

Accurate Determination of Phonon Lifetimes from the Spectral Domain

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Concerningly, numerous incorrect expressions for extracting phonon lifetimes from spectroscopic linewidths remain prevalent in the literature, resulting in misinterpretation of experimental data. This has significant consequences in the broader energy materials field, where erroneous lifetimes risk misinforming materials design and device modeling. We outline the origin of this ambiguity, provide historical context on how this error has propagated, and present the correct formula. In addition, we highlight a frequently overlooked source of error: the contribution of phase relaxation to spectral broadening. A unified approach enables fair comparison of values obtained from different spectroscopic methods, accurate benchmarking against first-principles calculations, and, most importantly, precise determination of macroscopic physical properties derived from phonon lifetime measurements.

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I. INTRODUCTION

Many fundamental properties of solids are governed by phonons, the quantized vibrations of a crystal lattice. The phonon dispersion relation dictates the elastic response of a material and, together with phonon lifetimes, determines the thermal conductivity, the control of which is critical for achieving high-figure-of-merit thermoelectric devices [1,2]. In semiconductors, electron-phonon coupling controls charge-carrier mobility and sets the defect capture cross sections that drive Shockley-Read-Hall recombination [3,4]. Nonradiative recombination lowers the open-circuit voltage and low charge-carrier mobility hinders their extraction with both processes being central to photovoltaic performance. Phonons also play a key role in determining ionic migration pathways and transport activation energies in solid electrolytes; thus, understanding these processes is essential for developing next-generation battery materials [5]. Reliable and accurate procedures for measuring phonon lifetimes are therefore crucial for

advancing energy materials research, including photovoltaics, thermoelectrics, and batteries, and for preventing the spread of errors into other fields in which phonons fundamentally govern macroscopic physical properties of importance.

The widespread use of incorrect spectral-domain formulas within the inelastic neutron scattering community has caused lifetime estimates for coherent excitations (phonons) to differ from the correct values by factors of 2 or π , depending on the formulas chosen. This error is present in the formulas that are derived under the assumption that phonons scatter only inelastically, i.e., when their population decays through energy relaxation. However, it is often overlooked that spectral broadening can also occur in the absence of energy relaxation, when elastic scattering disrupts the phase coherence of the oscillators. This leads to an additional contribution to the linewidth, known as pure dephasing. Pure dephasing processes (with lifetime T_ϕ) randomize the phase of the population of a phonon state without affecting its occupation number [6]. This mechanism is distinct from energy relaxation, the characteristic lifetime of which is conventionally defined as the phonon lifetime (τ_p).

In this paper, we consider only the contributions to spectral linewidths arising from homogeneous broadening, which occurs when all elements of the system exhibit a uniform response. Both energy relaxation and pure dephasing contribute to homogeneous broadening. In real

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materials, yet, variations in the local atomic environment, such as those introduced by, e.g., defects, strain, and compositional fluctuations, give rise to inhomogeneous broadening. This manifests as Gaussian components in the spectral line shapes, which are convolved with the Lorentzian components originating from homogeneous broadening. Given the multiple inconsistent approaches found in the literature, this paper aims to provide a clear explanation of the correct approach.

A. The historical origin of the incorrect phonon lifetime expression

When discussing phonon lifetimes in the spectral domain, references are frequently made to the seminal work of Klemens [7]. Klemens inherits the expression from the lifetime formulation of emitted radiation in laser physics, as originally presented by Heitler and Demtroder [8,9]. In this formulation, only population relaxation has been considered to contribute to the spectral response and the phase relaxation term has been omitted. The widespread overestimation of phonon lifetimes, commonly reported as being twice the actual value, originates from later misinterpretation of terminology used in Klemens's 1966 paper [7]. Klemens cites Heitler's 1936 work [8] and refers to the spectral broadening factor as the "half-width." This term was later misinterpreted as the half width at half maximum (HWHM), while in actual fact, Klemens's original definition corresponds to the full width at half maximum (FWHM). Nilsson [10] later acknowledged the confusion in the literature but ironically adopted the wrong interpretation of the phonon lifetimes, using the erroneous expression of HWHM. Nilsson's work [10] remains highly cited and, as a result, much of the modern inelastic neutron scattering community, particularly in the field of energy materials, continues to use this erroneous expression. By contrast, communities utilizing other spectroscopic techniques such as inelastic x-ray scattering, Raman (with the exception in Ref. [11]), Brillouin scattering, and infrared spectroscopy have largely retained the correct formulation. Thus, in this paper we derive the correct expression for phonon lifetimes as originally identified by Klemens and clarify the nuances that have led to misinterpretations in the literature.

B. The derivation of phonon lifetime expression

Consider a phonon excitation in the system as a damped linear harmonic oscillator (DHO) with a damping constant γ and a natural (resonant) angular frequency ω_0 . The equation of motion is a linear ordinary differential equation:

$$\ddot{x} + \gamma\dot{x} + \omega_0^2 x = 0. \quad (1)$$

In the case of underdamped oscillations when $\gamma \ll \omega_0$ and under initial conditions (the detailed derivation is provided

in Sec. i of the Supplemental Material [12]), the solution for the elongation of the oscillator becomes

$$x(t) = x_0 e^{-\gamma t/2} \cos(\omega_0 t). \quad (2)$$

Our interest lies in how the energy of the system relaxes over time. Therefore, we can calculate the total energy of the oscillator, which is the summation of the potential and kinetic energy, by inserting $x(t)$ in the equation

$$\langle W(t) \rangle = \left\langle \frac{1}{2} m (\dot{x}^2 + \omega_0^2 x^2) \right\rangle = W_0 e^{-\gamma t}, \quad (3)$$

where $W_0 = \frac{1}{2} m x_0^2 (\omega_0^2 + \gamma^2/8)$ (the full derivation is given in Sec. ii of the Supplemental Material [12]). Owing to the damping factor, energy will be dissipated from the system, which, in the case of a phonon gas, is due to various inelastic phonon scattering mechanisms. The time needed for the energy to decrease to $1/e$ of its initial value is $\tau_p = 1/\gamma$, thereby defining the phonon lifetime. For a DHO defined by Eq. (2), we can also identify the amplitude lifetime (T_2), i.e., the time required for the asymptotic exponential amplitude to decrease to $1/e$ of its initial value. This lifetime is $2/\gamma$, twice the value of the energy relaxation lifetime (the phonon lifetime, τ_p). This relationship is illustrated in Fig. 1(a). The phonon lifetime is typically experimentally measured in the spectral domain using Brillouin, Raman, inelastic x-ray, helium, or neutron scattering techniques. Relating the phonon lifetime, a time-domain property, to the linewidth of a spectral feature requires the use of a Fourier transform. As a result, what is empirically observed is the power spectral density [Eq. (5)], which corresponds to the squared magnitude of the Fourier transform of the elongation expression for the DHO, given in Eq. (2) ($x(t)$). A similar derivation applies to the lifetime of light radiation [8]. The Fourier transform of $x(t)$ is (a detailed derivation is given in Sec. iii of the Supplemental Material [12])

$$\begin{aligned} x(\omega) &= \frac{1}{\sqrt{2\pi}} \int_0^{+\infty} x(t) e^{-i\omega t} dt \\ &= \frac{1}{2\sqrt{2\pi}} x_0 \left(\frac{1}{i(\omega - \omega_0) + \gamma/2} + \frac{1}{i(\omega + \omega_0) + \gamma/2} \right). \end{aligned} \quad (4)$$

The power spectral density is then given by

$$I(\omega) = |x(\omega)|^2 \simeq I_0 \frac{1}{(\omega - \omega_0)^2 + (\gamma/2)^2}, \quad I_0 = \frac{x_0^2}{4\pi}. \quad (5)$$

As the phonon lifetime τ_p is defined as $1/\gamma$, it is necessary to determine γ from the experimentally measured intensity distribution $I(\omega)$. It can be readily demonstrated that the

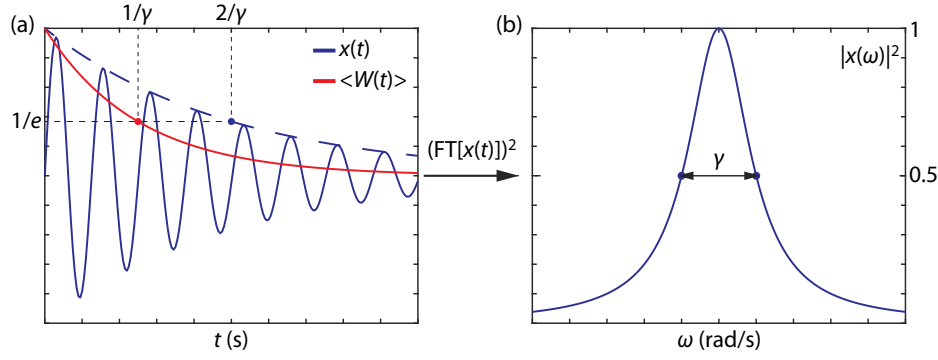


FIG. 1. An illustration of the time and frequency domains of a DHO. (a) $x(t)$ and $W(t)$ represent the amplitude and energy signals of a DHO in the time domain (solid lines). The dashed blue line shows the envelope of $x(t)$. The signals $x(t)$ and $W(t)$ decay to $1/e$ of their initial values at $t = 2/\gamma$ and $t = 1/\gamma$, which are conventionally defined as the amplitude and energy relaxation lifetimes of a DHO, respectively. It is the energy relaxation lifetime that is conventionally defined as the phonon lifetime (τ_p). (b) The spectral response of a DHO, $|x(\omega)|^2$, follows a Lorentzian function, normalized here to 1. The dots indicate points at which $y = |x(\omega)|^2 = 0.5$. The separation between the two corresponding ω values that satisfy this condition is equal to γ , which represents the full width at half maximum (FWHM) of the function.

FWHM of $I(\omega)$ satisfies $\text{FWHM}(I(\omega)) = \gamma$ [Fig. 1(b)], from which it clearly follows that

$$\tau_p = \frac{1}{\gamma} = \frac{1}{\text{FWHM}}. \quad (6)$$

If spectra are measured in frequency rather than angular frequency, the phonon lifetime is given by $\tau_p = 1/(2\pi \text{ FWHM [1/s]})$. However, if spectra are measured in the energy domain, as is often the case in inelastic neutron or x-ray scattering, the relevant expression becomes $\tau_p = \hbar/(\text{FWHM [J]})$. In Table I, we outline the correct lifetime expressions depending on the domain in which the spectral response is measured. Our derived expressions align

TABLE I. Correct versus incorrect phonon lifetime relations (commonly present in the literature) across spectral domains. $\delta\omega$, $\delta\nu$, δE , and $\delta\tilde{\nu}$ correspond to the FWHM of Lorentzians in the angular frequency, frequency, energy, and wavenumber domain, respectively. c is the speed of light. Note that the “Correct formulas” column presents phonon lifetime expressions under the assumption that elastic scattering between phonons is absent. If elastic scattering is included, the equality sign ($=$) in the expression should be replaced by an inequality ($\geq$).

Domain (units)	Correct formulas	Incorrect formulas
Angular frequency ω (rad/s)	$\tau_p = \frac{1}{\delta\omega}$	$\tau_p = \frac{2}{\delta\omega}$
Frequency ν (Hz)	$\tau_p = \frac{1}{2\pi\delta\nu}$	$\tau_p = \frac{1}{\pi\delta\nu}, \tau_p = \frac{1}{2\delta\nu}$
Energy E (J)	$\tau_p = \frac{\hbar}{\delta E}$	$\tau_p = \frac{2\hbar}{\delta E}, \tau_p = \frac{\hbar}{\pi\delta E}$
Wavenumber $\tilde{\nu}$ (m^{-1})	$\tau_p = \frac{1}{2\pi c\delta\tilde{\nu}}$	$\tau_p = \frac{2}{c\delta\tilde{\nu}}$

with those formulated for the lifetime of emitted radiation in laser physics, as referenced in the works of Heitler and Demtroder [8,9]. It is important to note that these expressions are valid only under the assumption that there is no elastic scattering between the phonon and the surrounding thermal bath. This implies that the phonon phase does not undergo stochastic fluctuations over time. However, this assumption is generally invalid at room temperature, where the thermal bath is significantly populated. Elastic scattering processes of this kind are governed by fourth- or higher-order terms in the Hamiltonian of the system [6].

Modern spectroscopic studies, based largely on inelastic neutron scattering, often use an erroneous expression for the phonon lifetime. These studies, focusing on energy-related materials, predominantly halide perovskites [13–18], thermoelectrics [19,20], and certain battery materials [21], mistakenly use the phonon amplitude lifetime instead of the phonon energy lifetime. This misinterpretation has resulted in the erroneous formula $\tau_p = \hbar/(\text{HWHM [J]})$ becoming widespread in modern literature. Clearly, using the HWHM in the denominator rather than the FWHM introduces a factor-of-2 difference in the estimated values, leading to a doubling of measured phonon lifetimes. This directly implies that influence of phonon lifetimes on macroscopic properties of importance for energy materials, such as thermal conductivity or charge-carrier mobility, would be overestimated.

We have identified that, when other techniques are used that probe phonons in spectral domain, such as Raman, Brillouin, and helium inelastic scattering, the correct expression (under the assumption of no dephasing contributions) is typically being used [22,23]. Additional care should be taken when estimating phonon lifetimes computationally from first principles. A common approach

involves utilizing many-body perturbation theory to calculate the Green's function for a particular phonon mode. This method inherently captures anharmonic interactions and higher-order phonon scattering, typically derived from first principles by computing force constants up to a certain order. The amplitude lifetime is inversely proportional to the imaginary part (Γ) of the phonon self-energy (a complex term in the Green's function), expressed in the angular frequency domain. To ensure a fair comparison with experiments, the phonon energy lifetime should be calculated instead. The correct phonon energy lifetime is given by

$$\tau_p = \frac{1}{2\Gamma}, \quad (7)$$

as established by Maradudin and Fein [24] (for the full derivation, see Sec. iv of the Supplemental Material [12]). This expression has been correctly implemented in most computational packages that rely on density-functional-theory (DFT) calculations [25]. This computational approach operates under the assumption that pure dephasing is absent, which is equivalent to neglecting fourth- and higher-order terms in the Hamiltonian.

C. Pure dephasing contribution to spectral broadening

So far, the derived phonon lifetime expressions have considered only inelastic scattering processes between phonons, excluding elastic collisions. Inelastic scattering leads to energy loss in the phonon system and thus to population decay. In systems at finite temperatures, however, population decay is not the sole mechanism contributing to spectral broadening. In the case of elastic phonon-phonon scattering, there is no population decay but the collisions induce phase changes, or so-called dephasing, which also leads to spectral broadening [6,26,27].

The dephasing contribution to spectral broadening can be derived using a classical treatment of stochastic processes, as originally shown by Kubo [28,29], who built on the Anderson-Weiss model of random frequency modulation [30]. In Kubo's formulation, frequency noise plays a central role in line broadening, particularly in magnetic resonance phenomena such as nuclear magnetic resonance (NMR) [28]. In the presence of elastic scattering, the phase coherence of a DHO is disrupted. This can be modeled by modifying Eq. (2) to include a stochastic frequency fluctuation term $\zeta(t)$, which captures random shifts in the resonance frequency induced by elastic collisions:

$$\ddot{x} + \gamma\dot{x} + (\omega_0 + \zeta(t))^2 x = 0. \quad (8)$$

If the frequency noise $\zeta(t)$ is Gaussian white with spectral power $2\Gamma_\phi$ and satisfies the condition $2\Gamma_\phi < \omega_0$ (the weak-noise limit), then, under the assumption that fluctuations are fast compared to the oscillator frequency, Eq. (8)

reduces to a Langevin equation. The ensemble-averaged solution for the displacement is then given by

$$\langle x(t) \rangle = x_0 e^{-(\gamma_1/2 + \Gamma_\phi)t} = x_0 e^{-\gamma_2 t}. \quad (9)$$

Fourier transforming $\langle x(t) \rangle$ using the same method as in Eqs. (4) and (5) yields

$$T_2 = 1/\text{HWHM}, \quad (10)$$

where HWHM is the half width at half maximum of the resulting Lorentzian line shape. The total damping rate $\gamma_2 = \gamma_1/2 + \Gamma_\phi$ leads to the standard expression

$$\frac{1}{T_2} = \frac{1}{2T_1} + \frac{1}{T_\phi}, \quad (11)$$

where T_1 is the phonon energy relaxation lifetime [denoted τ_p in Eqs. (5) and (7)], T_ϕ is the pure dephasing time (associated with phase randomization without energy exchange), and T_2 is the total (amplitude) lifetime, governing the decay of the oscillation amplitude.

In the absence of pure dephasing, i.e., when $T_\phi \rightarrow \infty$, the second term vanishes and one obtains the lifetime-limited case:

$$\frac{1}{T_2} = \frac{1}{2T_1} \implies T_2 = 2T_1 = 2\tau_p. \quad (12)$$

This result demonstrates that when phase scrambling is negligible, the amplitude relaxation time is twice the energy relaxation time, as shown in Fig. 1, based on the classical DHO model. When, however, random phase fluctuations cannot be neglected, extracting the true phonon energy relaxation lifetime τ_p from spectroscopic data becomes nontrivial. The measured linewidth reflects both energy relaxation and dephasing contributions, such that only a lower bound on τ_p can be inferred:

$$T_1 = \tau_p \geq \frac{T_2}{2}. \quad (13)$$

Depending on the ratio T_ϕ/T_1 in Eq. (11), different regimes emerge. When $T_\phi = 2T_1$, the amplitude and energy relaxation times are equal, $T_2 = T_1$. In the case of strong dephasing ($T_\phi < 2T_1$), the amplitude decays faster than the energy, $T_2 < T_1$. In contrast, for weak dephasing ($\infty \geq T_\phi > 2T_1$), the total amplitude lifetime lies between the energy and twice the energy relaxation lifetime, i.e., $T_1 < T_2 < 2T_1$.

In the most general case, spectroscopic measurements yield the amplitude relaxation lifetime, extracted from the HWHM of a Lorentzian line shape ($T_2 = 1/\text{HWHM}$). This amplitude lifetime equals twice the energy relaxation lifetime only when phase relaxation (pure dephasing) is absent. When relying solely on the spectral linewidth,

the energy relaxation time may be either underestimated or overestimated, depending on the relative strength of pure dephasing processes compared to energy relaxation. Decoupling pure dephasing from energy relaxation is, with careful experiment design, possible in time-domain measurements with two-dimensional ultrafast spectroscopes [31].

Equation (11) can also be derived using a quantum mechanical formalism, where the pure dephasing term arises from the decay of off-diagonal elements of the density matrix, while the population decay corresponds to the decay of the diagonal elements. This result follows from the optical Bloch equations [26]. The resulting expression is identical to the classical one given in Eq. (11) and is widely used in the quantum information field to describe coherence times of single qubits. It is also well established in NMR, where it describes longitudinal and transverse magnetic relaxation times.

In computational approaches based on the Green's function formalism, the dephasing term is effectively accounted for by extending the Maradudin and Fein [24] self-energy expression to include quartic (four-phonon) interactions. These predominantly capture elastic scattering processes that contribute to spectral broadening induced by pure dephasing processes. Recent studies have highlighted that four-phonon scattering plays a crucial role in describing the additional broadening of phonon spectral line shapes necessary to adequately explain experimental observations [32–34]. This further underscores the importance of recognizing the presence of pure dephasing contributions when interpreting experimental linewidths.

II. CONCLUSIONS

In conclusion, we have identified several factors contributing to the systematic miscalculation of phonon lifetimes. The first stems from historical context and a common misunderstanding of the distinction between amplitude and energy lifetimes and which of these enters the equations that govern macroscopic properties relevant to applications. The second issue arises from classical derivations of phonon lifetimes and spectral linewidths, which often omit elastic scattering processes that can substantially affect spectral broadening through dephasing. In practice, spectroscopic techniques measure amplitude lifetimes, as these are directly accessible from linewidths. However, isolating the contribution of energy relaxation requires additional analysis to separate it from pure dephasing. A further layer of complexity appears in systems with static disorder, where inhomogeneous broadening introduces Gaussian components into the spectral line shape. Thus, when macroscopic properties such as thermal conductivity, charge-carrier mobility, or other quantities are inferred from experimental spectroscopic observations, it is essential first to determine whether the energy

relaxation lifetime ($T_1 = \tau_p$) or the amplitude relaxation lifetime (T_2) enters the governing equations and then to carefully interpret the spectroscopic linewidth to extract the appropriate lifetime.

We hope that this systematic derivation and clarification of the meaning of phonon lifetime, along with this precise identification of the origins of the incorrect expression and its continued use, will lead to consistent reporting of phonon lifetimes in the future, not only in the energy materials community but also across all spectroscopic disciplines. This, in turn, will enable more reliable predictions of macroscopic material properties.

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M.D. identified the presence of erroneous expressions in the literature and wrote the original version of the manuscript. A.P., M.P.N., and S.D.S. supervised the project, discussed the findings, and contributed to reviewing and editing of the manuscript.

DATA AVAILABILITY

No data were created or analyzed in this study.

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