_{\mathbf{q}\nu}^{\text{SL}}$ [68]. As illustrated in the SM [40], it can be expressed as

$$\Gamma_{\mathbf{q}\nu}^{\text{SL}} = \pi \sum_{m\mathbf{k}} |g_{m\nu}^s(\mathbf{k}, \mathbf{q})|^2 [f(\varepsilon_{n\mathbf{k}}) - f(\varepsilon_{n\mathbf{k}} + \omega_{\mathbf{q}\nu})] \times \delta_\gamma(\varepsilon_{m\mathbf{k}+\mathbf{q}} - \varepsilon_{n\mathbf{k}} - \omega_{\mathbf{q}\nu}), \quad (25)$$

where the electron-phonon coupling matrix element obtained from DFPT calculations $g_{m\nu}^s(\mathbf{k}, \mathbf{q}) = \sqrt{\frac{1}{2\omega_{\mathbf{q}\nu}}} \langle m\mathbf{k} + \mathbf{q} | \partial_{\mathbf{q}\nu} V^{\text{scf}} | n\mathbf{k} \rangle$ is screened by Coulomb and exchange-correlation potentials. $f(\varepsilon) = \{\exp[(\varepsilon - \mu)/k_B T_{\text{el}}] + 1\}^{-1}$ denotes the Fermi-Dirac function with chemical potential μ . In principle, the phonon linewidth can also be calculated with the nonequilibrated electronic population; we hereby assume that the excited electrons quickly thermalize following laser excitation. δ_γ is a Lorentzian function with width $\gamma = 15$ meV, representative of the electron linewidths in the vicinity of the Fermi surface. In the limit of vanishing electron linewidth ($\gamma \rightarrow 0$), δ_γ reduces to the Dirac δ , and one recovers from Eq. (25) the decoherence rate in the Fan-Migdal approximation $\Gamma_{\mathbf{q}\nu}^{\text{FM}}$ [25,26,28,69] [black line in Figs. 2(b) and 2(e)].

The decoherence rate $\Gamma_{\mathbf{q}\nu}^{\text{SL}}$ of the high-frequency optical phonon of Bi and Sb obtained from the evaluation of Eq. (25) is reported in red in Figs. 2(b) and 2(e) for momenta along the T - Γ - K path. Away from Γ , the decoherence rate $\Gamma_{\mathbf{q}\nu}^{\text{SL}}$ coincides with the result of the Fan-Migdal approximation $\Gamma_{\mathbf{q}\nu}^{\text{FM}}$; however, the two approximations differ significantly at the zone center. Specifically, $\Gamma_{\mathbf{q}\nu}^{\text{SL}}$ lifts the unphysical discontinuity at Γ , leading to finite decoherence rates. This is critical for investigating the dissipation of coherent phonons induced by electron-phonon coupling, and it enables us to estimate the decoherence rate of the A_{1g} mode of Bi (Sb) to 0.9 ps^{-1} (0.4 ps^{-1}) for an effective electronic temperature of $T_{\text{el}} = 3000 \text{ K}$ (1450 K). To investigate the influence of the electronic excitation on phonon decoherence, we report in Figs. 2(c) and 2(f) the decoherence rate $\Gamma_{\mathbf{q}\nu}^{\text{SL}}$ for electronic temperatures ranging up to 2000 and 4500 K. We observe a significant increase of the decoherence rate with increasing electronic temperature. This behavior can be attributed to the larger phase space available for phonon-assisted electronic transitions at large electronic temperatures, giving rise to faster decoherence.

The dependence of the calculated coherent phonon lifetime $\tau_{A_{1g}}^{\text{SL}} = (\Gamma_{A_{1g}}^{\text{SL}})^{-1}$ for the A_{1g} mode on the effective electronic temperature T_{el} is illustrated in Figs. 3(a) and 3(b) (orange diamonds). For comparison, we report the experimental lifetime (red circles) extracted from fluence-dependent pump-probe optical and photoemission experiments of Bi and Sb, respectively [18,35]. The fluence is expressed in terms of the

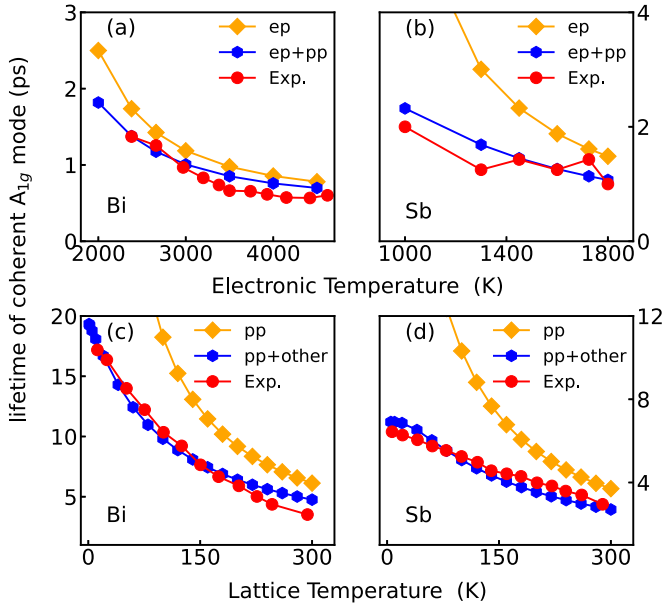


FIG. 3. Coherent phonon lifetime of the A_{1g} mode for Bi and Sb as a function of (a) and (b) electronic temperature and (c) and (d) lattice temperature. The experimental data are taken from Refs. [18,57] for Bi and Refs. [35,58] for Sb.

effective electronic temperature according to the prescription outlined in the SM. The dependence of the lifetime $\tau_{A_{1g}}^{\text{SL}}$ on T_{el} is well approximated by a linear relation of the form $\tau^{\text{SL}} = \alpha T_{\text{el}}^{-1}$, with $\alpha \sim 2.6$ and $\sim 1.6 \text{ ns K}$ for Bi and Sb, respectively. Overall, by accounting for the electron-phonon interactions our simulations capture the decrease of the coherent phonon lifetime as a function of the electronic temperature (fluence) observed in experiments. These findings provide strong evidence that electron-phonon coupling is the primary mechanism behind the fluence-dependent decoherence. However, calculations based on Eq. (25) alone systematically overestimate the experimental lifetime. Inspection of the decoherence rate (Fig. S1 in the SM [40]) reveals underestimation by a constant offset, suggesting the presence of additional interaction mechanisms—independent of T_{el} —contributing to the decoherence rate. As discussed below, this discrepancy can be partially resolved by including the effects of phonon-phonon interactions, which introduce an additional damping channel beyond electron-phonon coupling.

The contribution of phonon-phonon scattering to the decoherence rate $\Gamma_{\text{qv}}^{\text{pp}}$ can be accounted for through Eqs. (22) and (24), leading to

$$\Gamma_{\text{qv}}^{\text{pp}} = \frac{\pi}{2} \sum_{\mathbf{q}'\mathbf{q}''\mathbf{v}'\mathbf{v}''} |\Psi_{-\mathbf{q},\mathbf{q}',\mathbf{q}''}^{\mathbf{v},\mathbf{v}',\mathbf{v}''}|^2 \times [(n_{\mathbf{q}'\mathbf{v}'} + n_{\mathbf{q}''\mathbf{v}''} + 1)\delta(\omega_{\mathbf{q}'\mathbf{v}'} + \omega_{\mathbf{q}''\mathbf{v}''} - \omega_{\mathbf{qv}}) + 2(n_{\mathbf{q}'\mathbf{v}''} - n_{\mathbf{q}'\mathbf{v}'})\delta(\omega_{\mathbf{q}'\mathbf{v}'} - \omega_{\mathbf{q}''\mathbf{v}''} - \omega_{\mathbf{qv}})]. \quad (26)$$

Numerical evaluation of $\Gamma_{A_{1g}}^{\text{pp}}$ at room temperature ($T_{\text{ph}} = 300 \text{ K}$) for the A_{1g} mode of Sb and Bi yields 0.26 and 0.15 ps^{-1} , respectively. According to Matthiessen's rule, the

total lifetime $\tau_{A_{1g}}^{\text{ep+pp}}$ can be obtained by adding the contributions due to electron-phonon and phonon-phonon scattering: $\tau_{A_{1g}}^{\text{ep+pp}} = [\Gamma_{A_{1g}}^{\text{SL}}(T_{\text{el}}) + \Gamma_{A_{1g}}^{\text{pp}}(T_{\text{ph}})]^{-1}$. Since $\Gamma_{A_{1g}}^{\text{pp}}$ does not depend on the electronic occupations f_{nk} , we neglect its dependence on T_{el} . This approximation omits possible changes in the phonon distribution n_{qv} induced by electronic excitations [70–74] and their effects on the phonon-phonon decoherence rate. The total lifetime $\tau_{A_{1g}}^{\text{ep+pp}}$, marked by blue hexagons in Figs. 3(a) and 3(b), reduces the discrepancy between experiments and theory, indicating that both electron-phonon scattering and phonon scattering can contribute to decoherence.

To illustrate the influence of phonon-phonon interaction on decoherence, we report in Figs. 3(c) and 3(d) the quantity $\tau_{A_{1g}}^{\text{pp}}$ for lattice temperatures T_{ph} ranging between 0 and 300 K (orange diamonds). The temperature-dependent coherent phonon lifetime extracted from pump-probe experiments at constant fluence are shown as red circles [57,58]. Our calculations accurately reproduce the decrease of lifetime with lattice temperature, in agreement with experiments. The temperature dependence is well captured by the fitting function $\tau_{A_{1g}}^{\text{pp}} \simeq \tau_0^{\text{pp}}[2n(T_{\text{ph}}) + 1]^{-1}$, where $n(T_{\text{ph}})$ is the Bose-Einstein distribution, which accounts for the increasing phonon population with temperature. After an initial nonlinear transient at low temperatures, $n(T_{\text{ph}})$ varies linearly with T_{ph} , and the lifetime is well described by the fitting function $\tau_{A_{1g}}^{\text{pp}} = [\Gamma_0 + \alpha_{\text{anh}}T_{\text{ph}}]^{-1}$, with $\alpha_{\text{anh}} = 5.0 \times 10^{-4}$ for Bi and $\alpha_{\text{anh}} = 7.6 \times 10^{-4}$ for Sb.

Simulations of $\Gamma_{A_{1g}}^{\text{pp}}$ underestimate the experiments by a constant offset, as shown in Fig. S1 in the SM [40], indicating the presence of additional contributions to the decoherence rates that do not depend significantly on the lattice temperature. These effects were attributed to electron-phonon and impurity scattering, and they were estimated to be 0.05 ps^{-1} [57] for Bi and 0.1 ps^{-1} for Sb [58]. To account for these contributions, we introduce an effective constant decoherence rate $\Gamma_{A_{1g}}^{\text{eff}}$, leading to a total decoherence rate $\Gamma_{A_{1g}}^{\text{tot}} = \Gamma_{A_{1g}}^{\text{pp}} + \Gamma_{A_{1g}}^{\text{eff}}$. The total lifetime $\tau_{A_{1g}}^{\text{tot}}$ is reported in Figs. 3(c) and 3(d) (blue hexagons), and its improved agreement with experiments provides a robust interpretation of the residual discrepancy. Overall, these findings provide strong evidence that the temperature dependence of decoherence is governed by phonon-phonon scattering.

IV. CONCLUSIONS

In conclusion, we investigated the process of phonon decoherence in crystalline solids on the basis of a first-principles many-body framework. Starting from a many-body formulation of the lattice equation of motion, we obtained explicit expressions for the decoherence rate and frequency renormalization of coherent phonons arising from electron-phonon and phonon-phonon coupling. These results rigorously link the process of phonon decoherence to the phonon self-energy. We validated this approach by conducting first-principles calculations of phonon decoherence induced by electron-phonon and phonon-phonon interactions. Specifically, we estimated the fluence and temperature dependence of the phonon de-

coherence time for the elemental semimetals Bi and Sb. The good quantitative agreement with recent pump-probe experimental data corroborates the theoretical approach introduced here. These findings further indicate that far from equilibrium both electron-phonon scattering and phonon-phonon scattering can contribute substantially to the process of phonon decoherence. Besides advancing the fundamental understanding of the nonequilibrium lattice dynamics, these results constitute the basis to understand and assess the characteristic timescales for phonon decoherence in driven solids, with implications for light-induced structural control and phase transitions.

Note added. Recently, we became aware of Ref. [42], where Eqs. (16)–(18) were independently derived.

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DATA AVAILABILITY

The data that support the findings of this article are openly available [75].

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