Sound waves and flexural mode dynamics in two-dimensional crystals

K. H. Michel,* P. Scuracchio,† and F. M. Peeters‡

Universiteit Antwerpen, Department of Physics, Groenenborgerlaan 171, Be-2020 Antwerpen, Belgium

(Received 24 May 2017; revised manuscript received 17 August 2017; published 5 September 2017)

Starting from a Hamiltonian with anharmonic coupling between in-plane acoustic displacements and out-of-plane (flexural) modes, we derived coupled equations of motion for in-plane displacements correlations and flexural mode density fluctuations. Linear response theory and time-dependent thermal Green's functions techniques are applied in order to obtain different response functions. As external perturbations we allow for stresses and thermal heat sources. The displacement correlations are described by a Dyson equation where the flexural density distribution enters as an additional perturbation. The flexural density distribution satisfies a kinetic equation where the in-plane lattice displacements act as a perturbation. In the hydrodynamic limit this system of coupled equations is at the basis of a unified description of elastic and thermal phenomena, such as isothermal versus adiabatic sound motion and thermal conductivity versus second sound. The general theory is formulated in view of application to graphene, two-dimensional h-BN, and 2H-transition metal dichalcogenides and oxides.

DOI: 10.1103/PhysRevB.96.094302

I. INTRODUCTION

Besides remarkable electronic properties [1,2], two-dimensional (2D) crystals exhibit extraordinary elastic and thermal properties, such as the large intrinsic strength of suspended graphene [3], its high thermal conductivity [4], and its negative thermal expansion [5,6]. From an analytical point of view, the latter two phenomena are closely related to the existence of flexural phonon modes due to out-of-plane displacements (see, e.g., [7]) and the anharmonic coupling of these modes to in-plane displacements. Such a coupling has been originally suggested as a membrane effect [8] in order to explain the negative thermal expansion in layered structures.

More recently, the thermal contraction and expansion in graphene has been studied by *ab initio* density functional theory [9] and by Monte Carlo simulations[10]. Continuum-field-theory–based studies [11] have exploited the topological equivalence [12] between graphene and a crystalline polymerized membrane [13]. The competition between thermal contraction and expansion has been studied by applying the unsymmetrized self-consistent field method to graphene [14] and by using anharmonic lattice dynamics [15,16].

Acoustic phonon lifetimes in freestanding and in strained graphene have been calculated by density functional theory [17,18], and relaxation time analysis has been used to estimate the intrinsic thermal conductivity [18]. Phonon lifetimes were obtained by lattice dynamical methods adapted to the case of 2D crystals [15,16]. In Ref. [15] it has been shown that in a broad temperature range the decay rate of flexural modes is much less affected by umklapp processes than the decay rate of in-plane modes.

Experimental studies have revealed still relatively large values of the thermal conductivity also in supported graphene [19], a result that might be relevant for the evacuation of heat in nanoelectronic devices. Theoretical investigations have predicted an anomalous size-dependent thermal conductivity of graphene ribbons and slabs [20]. A review of experimental and theoretical work on phonon transport in graphene up to 2012 has been given in Ref. [21]. Although many experimental results and their interpretation by theory are still debated, there is consensus that the existence of flexural modes plays a crucial role in determining the intrinsic thermal conductivity in 2D materials [22] and that the solution of a Boltzmann-type transport equation provides the most reliable method for the interpretation of theoretical results [23,24].

Recently, thermal phonon transport and related hydrodynamic phenomena such as Poiseuille flow and second sound have been studied by *ab initio* methods combined with the solution of the Boltzmann transport equation for phonons [25–27]. In particular, it is predicted that in 2D crystals the dominance of momentum-conserving normal processes over momentum-destroying umklapp processes in a large temperature range favors the possible occurrence of second sound in these materials. The competition between normal processes versus umklapp processes as a necessary condition for the existence of second sound in three-dimensional (3D) dielectric solids has been studied by theory [28–30] and confirmed by experiments in solid He^4 [31,32].

A description of hydrodynamic phenomena based on the solution of a Peierls-Boltzmann transport equation is incomplete. As is well known in classical liquids, heat conduction not only changes the sound velocity but, as a consequence of thermal expansion, also gives rise to an undisplaced line in addition to the Brillouin doublet in the spectrum of density oscillations [33]. Likewise, in 3D dielectric solids in the regime of low-frequency and long-wavelength, hydrodynamic phenomena such as heat conduction or second sound appear as additional resonances in the displacement-displacement correlation function [34–36]. Given the peculiarities of lattice dynamics related to the existence of in-plane phonons and flexural modes in 2D crystals, a theoretical study that comprehends acoustic and thermal dynamic phenomena should be in order.

In the present paper we will derive, by microscopic theory, transport equations for in-plane sound waves coupled to the

*ktdm@skynet.be

†pablo.scuracchio@uantwerpen.be

‡francois.peeters@uantwerpen.be

nonequilibrium flexural phonon density distribution. Such equations should be the basis of a unified description of various nonequilibrium thermoelastic and hydrodynamic phenomena such as ordinary (first) sound attenuation, time-dependent thermal conductivity, and second sound.

The content of the paper is the following. In Sec. II we recall first some basics from lattice dynamics of nonprimitive crystals. Taking into account symmetry properties of the planar hexagonal crystal, we construct an anharmonic Hamiltonian where in-plane acoustic lattice displacements are coupled to out-of-plane flexural phonons. To obtain an analytically tractable model where all parameters are specified, we restrict ourselves to nearest-neighbor interactions. The anharmonic interactions include normal and umklapp processes. In Sec. III, we recall some principles of linear response theory and of thermal Green's functions techniques [37,38]. As external perturbations we consider a stress tensor and a temperature source. Next, in Sec. IV, the response function for in-plane lattice displacements is obtained in the form of a Dyson equation where the nonequilibrium flexural density distribution occurs as an inhomogeneous term. In Sec. V we derive a kinetic equation for the nonequilibrium flexural density distribution. This equation has the form of a linearized Peierls-Boltzmann–type integrodifferential equation, where in turn the in-plane displacement correlations enter as an inhomogeneous term. In Sec. VI we discuss the physical meaning of the previous results and establish the link with phenomenological theory. Concluding remarks are made in Sec. VII. Additional information on the mathematical techniques are provided in the Appendix.

II. LATTICE DYNAMICS

We recall some elements of anharmonic lattice dynamics of nonprimitive crystals [39,40] and apply these concepts to 2D or monolayer hexagonal crystals. We will calculate changes of the renormalized flexural mode frequencies by in-plane lattice strains. The crystal consists of N unit cells with position vectors

$$\vec{X}(\vec{n}) = n_1\vec{a}_1 + n_2\vec{a}_2, \tag{1}$$

where $\vec{a}_1$, $\vec{a}_2$ are basis vectors of the hexagonal lattice (see Fig. 1) and n_1,n_2 integers with $\vec{n} = (n_1,n_2)$. Each unit cell contains two atoms labeled by an index $\kappa = \{1,2\}$. The equilibrium position of atom κ in the cell $\vec{n}$ is given by

$$\vec{X}(\vec{n}\kappa) = \vec{X}(\vec{n}) + \vec{r}(\kappa), \tag{2}$$

where $\vec{r}(\kappa)$ specifies the position of atom κ with mass m_κ. Lattice dynamics is formulated in terms of atomic displacements $u_i(\vec{n}\kappa)$ and conjugate momenta $p_i(\vec{n}\kappa)$. The Cartesian indices $i = \{x,y\}$ refer to in-plane displacements and $i = z$ to out-of-plane or flexural displacements. The vibrational Hamiltonian of the crystal is given by $H = K + V$, with kinetic energy

$$K = \sum_{\vec{n},\kappa,i} \frac{p_i^2(\vec{n}\kappa)}{2m_\kappa}, \tag{3}$$

and potential energy V taken as a function of the instantaneous atomic positions $\vec{R}(\vec{n}\kappa) = \vec{X}(\vec{n}\kappa) + \vec{u}(\vec{n}\kappa)$. Expansion around

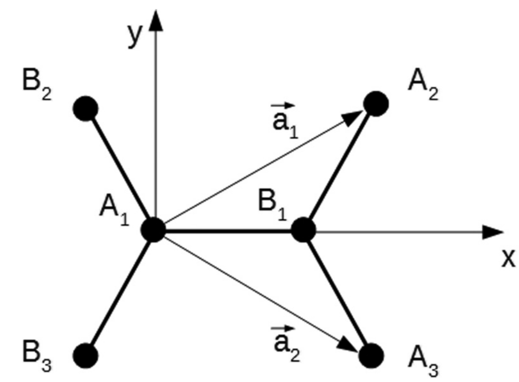

FIG. 1. Excerpt of the 2D hexagonal crystal configuration with lattice vectors $\vec{a}_1$ and $\vec{a}_2$. The unit cell consists of atoms A_1 and B_1 which, in general, can be of different nature. For each of these atoms there exist three nearest neighbors: B_1, B_2, and B_3 for A_1, and A_1, A_2, and A_3 for B_1.

the equilibrium positions leads to the Taylor series

$$V = \Phi^{(0)} + \Phi^{(1)} + \Phi^{(2)} + \Phi^{(3)} + \Phi^{(4)} + \cdots, \tag{4}$$

where

$$\Phi^{(\nu)} = \frac{1}{\nu!}\sum \Phi^{(\nu)}_{i_1,i_2,\ldots,i_\nu}(\vec{n}_1\kappa_1,\vec{n}_2\kappa_2,\ldots,\vec{n}_\nu\kappa_\nu) \times u_{i_1}(\vec{n}_1\kappa_1)u_{i_2}(\vec{n}_2\kappa_2)\ldots u_{i_\nu}(\vec{n}_\nu\kappa_\nu), \tag{5}$$

with summation over repeated indices $i,\vec{n},\kappa$. The coefficients $\Phi^{(\nu)}$, also called coupling parameters, are the νth-order derivatives of the potential energy V with respect to the atomic displacements. The terms $\Phi^{(0)}$ and $\Phi^{(1)}$ will play no role in the derivation of the dynamic equations. Usually, the harmonic Hamiltonian $H_h = K + \Phi^{(2)}$ can be described as a gas of noninteracting phonons while the anharmonic terms $\Phi^{(\nu)}$ with $\nu > 2$ are treated as perturbation. We will show that this point of view has to be modified in the case of flexural phonons.

It is standard to introduce normal coordinates $Q(\vec{q},\alpha)$ and conjugate momenta $P(\vec{q},\alpha)$ by considering the plane-wave expansions

$$u_i(\vec{n}\kappa) = \frac{1}{\sqrt{m_\kappa N}}\sum_{\vec{q},\alpha} \mathrm{e}_i^\kappa(\vec{q},\alpha) Q\binom{\alpha}{\vec{q}} \mathrm{e}^{i\vec{q}\cdot\vec{X}(\vec{n}\kappa)},$$

$$p_i(\vec{n}\kappa) = \sqrt{\frac{m\kappa}{N}}\sum_{\vec{q},\alpha} \mathrm{e}_i^\kappa(\vec{q},\alpha) P\binom{\alpha}{\vec{q}} \mathrm{e}^{i\vec{q}\cdot\vec{X}(\vec{n}\kappa)}. \tag{6}$$

Here $\vec{q}$ is a vector in the 2D Brillouin zone (BZ). The quantities $\mathrm{e}_i^\kappa(\vec{q},\alpha)$ with polarization index $\alpha = \{1,2,\ldots,6\}$ are the components of a complete set of orthonormalized eigenvectors, also called polarization vectors, that diagonalize the dynamical matrix

$$D_{ij}^{\kappa\kappa'}(\vec{q}) = \sum_{\vec{n}'} \frac{\Phi_{ij}^{(2)}(\vec{n}\kappa;\vec{n}'\kappa')}{\sqrt{m_\kappa m_{\kappa'}}} \mathrm{e}^{i\vec{q}\cdot[\vec{X}(\vec{n}'\kappa')-\vec{X}(\vec{n}\kappa)]}, \tag{7}$$

with the corresponding $\omega^2(\vec{q},\alpha)$ eigenvalues

$$\mathrm{e}_i^{\kappa\dagger}(\vec{q},\alpha) D_{ij}^{\kappa\kappa'}(\vec{q}) \mathrm{e}_j^{\kappa'}(\vec{q},\beta) = \omega^2(\vec{q},\alpha)\delta_{\alpha\beta}. \tag{8}$$

The harmonic Hamiltonian then reads

$$H_h = \frac{1}{2}\sum_{\vec{q},\alpha}\left[P^\dagger\binom{\alpha}{\vec{q}}P\binom{\alpha}{\vec{q}} + \omega^2(\vec{q},\alpha)Q^\dagger\binom{\alpha}{\vec{q}}Q\binom{\alpha}{\vec{q}}\right]. \quad (9)$$

In terms of phonon creation and annihilation operators $b_{\vec{q}}^{\alpha\dagger}$ and $b_{\vec{q}}^{\alpha}$, defined by

$$Q\binom{\alpha}{\vec{q}} = \sqrt{\frac{\hbar}{2\omega(\vec{q},\alpha)}}\left(b_{-\vec{q}}^{\alpha\dagger} + b_{\vec{q}}^{\alpha}\right),$$
$$P\binom{\alpha}{\vec{q}} = -i\sqrt{\frac{\hbar\omega(\vec{q},\alpha)}{2}}\left(b_{\vec{q}}^{\alpha} - b_{-\vec{q}}^{\alpha\dagger}\right), \quad (10)$$

one obtains

$$H_h = \sum_{\vec{q},\alpha}\hbar\omega(\vec{q},\alpha)\left[b_{\vec{q}}^{\alpha\dagger}b_{\vec{q}}^{\alpha} + \frac{1}{2}\right], \quad (11)$$

together with the usual commutation rules for Bose operators,

$$\left[b_{\vec{q}}^{\alpha}, b_{\vec{k}}^{\alpha'\dagger}\right] = \delta_{\vec{q}\vec{k}}\,\delta_{\alpha\alpha'}$$
$$\left[b_{\vec{q}}^{\alpha}, b_{\vec{k}}^{\alpha'}\right] = \left[b_{\vec{q}}^{\alpha\dagger}, b_{\vec{k}}^{\alpha'\dagger}\right] = 0. \quad (12)$$

In the following we will study low-frequency and long-wavelength phenomena. Out of the six phonon modes we restrict ourselves to the three acoustic modes which we label by $\alpha = \{1,2\}$ for the in-plane displacements (LA and TA, namely) and by $\alpha = \zeta$ for flexural displacements. While the in-plane acoustic phonon frequencies vanish linearly with $\vec{q}$ near the $\vec{q} = 0$ or Γ point of the BZ, the flexural phonon frequency vanishes quadratically [7], $\omega(\vec{q},\zeta) = \sqrt{\kappa_0}q^2$, where $\kappa_0 = \kappa_B/\rho_{2D}$ is the ratio of the bending rigidity κ_B to the surface density ρ_{2D}. We approximate the acoustic eigenvectors by their value at the Γ point, viz.,

$$\mathrm{e}_i^\kappa(\vec{q},\alpha) \approx \mathrm{e}_i^\kappa(0,\alpha) = \sqrt{\frac{m_\kappa}{m}}\delta_{\alpha i}, \quad (13)$$

where $m = \sum_\kappa m_\kappa$ is the total mass per unit cell. Our model not only describes graphene, where $m_1 = m_2 = m_C$ or 2D h-BN, where $m_1 = m_B$ and $m_2 = m_N$, but also 2D transition metal dichalcogenides and transition metal dioxides. In these crystals we assume that the two chalcogen or oxygen atoms move in unison (in each cell) and consider them as one single effective particle with twice their mass, in addition to the metal atom, e.g., $m_1 = m_{Mo}$ and $m_2 = 2m_S$ in the case of 2H MoS_2. Each of the acoustic polarization vectors then has six components; the first three of them refer to the (x,y,z) components of the $\kappa = 1$ atom, while the last three refer to the $\kappa = 2$ atom:

$$\vec{e}(0,1) = \left(\sqrt{\frac{m_1}{m}},0,0,\sqrt{\frac{m_2}{m}},0,0\right),$$
$$\vec{e}(0,2) = \left(0,\sqrt{\frac{m_1}{m}},0,0,\sqrt{\frac{m_2}{m}},0\right),$$
$$\vec{e}(0,\zeta) = \left(0,0,\sqrt{\frac{m_1}{m}},0,0,\sqrt{\frac{m_2}{m}}\right). \quad (14)$$

These expressions are valid approximations for the acoustic polarization vectors at long wavelengths (sound waves). Further out in the BZ one has to resort to the diagonalization of the dynamical matrix by numerical methods [15,16]. The Fourier-transformed center-of-mass displacements per unit cell are then related to the normal coordinates by

$$s_i(\vec{q}) = \frac{1}{\sqrt{m}}Q\binom{\alpha}{\vec{q}}\delta_{\alpha i} \quad (15)$$

and to uniform in-plane strains by

$$\epsilon_{ij} = \frac{i}{\sqrt{Nm}}\lim_{\vec{q}\to 0} q_j Q\binom{i}{\vec{q}}. \quad (16)$$

Turning to the anharmonic contributions, we restrict ourselves to third- and fourth-order processes $\Phi^{(3)}$ and $\Phi^{(4)}$, where one and two in-plane phonons, respectively, scatter with two flexural modes. Using Eqs. (5) and (6) we obtain

$$\Phi^{(3)} = \frac{1}{2}\sum_{\vec{p},\vec{k},\vec{q},i}\Phi^{(3)}\begin{pmatrix} i & \zeta & \zeta \\ \vec{q} & \vec{k} & \vec{p}\end{pmatrix}Q\binom{i}{\vec{q}}Q\binom{\zeta}{\vec{k}}Q\binom{\zeta}{\vec{p}},$$
$$\Phi^{(4)} = \frac{1}{2}\sum_{\vec{p},\vec{k},\vec{q},\vec{h},i,j}\Phi^{(4)}\begin{pmatrix} i & j & \zeta & \zeta \\ \vec{p} & \vec{q} & \vec{k} & \vec{h}\end{pmatrix}$$
$$\times Q\binom{i}{\vec{p}}Q\binom{j}{\vec{q}}Q\binom{\zeta}{\vec{k}}Q\binom{\zeta}{\vec{h}}. \quad (17)$$

Due to planar symmetry, the flexural mode operators appear in pairs [23]. We deliberately neglect cubic in-plane anharmonicities, since at low temperature in-plane phonons are less likely to be excited than flexural phonons. Invariance of the crystal with respect to translation by a lattice vector implies that $\Phi^{(3)}(\vec{q}i,\vec{k}\zeta,\vec{p}\zeta)$ is proportional to a factor

$$\Delta(\vec{q}+\vec{k}+\vec{p}) = \sum_{\vec{g}}\delta_{\vec{q}+\vec{k}+\vec{p},\vec{g}}\,, \quad (18)$$

where $\vec{g}$ is a reciprocal 2D lattice vector. Scattering processes with $\vec{g} = 0$ are called normal processes and those with $\vec{g} \neq 0$ are umklapp processes [41]. Invariance of the potential energy of the crystal against infinitesimal rigid body translations implies

$$\Phi^{(3)}\begin{pmatrix} i & \zeta & \zeta \\ \vec{q}=0 & \vec{k} & -\vec{k}\end{pmatrix} = 0. \quad (19)$$

Properties similar to Eqs. (18) and (19) hold for $\Phi^{(4)}(\vec{p}i,\vec{q}j,\vec{k}\zeta,\vec{h}\zeta)$.

We show now that as a consequence of anharmonicity the presence of in-plane strains leads to a change of the flexural mode frequency $\omega(\vec{k},\zeta)$. From Eq. (9) it follows that the contribution of the flexural modes to the harmonic potential energy is given by

$$\Phi^{(2)}(\zeta) = \frac{1}{2}\sum_{\vec{k}}\omega^2(\vec{k},\zeta)Q^\dagger\binom{\zeta}{\vec{k}}Q\binom{\zeta}{\vec{k}}. \quad (20)$$

Likewise, for a given in-plane long-wavelength lattice displacement with wave vector $\vec{q}$ and polarization i, the contribution of the flexural mode to the third-order anharmonic potential reads

$$\Phi^{(3)}(\zeta) = \frac{1}{2}\sum_{\vec{k}} \Phi^{(3)}\begin{pmatrix} i & \zeta & \zeta \\ \vec{q} & -\vec{k} & \vec{k}-\vec{q} \end{pmatrix} \times Q\begin{pmatrix} i \\ \vec{q} \end{pmatrix} Q^{\dagger}\begin{pmatrix} \zeta \\ \vec{k} \end{pmatrix} Q\begin{pmatrix} \zeta \\ \vec{k} \end{pmatrix}. \quad (21)$$

Adding Eqs. (20) and (21), we infer that the square of the flexural mode frequency in the deformed lattice is given by

$$\tilde{\omega}^2(\vec{k},\zeta)|_{\vec{q}i} = \omega^2(\vec{k},\zeta) + \Phi^{(3)}\begin{pmatrix} i & \zeta & \zeta \\ \vec{q} & -\vec{k} & \vec{k}-\vec{q} \end{pmatrix} Q\begin{pmatrix} i \\ \vec{q} \end{pmatrix}. \quad (22)$$

At small $\vec{q}$ the anharmonic coupling is linear in $\vec{q}$, and using Eq. (16) we get in the limit $\vec{q} \to 0$

$$\tilde{\omega}^2(\vec{k},\zeta) = \omega^2(\vec{k},\zeta) - i\sqrt{Nm}\,\epsilon_{ij}\frac{\partial}{\partial q_i}\Phi^{(3)}\begin{pmatrix} j & \zeta & \zeta \\ \vec{q} & -\vec{k} & \vec{k} \end{pmatrix}\Bigg|_{\vec{q}=0}. \quad (23)$$

We notice that $\Phi^{(3)}(\vec{q}\,j,-\vec{k}\zeta,\vec{k}\zeta)$ varies as k^2 in the limit $\vec{k} \to 0$, while $\omega^2(\vec{k},\zeta)$ varies as k^4. Hence, in contradistinction to the case of nonlayered 3D crystals, the anharmonic term cannot be considered as a perturbation to $\omega^2(\vec{k},\zeta)$ in Eq. (23) at long wavelengths $2\pi/k$. A further deficiency of result (23) is the divergence of the generalized Grüneisen coefficient

$$\gamma_{ij}(\vec{k}) = -\frac{\partial \ln(\tilde{\omega}(\vec{k},\zeta))}{\partial \epsilon_{ij}} \quad (24)$$

for $\vec{k} = 0$. In order to correct these shortcomings that have their origin in the k^2 dependence of the bare flexural mode frequency $\omega(\vec{k},\zeta)$, we will replace the latter by the renormalized frequency $\omega_R(\vec{k},\zeta)$, obtained in the lowest order renormalized harmonic approximation (RHA).

Henceforth we will derive an expression for $\omega_R(\vec{k},\zeta)$ starting from the fourth-order anharmonic interaction $\Phi^{(4)}$ defined by Eq. (17). We replace there the product of the in-plane displacement operators by their thermal average

$$\left\langle Q^{\dagger}\begin{pmatrix} i \\ \vec{p} \end{pmatrix} Q\begin{pmatrix} j \\ \vec{q} \end{pmatrix}\right\rangle = \delta_{ij}\,\delta_{-\vec{p},\vec{q}}\left\langle Q^{\dagger}\begin{pmatrix} i \\ \vec{q} \end{pmatrix} Q\begin{pmatrix} i \\ \vec{q} \end{pmatrix}\right\rangle. \quad (25)$$

The thermal average is rewritten in terms of phonon creation and annihilation operators Eq. (10) with the result

$$\left\langle Q^{\dagger}\begin{pmatrix} i \\ \vec{q} \end{pmatrix} Q\begin{pmatrix} i \\ \vec{q} \end{pmatrix}\right\rangle = \frac{\hbar}{\omega(\vec{q},i)}\left[n(\vec{q},i) + \frac{1}{2}\right], \quad (26)$$

where $n(\vec{q},i) = [\exp(\hbar\omega(\vec{q},i)/k_B T) - 1]^{-1}$ is the in-plane phonon thermal density distribution. Taking into account Eq. (25) and noting that now $\Delta(\vec{p}+\vec{q}+\vec{k}+\vec{h}) = \delta_{-\vec{h},\vec{k}}$, we obtain for $\Phi^{(4)}$ the approximation

$$\Phi^{(4)}(\zeta) = \frac{1}{2}\sum_{\vec{k},\vec{q},i} \Phi^{(4)}\begin{pmatrix} i & i & \zeta & \zeta \\ \vec{q} & -\vec{q} & \vec{k} & -\vec{k} \end{pmatrix} \times \left\langle Q^{\dagger}\begin{pmatrix} i \\ \vec{q} \end{pmatrix} Q\begin{pmatrix} i \\ \vec{q} \end{pmatrix}\right\rangle Q^{\dagger}\begin{pmatrix} \zeta \\ \vec{k} \end{pmatrix} Q\begin{pmatrix} \zeta \\ \vec{k} \end{pmatrix}. \quad (27)$$

Addition of this expression member by member with Eq. (20) yields

$$\Phi^{(2)}(\zeta) + \Phi^{(4)}(\zeta) = \frac{1}{2}\sum_{\vec{k}} \omega_R^2(\vec{k},\zeta) Q^{\dagger}\begin{pmatrix} \zeta \\ \vec{k} \end{pmatrix} Q\begin{pmatrix} \zeta \\ \vec{k} \end{pmatrix}, \quad (28)$$

where we have defined the RHA squared flexural phonon frequency

$$\omega_R^2(\vec{k},\zeta) = \omega^2(\vec{k},\zeta) + \sum_{\vec{q},i} \Phi^{(4)}\begin{pmatrix} i & i & \zeta & \zeta \\ \vec{q} & -\vec{q} & \vec{k} & -\vec{k} \end{pmatrix} \times \left\langle Q^{\dagger}\begin{pmatrix} i \\ \vec{q} \end{pmatrix} Q\begin{pmatrix} i \\ \vec{q} \end{pmatrix}\right\rangle. \quad (29)$$

In the regime of small $\vec{k}$ the first term is $\propto k^4$ while the second term is $\propto k^2$. Hence the renormalized frequency $\omega_R(\vec{k},\zeta)$ becomes linear in $\vec{k}$ for small wave vectors. Adding now Eqs. (21) and (28), we can treat the third-order anharmonic term as a perturbation to $\omega_R(\vec{k},\zeta)$. Taking the square root we obtain for the renormalized phonon frequency in presence of uniform in-plane strains the result

$$\tilde{\omega}_R(\vec{k},\zeta) = \omega_R(\vec{k},\zeta) - \epsilon_{ij} h_{ij}(\vec{k},\zeta), \quad (30)$$

where

$$h_{ij}(\vec{k},\zeta) = \frac{i\sqrt{Nm}}{2\omega_R(\vec{k},\zeta)}\frac{\partial}{\partial q_i}\Phi^{(3)}\begin{pmatrix} j & \zeta & \zeta \\ \vec{q} & -\vec{k} & \vec{k} \end{pmatrix}\Bigg|_{\vec{q}=0}. \quad (31)$$

The generalized Grüneisen coefficient within RHA,

$$\gamma_{ij}(\vec{k},\zeta) = -\frac{\partial \ln(\tilde{\omega}_R(\vec{k},\zeta))}{\partial \epsilon_{ij}} = \frac{h_{ij}(\vec{k},\zeta)}{\omega_R(\vec{k},\zeta)}, \quad (32)$$

remains finite for $\vec{k} \to 0$.

From the continuum theory of crystalline membranes [13], one knows that the anharmonic coupling between stretching and bending modes leads to a phonon-mediated interaction between Gaussian curvatures that in turn increases the bending rigidity. These concepts have been applied to the study of thermal and elastic properties of graphene and related systems [42,43]. On the other hand, within the lattice dynamical theory, renormalization of the flexural phonon frequencies is due to third- and fourth-order anharmonic interactions [15,16]. In Refs. [15,16], anharmonic coupling parameters have been calculated for the case of a central force interatomic potential between nearest neighbors. By applying these concepts here with approximation (13) for the polarization vectors we obtain

$$\Phi^{(3)}\begin{pmatrix} i & \zeta & \zeta \\ \vec{q} & \vec{k} & \vec{p} \end{pmatrix} = \frac{i8}{\sqrt{Nm^3}}\sum_s \phi^{(3)}_{izz}(A_1;B_s) \times \cos\left[\frac{(\vec{q}+\vec{k}+\vec{p})\cdot\vec{\rho}(B_s)}{2}\right] \times \sin\left[\frac{\vec{q}\cdot\vec{r}(B_s)}{2}\right]\sin\left[\frac{\vec{k}\cdot\vec{r}(B_s)}{2}\right] \times \sin\left[\frac{\vec{p}\cdot\vec{r}(B_s)}{2}\right]\Delta(\vec{q}+\vec{k}+\vec{p}), \quad (33)$$

TABLE I. Third-order anharmonic force constants for nearest-neighbor atoms. Numerical values $h^{(3)} = -3.35$ for graphene and 2D h-BN, $h^{(3)} = -0.335$ for 2H MoS_2 have been estimated [15] from acoustic mode Grüneisen parameters. Units in 10^{12} erg/cm^3.

B_s	B_1	B_2	B_3
$\phi^{(3)}_{xzz}(A;B_s)$	$h^{(3)}$	$\frac{-1}{2}h^{(3)}$	$\frac{-1}{2}h^{(3)}$
$\phi^{(3)}_{yzz}(A;B_s)$	0	$\frac{\sqrt{3}}{2}h^{(3)}$	$\frac{-\sqrt{3}}{2}h^{(3)}$

where $\phi^{(3)}_{izz}$ is the third-order derivative of the two-body potential (see Table I). Here $\vec{r}(B_s)$, $s = 1,2,3$, is the position vector of particle B_s counted from A_1 origin as shown in Fig. 1. We have defined $\vec{\rho}(B_s) = \vec{r}(B_1) - \vec{r}(B_s)$ and hence $\vec{\rho}(B_1) = 0$, $\vec{\rho}(B_2) = \vec{a}_2$, and $\vec{\rho}(B_3) = \vec{a}_1$. The derivative of the renormalized flexural mode frequency with respect to ϵ_{ij} is now given by

$$h_{ij}(\vec{k},\zeta) = \frac{-\partial\tilde{\omega}_R(\vec{k},\zeta)}{\partial\epsilon_{ij}} = \frac{1}{2m\omega_R(\vec{k},\zeta)}\sum_s \phi^{(3)}_{izz}(A_1;B_s)(\vec{k}\cdot\vec{r}(B_s))^2 r_j(B_s). \tag{34}$$

Using the force constants from Table I we obtain

$$h_{11}(\vec{k},\zeta) = \frac{\sqrt{3}h^{(3)}a^3}{16m\omega_R(\vec{k},\zeta)}\left(k_x^2 + \frac{k_y^2}{3}\right), \tag{35}$$

and $h_{22}(\vec{k},\zeta)$ by interchanging k_x with k_y. This expression differs by a factor of 2 from Eq. (43) in Ref. [15] where this factor has been omitted. The same mistake should be corrected also in Eq. (48). Following Refs. [15] and [16] we take $h^{(3)} < 0$. It then follows from Eq. (30) that a positive strain leads to an increase of the flexural mode frequency, which is equivalent to a negative ZA Grüneisen constant [9] and hence to a negative thermal expansion [8]. The change of phonon frequencies under strains can be traced back to Mie and Grüneisen [44,45], who assumed temperature-dependent lattice constants in cubic crystals. These concepts have been generalized to time-dependent phenomena by the theoretical work of Akhiezer [46] and Woodruff and Ehrenreich [47], who studied the absorption of sound waves and the concomitant change of thermal phonon frequencies. Early experiments on sound attenuation giving information on phonon-phonon relaxation precesses were carried out by Bömmel and Dransfeld [48]. In the following Secs. IV and V we will show that the quantity $h_{ij}(\vec{k},\zeta)$ accounts for the coupling of hydrodynamic in-plane lattice displacements (sound waves) to the kinetics of flexural mode density fluctuations and vice versa.

III. RESPONSE FUNCTIONS

Since we want to study the crystal under the influence of external perturbations such as mechanical stresses and heat sources we recall some concepts of linear response theory [37,49]. One adds to the crystal Hamiltonian $H = K + V$ a time-dependent external perturbation:

$$H'(t) = -\sum_{\vec{q}} F(\vec{q},t)B(-\vec{q}). \tag{36}$$

Here $F(\vec{q},t)$ is an external field switched adiabatically at $t = -\infty$ and $B(-\vec{q})$ is the operator of an observable of the system. We define wave-vector and frequency-dependent Fourier transforms by

$$F(\vec{q},\omega) = \frac{1}{\sqrt{N}}\sum_{\vec{n}}\int dt\; e^{i(\omega t - \vec{q}\cdot\vec{X}(\vec{n}))}F(\vec{n},t). \tag{37}$$

In the case of a periodic field varying with frequency ω, the change of a physical quantity characterized by an operator $A(\vec{q})$ is given in linear approximation with respect to $F(\vec{q},\omega)$ by

$$\delta\langle A(\vec{q})\rangle_\omega = -\langle\langle A(\vec{q})\,;B(-\vec{q})\rangle\rangle_z F(\vec{q},\omega), \tag{38}$$

where $\langle\langle A\,;B\rangle\rangle_z$ is the retarded Green's function (GF)

$$\langle\langle A\,;B\rangle\rangle_z = -\frac{i}{\hbar}\int_0^\infty dt\; e^{izt}\langle[A(t),B(0)]\rangle, \tag{39}$$

with $z = \omega + i\epsilon$, $\epsilon \to 0+$, and $A(t) = e^{iHt/\hbar}Ae^{-iHt/\hbar}$, where H refers to the unperturbed system. The retarded GF is completely determined by the Hamiltonian H and by the temperature T at $t = -\infty$. Of special interest is the static response to a wave-vector-dependent field $F(\vec{q})$ switched on arbitrarily slowly, i.e., $F(\vec{q})e^{\epsilon t}$, $t < 0$, $\epsilon \to 0^+$. The wave-number-dependent response at time $t = 0$ is then given by

$$\delta\langle A(\vec{q})\rangle = \chi_{AB}(\vec{q})F(\vec{q}), \tag{40}$$

where

$$\chi_{AB}(\vec{q}) = -\langle\langle A(\vec{q})\,;B(-\vec{q})\rangle\rangle_{z=0} \tag{41}$$

is the static susceptibility [50]. For the present case we consider a perturbation of the form [51]

$$H'(t) = -s_i(-\vec{q})F_i(\vec{q},t) - \frac{\epsilon(-\vec{q})}{T}\Theta(\vec{q},t), \tag{42}$$

where $F_i(\vec{q},t)$, with $i = \{x,y\}$, is an external in-plane displacement force, $\Theta(\vec{q},t)$ an external temperature, and $\epsilon(\vec{q})$ the energy-density operator of phonons

$$\epsilon(\vec{q}) = \frac{1}{\sqrt{N}}\sum_{\vec{k},\alpha}\hbar\omega(\vec{k},\alpha)b^{\alpha\dagger}_{\vec{k}-\vec{q}}\,b^{\alpha}_{\vec{k}}. \tag{43}$$

Here $\omega(\vec{k},\zeta)$ has to be replaced by $\omega_R(\vec{k},\zeta)$ for the case $\alpha = \zeta$. Application of Eq. (38) then gives the in-plane lattice displacement response

$$s_i(\vec{q},\omega) = -D^s_{ij}(\vec{q},z)F_j(\vec{q},\omega) - D^\epsilon_i(\vec{q},z)\frac{\Theta(\vec{q},\omega)}{T}, \tag{44}$$

where

$$D^s_{ij}(\vec{q},z) = \langle\langle s_i(\vec{q})\,;s_j(-\vec{q})\rangle\rangle_z,$$
$$D^\epsilon_i(\vec{q},z) = \langle\langle s_i(\vec{q})\,;\epsilon(-\vec{q})\rangle\rangle_z, \tag{45}$$

are the displacement-displacement correlation and the displacement-energy correlation, respectively. We will also need the following phonon density correlations:

$$G_j^s(\vec{k},\alpha;\vec{q},z) = \langle\langle b_{\vec{k}-\vec{q}}^{\alpha\dagger} b_{\vec{k}}^{\alpha}\,;s_j(-\vec{q})\rangle\rangle_z \sqrt{N},$$
$$G^{\epsilon}(\vec{k},\alpha;\vec{q},z) = \langle\langle b_{\vec{k}-\vec{q}}^{\alpha\dagger} b_{\vec{k}}^{\alpha}\,;\epsilon(-\vec{q})\rangle\rangle_z \sqrt{N}. \tag{46}$$

Starting from the equation of motion for retarded GFs,

$$z\langle\langle A\,;B\rangle\rangle_z = \frac{1}{\hbar}\langle[A,B]\rangle + \frac{1}{\hbar}\langle\langle[A,H]\,;B\rangle\rangle_z, \tag{47}$$

we will derive in the following Secs. IV and V a set of coupled dynamic equations for the in-plane phonon displacements and the flexural phonon density distribution function.

IV. DISPLACEMENTS RESPONSE

The derivation of dynamic equations for $s_i(\vec{q},\omega)$ amounts to a derivation of the equations of motion for the correlation functions $D_{ij}^s(\vec{q},z)$ and $D_i^{\epsilon}(\vec{q},z)$. Thereby we will take the crystal Hamiltonian $H = H_{hR} + \Phi_R^{(3)}$, where H_{hR} and $\Phi_R^{(3)}$ are obtain from Eqs. (11) and (17), respectively, by replacing $\omega(\vec{k},\zeta)$ by $\omega_R(\vec{k},\zeta)$. Here and in the following Secs. V and VI it is understood that we consider the flexural mode with frequency $\omega_R(\vec{k},\zeta)$. However, for the sake of notation we will drop the subscript R and write $\omega(\vec{k},\zeta)$ instead. It is convenient to formulate the problem in terms of normal coordinates and to consider instead

$$D_{ij}^{Q}(\vec{q},z) = \left\langle\left\langle Q\begin{pmatrix} i \\ \vec{q}\end{pmatrix}; Q\begin{pmatrix} j \\ -\vec{q}\end{pmatrix}\right\rangle\right\rangle_z,$$
$$D_{i}^{Q\epsilon}(\vec{q},z) = \left\langle\left\langle Q\begin{pmatrix} i \\ \vec{q}\end{pmatrix}; \epsilon(-\vec{q})\right\rangle\right\rangle_z. \tag{48}$$

We start with a study of the displacement-displacement correlation $D_{ij}^{Q}(\vec{q},z)$. As shown in the Appendix, D_{ij}^{Q} satisfies the following equation of motion:

$$[z^2 - \omega^2(\vec{q},i)]D_{ij}^{Q}(\vec{q},z) = \delta_{ij} + \frac{1}{2}\sum_{\vec{k}}\Phi^{(3)}\begin{pmatrix} i & \zeta & \zeta \\ -\vec{q} & \vec{k} & -\vec{k}+\vec{q}\end{pmatrix}\left\langle\left\langle Q\begin{pmatrix} \zeta \\ \vec{k}\end{pmatrix} Q\begin{pmatrix} \zeta \\ \vec{q}-\vec{k}\end{pmatrix}; Q\begin{pmatrix} j \\ -\vec{q}\end{pmatrix}\right\rangle\right\rangle_z. \tag{49}$$

Here we have taken into account that for small wave vectors $\vec{q}$, the crystal momentum is conserved and there remains a single $\vec{k}$ summation over the BZ. To proceed further we disentangle the GF on the right-hand side (rhs) of Eq. (49) by writing

$$\left\langle\left\langle Q\begin{pmatrix} \zeta \\ \vec{k}\end{pmatrix} Q\begin{pmatrix} \zeta \\ \vec{q}-\vec{k}\end{pmatrix}; Q\begin{pmatrix} j \\ -\vec{q}\end{pmatrix}\right\rangle\right\rangle_z = \frac{\hbar}{2\sqrt{\omega(\vec{k},\zeta)\omega(\vec{k}-\vec{q},\zeta)}}\left[\left\langle\left\langle b_{-\vec{k}}^{\zeta\dagger}\, b_{-\vec{k}+\vec{q}}^{\zeta}; Q\begin{pmatrix} j \\ -\vec{q}\end{pmatrix}\right\rangle\right\rangle_z + \left\langle\left\langle b_{\vec{k}}^{\zeta}\, b_{\vec{k}-\vec{q}}^{\zeta\dagger}; Q\begin{pmatrix} j \\ -\vec{q}\end{pmatrix}\right\rangle\right\rangle_z\right.$$
$$\left. + \left\langle\left\langle b_{-\vec{k}}^{\zeta\dagger}\, b_{\vec{k}-\vec{q}}^{\zeta\dagger}; Q\begin{pmatrix} j \\ -\vec{q}\end{pmatrix}\right\rangle\right\rangle_z + \left\langle\left\langle b_{\vec{k}}^{\zeta}\, b_{-\vec{k}+\vec{q}}^{\zeta}; Q\begin{pmatrix} j \\ -\vec{q}\end{pmatrix}\right\rangle\right\rangle_z\right], \tag{50}$$

and calculate separately the four GFs on the rhs. Due to lattice anharmonicities, the corresponding equations of motion lead to higher-order GFs. In order to close this hierarchy and to obtain a closed expression for the initial function $D_{ij}^{Q}(\vec{q},z)$, we follow the method of Ref. [38] and approximate the higher-order GFs by decoupling techniques. In first-order perturbation theory we obtain closed expressions for all GFs on the rhs of Eq. (50) that are proportional to $D_{ij}^{Q}(\vec{q},z)$ (see Appendix). By substituting the results back into Eq. (49) we arrive at the Dyson equation

$$[z^2\delta_{ij} - M_{ik}(\vec{q},z)]D_{kj}^{Q}(\vec{q},z) = \delta_{ij}. \tag{51}$$

The self-energy matrix has the elements

$$M_{ik}(\vec{q},z) = \omega^2(\vec{q},i)\delta_{ik} + \frac{\hbar}{8}\sum_{\vec{k}}\frac{\Phi^{(3)}\begin{pmatrix} i & \zeta & \zeta \\ -\vec{q} & \vec{k} & \vec{q}-\vec{k}\end{pmatrix}\Phi^{(3)}\begin{pmatrix} k & \zeta & \zeta \\ \vec{q} & -\vec{k} & \vec{k}-\vec{q}\end{pmatrix}}{\omega(\vec{k},\zeta)\omega(\vec{k}-\vec{q},\zeta)}$$
$$\times\left[2\left(\frac{n(\vec{k}-\vec{q},\zeta)-n(\vec{k},\zeta)}{z+\omega(\vec{k}-\vec{q},\zeta)-\omega(\vec{k},\zeta)}\right) + \frac{1+n(\vec{k}-\vec{q},\zeta)+n(\vec{k},\zeta)}{z-\omega(\vec{k}-\vec{q},\zeta)-\omega(\vec{k},\zeta)} - \frac{1+n(\vec{k}-\vec{q},\zeta)+n(\vec{k},\zeta)}{z+\omega(\vec{k}-\vec{q},\zeta)+\omega(\vec{k},\zeta)}\right]. \tag{52}$$

Here, $\omega(\vec{k},\zeta)$ is the renormalized flexural phonon frequency and $n(\vec{k},\zeta) = [\exp(\hbar\omega(\vec{k},\zeta)/k_BT)-1]^{-1}$ the thermal equilibrium density distribution. In Eq. (52) the first term within square brackets is due to the sum of the first two terms on the rhs of Eq. (50), while the second and third term are due to the fourth and third term, respectively.

Expression (51) with $M_{ik}(\vec{q},z)$ given by Eq. (52) is a valid approximation of Eq. (49) for $z \neq 0$ and $\vec{q} \neq 0$, such that the resonances of $D_{kj}^{Q}(\vec{q},z)$ are high-frequency sound waves. However, this approximation breaks down in the hydrodynamic regime in the simultaneous limits $z \to 0$ and $\vec{q} \to 0$. While the second and third term within square brackets in Eq. (52) are regular, the first term depends on the order in which the limits are taken. In lowest order of $\Phi^{(3)}$ we obtain for the change of the phonon density distribution

$$\left\langle\left\langle b_{\vec{k}-\vec{q}}^{\zeta\dagger}\, b_{\vec{k}}^{\zeta}; Q\begin{pmatrix} j \\ -\vec{q}\end{pmatrix}\right\rangle\right\rangle_z = \frac{f(\vec{k},\zeta;\vec{q},z)}{2\sqrt{\omega(\vec{k},\zeta)\omega(\vec{k}-\vec{q},\zeta)}}\Phi^{(3)}\begin{pmatrix} k & \zeta & \zeta \\ \vec{q} & -\vec{k} & \vec{k}-\vec{q}\end{pmatrix}D_{kj}^{Q}(\vec{q},z), \tag{53}$$

with f defined by

$$f(\vec{k},\zeta;\vec{q},z) = \frac{n(\vec{k}-\vec{q},\zeta) - n(\vec{k},\zeta)}{z + \omega(\vec{k}-\vec{q},\zeta) - \omega(\vec{k},\zeta)}. \tag{54}$$

Obviously this expression is a discontinuous function for $(\vec{q},z) \to 0$: $f(\vec{k},\zeta;\vec{0},z) = 0$, while $-f(\vec{k},\zeta;\vec{q},0)/\hbar = n(\vec{k},\zeta)[1+n(\vec{k},\zeta)]/k_BT \equiv n'(\vec{k},\zeta)$. In order to avoid this singularity that appears only at $T \neq 0$ in the self-energy of the Dyson equation for D^Q_{ij} we have to improve our approximation in Eq. (53) for the phonon density function $\langle\langle b^{\zeta\dagger}_{\vec{k}-\vec{q}} b^{\zeta}_{\vec{k}}; Q(j,-\vec{q})\rangle\rangle_z$. Indeed, in every order of perturbation theory one finds contributions with a vanishing energy denominator. The physical reason for the singular behavior is that at nonzero T the self-energy of D^Q_{ij} has poles due to hydrodynamic phenomena such as heat conduction and second sound [34,36,38]. Such poles cannot be described by first-order perturbation theory. In Sec. V we will show that the summation of leading divergent terms that contribute to the singularities of the self-energy amounts to the derivation of kinetic equations for the phonon density distribution functions. In the following we use the notation

$$G^Q_j(\vec{k},\zeta;\vec{q},z) = \left\langle\!\!\left\langle b^{\zeta\dagger}_{\vec{k}-\vec{q}}\, b^{\zeta}_{\vec{k}}; Q\begin{pmatrix} j \\ -\vec{q}\end{pmatrix}\right\rangle\!\!\right\rangle_z \sqrt{N}. \tag{55}$$

We write G^Q_j as a sum of two terms, the first one proportional to $D^Q_{kj}(\vec{q},z)$ is obtained from the long-wavelength expansion of the rhs of Eq. (53) with $f(\vec{k},\zeta;\vec{q},0)$; the second one denoted by Γ^Q_j has still to be determined:

$$G^Q_j(\vec{k},\zeta;\vec{q},z) = \frac{i\hbar}{\sqrt{m}} n'(\vec{k},\zeta) q_i h_{ik}(\vec{k},\zeta) D^Q_{kj}(\vec{q},z) + \Gamma^Q_j(\vec{k},\zeta;\vec{q},z). \tag{56}$$

We have again used the fact that at small $\vec{q}$ the anharmonic coupling $\Phi^{(3)}$ is a linear function of $\vec{q}$. The quantity $h_{ij}(\vec{k},\zeta)$ agrees with the definition (31). Hence the first term on the rhs of Eq. (56) accounts for a contribution to the nonequilibrium flexural phonon density distribution due to coherent dynamic lattice deformations that are characteristic for sound waves. This term has the meaning of a local equilibrium distribution. The Γ^Q_j term describes the deviations from local equilibrium; in Sec. V we will show that it satisfies a kinetic equation with a Peierls-Boltzmann–type collision term.

We return to Eq. (50) and replace the first two functions within square brackets by Eq. (56). As before, we keep the third and fourth functions in the linear approximation proportional to D^Q_{kj}. Finally, we insert the result into Eq. (49) and arrive at a modified Dyson equation for the in-plane displacement-displacement correlations:

$$[z^2\delta_{ik} - M_{ik}(\vec{q},0)] D^Q_{kj}(\vec{q},z) = \delta_{ij} + \frac{i\hbar}{N\sqrt{m}} \sum_{\vec{k}} q_l h_{li}(\vec{k},\zeta) \times \Gamma^Q_j(\vec{k},\zeta;\vec{q},z). \tag{57}$$

The function $\Gamma^Q_j(\vec{k},\zeta;\vec{q},z)$ accounts for the deviations from thermal equilibrium of the flexural mode density distribution and acts as a perturbation to the displacement correlations.

So far we have considered correlations D^Q_{ij} and G^Q_j that describe the linear response to an external mechanical displacement force [first term on the rhs of Eq. (42)]. In order to study the response to an external temperature perturbation [second term on the rhs of Eq. (42)], we have to start from $D^{Q\epsilon}_i(\vec{q},z)$, Eq. (48). We can repeat step by step the procedure that led to Eq. (57). The only changes will occur in the presence or absence of inhomogeneous terms, due to the expectation value $\langle[A,B]\rangle$ in the general equation of motion Eq. (47) for any GF. Instead of Eq. (57) the equation of motion for $D^{Q\epsilon}_i(\vec{q},z)$ reads

$$[z^2\delta_{ik} - M_{ik}(\vec{q},0)] D^{Q\epsilon}_k(\vec{q},z) = +\frac{i\hbar}{N\sqrt{m}} \sum_{\vec{k}} q_l h_{li}(\vec{k},\zeta) \times \Gamma^{\epsilon}(\vec{k},\zeta;\vec{q},z), \tag{58}$$

where $\Gamma^\epsilon(\vec{k},\zeta;\vec{q},z)$ has been defined by

$$G^{\epsilon}(\vec{k},\zeta;\vec{q},z) = \frac{i\hbar}{\sqrt{m}} n'(\vec{k},\zeta) q_l h_{lk}(\vec{k},\zeta) D^{Q\epsilon}_k(\vec{q},z) + \Gamma^{\epsilon}(\vec{k},\zeta;\vec{q},z). \tag{59}$$

We rewrite Eqs. (57) and (58) in terms of center-of-mass displacement operators $s_i(\vec{q})$, Eq. (15), and obtain

$$[z^2\delta_{ij} - M_{ij}(\vec{q},0)] D^s_{jl}(\vec{q},z) = \frac{1}{m}\delta_{il} + \frac{i\hbar}{Nm} \sum_{\vec{k}} q_j h_{ji}(\vec{k},\zeta) \times \Gamma^s_l(\vec{k},\zeta;\vec{q},z), \tag{60}$$

$$[z^2\delta_{ij} - M_{ij}(\vec{q},0)] D^{\epsilon}_j(\vec{q},z) = +\frac{i\hbar}{Nm} \sum_{\vec{k}} q_j h_{ji}(\vec{k},\zeta) \times \Gamma^{\epsilon}(\vec{k},\zeta;\vec{q},z). \tag{61}$$

Multiplying Eq. (60) by $-F_l(\vec{q},z)$ and Eq. (61) by $-\Theta(\vec{q},z)/T$ and adding the results, we arrive at the first coupled differential equation for the in-plane lattice displacement response:

$$[z^2\delta_{ij} - M_{ij}(\vec{q},0)] s_j(\vec{q},z) = \frac{-F_i(\vec{q},z)}{m} + \frac{i\hbar}{Nm} \sum_{\vec{k}} q_j h_{ji}(\vec{k},\zeta) \times n'(\vec{k},\zeta)\nu(\vec{k},\zeta;q,z). \tag{62}$$

Here, $\nu(\vec{k},\zeta;q,z)$ is the nonequilibrium flexural phonon density distribution function defined by

$$n'(\vec{k},\zeta)\nu(\vec{k},\zeta;q,z) = -\Gamma^s_l(\vec{k},\zeta;q,z) F_l(\vec{q},z) - \Gamma^{\epsilon}(\vec{k},\zeta;q,z) \frac{\Theta(\vec{q},z)}{T}. \tag{63}$$

In Sec. VI we show that the quantity $M_{ij}(\vec{q},0)$ is related to the isothermal elastic constants $C_{ij,kl}$. In case of 2D crystals we write

$$M_{ij}(\vec{q},0) = \frac{v_{2D}}{m} q_k q_l C_{ik,jl}, \tag{64}$$

where v_{2D} is the surface of the unit cell, and as a consequence of hexagonal symmetry one has $C_{11,11} \equiv \gamma_{11}$, $C_{11,22} \equiv \gamma_{12}$ with $\gamma_{11} - \gamma_{12} = 2\gamma_{66}$, $\gamma_{66} \equiv C_{12,12}$. These quantities have the dimension of surface tensions. It will be useful to express the external force $F_i(\vec{q},z)$ in terms of a second-rank stress tensor $\sigma_{ij}(\vec{q},z)$,

$$F_i(\vec{q},z) = -i v_{2D} q_j \sigma_{ij}(\vec{q},z), \tag{65}$$

and to rewrite Eq. (62) as a sound wave equation:

$$\left[z^2\delta_{ij} - \frac{v_{2D}}{m} q_k q_l C_{ik,jl}\right] s_j(\vec{q},z) = \frac{i q_j}{m}\left[v_{2D}\sigma_{ij}(\vec{q},z) + \frac{\hbar}{N}\sum_{\vec{k}} h_{ji}(\vec{k},\zeta) n'(\vec{k},\zeta)\nu(\vec{k},\zeta;\vec{q},z)\right]. \tag{66}$$

As we can observe here, due to the anharmonic coupling $h_{ij}(\vec{k},\zeta)$, the flexural mode density fluctuations act as additional stresses on the in-plane lattice displacements.

V. FLEXURAL DENSITY DISTRIBUTION KINETICS

In order to derive a kinetic equation for the flexural density distribution $\nu(\vec{k},\zeta;\vec{q},z)$ defined by Eq. (63), we derive the equations of motion for the functions $\Gamma_i^Q(\vec{k},\zeta;\vec{q},z)$ and $\Gamma^\epsilon(\vec{k},\zeta;\vec{q},z)$. We start from the equation of motion for the correlation $G_j^Q(\vec{k},\zeta;\vec{q},z)$, Eq. (55), applying twice the identity (47). The hierarchy of higher-order GFs thereby obtained is then closed by well-known decoupling techniques [38]. Details of the calculation are given in the Appendix. As a result we obtain an integrodifferential equation for $\Gamma_j^Q(\vec{k},\zeta;\vec{q},z)$ where the displacement-displacement correlation $D_{kj}^Q(\vec{q},z)$ occurs as an inhomogeneous term:

$$[z - q_i v_i(\vec{k},\zeta)]\Gamma_j^Q(\vec{k},\zeta;\vec{q},z) + \frac{i\hbar z}{\sqrt{m}} q_i n'(\vec{k},\zeta) \times h_{ik}(\vec{k},\zeta) D_{kj}^Q(\vec{q},z) = -iC^{(1)}\Gamma_j^Q(\vec{k},\zeta;\vec{q},z). \tag{67}$$

Here $C^{(1)}$ stands for the collision operator of a linearized Peierls-Boltzmann equation,

$$C^{(1)}\Gamma_j^Q(\vec{k},\zeta;\vec{q},z) = R_j^{Q(1)} + R_j^{Q(2)} + R_j^{Q(3)}, \tag{68}$$

where

$$R_j^{Q(1)} = 2\pi\hbar\sum_{\vec{p},\vec{h},i}\left|\Psi^{(3)}\begin{pmatrix} i & \zeta & \zeta \\ \vec{p} & \vec{k} & \vec{h}\end{pmatrix}\right|^2 \delta(\omega(\vec{k},\zeta) - \omega(\vec{h},\zeta) + \omega(\vec{p},i))\{[n(\vec{p},i) - n(\vec{h},\zeta)]\Gamma_j^Q(\vec{k},\zeta;\vec{q},z) - [1 + n(\vec{p},i) + n(\vec{k},\zeta)]\Gamma_j^Q(\vec{h},\zeta;\vec{q},z) + [n(\vec{k},\zeta) - n(\vec{h},\zeta)]\Gamma_j^Q(\vec{p},i;\vec{q},z)\}, \tag{69}$$

$R_j^{Q(2)}$ is obtained from $R_j^{Q(1)}$ by interchanging $(\vec{h},\zeta)$ with $(\vec{p},i)$, and

$$R_j^{Q(3)} = 2\pi\hbar\sum_{\vec{p},\vec{h},k}\left|\Psi^{(3)}\begin{pmatrix} i & \zeta & \zeta \\ \vec{p} & \vec{k} & \vec{h}\end{pmatrix}\right|^2 \delta(\omega(\vec{k},\zeta) - \omega(\vec{h},\zeta) - \omega(\vec{p},i))\{[n(\vec{k},\zeta) - n(\vec{p},i)]\Gamma_j^Q(\vec{h},\zeta;\vec{q},z) + [1 + n(\vec{p},i) + n(\vec{h},\zeta)]\Gamma_j^Q(\vec{k},\zeta;\vec{q},z) - [n(\vec{h},\zeta) - n(\vec{k},\zeta)]\Gamma_j^Q(\vec{p},i;\vec{q},z)\}. \tag{70}$$

Here the quantity $\Psi^{(3)}$ has been defined by Eq. (A11), and $n(\vec{p},i) = [\exp(\hbar\omega(\vec{p},i)/k_BT) - 1]^{-1}$ denotes the in-plane equilibrium phonon density distribution. We notice that in addition to the flexural mode contribution $\Gamma_j^Q(\vec{k},\zeta;\vec{q},z)$, the collision term comprehends the in-plane counterpart,

$$\Gamma_j^Q(\vec{k},i;\vec{q},z) = \left\langle\left\langle b_{\vec{k}-\vec{q}}^{i\dagger}\, b_{\vec{k}}^{i}; Q\begin{pmatrix} j \\ -\vec{q}\end{pmatrix}\right\rangle\right\rangle_z \sqrt{N}. \tag{71}$$

As a consequence of collisions between flexural and in-plane phonons, also the latter are dragged out of equilibrium by the former. Notice that the definition of $\Gamma_j^Q(\vec{k},i;\vec{q},z)$ differs from $\Gamma_j^Q(\vec{k},\zeta;\vec{q},z)$, Eq. (56). With the anharmonic interaction $\Phi^{(3)}$, Eq. (17), there is no linear coupling between in-plane phonon density fluctuations and in-plane or out-of-plane displacements. The coupling via the collision term is a quadratic effect in the interaction, reminiscent of the well-known phonon drag due to electron-phonon coupling in metals [52].

In analogy with the equation of motion for $\Gamma_j^Q(\vec{k},\zeta;\vec{q},z)$, we have derived the corresponding equation for $\Gamma^\epsilon(\vec{k},\zeta;\vec{q},z)$ with the result

$$[z - q_i v_i(\vec{k},\zeta)]\Gamma^\epsilon(\vec{k},\zeta;\vec{q},z) + \frac{i\hbar z}{\sqrt{m}} n'(\vec{k},\zeta) q_i h_{ik}(\vec{k},\zeta) D_k^{Q\epsilon} \times (\vec{q},z) - n'(\vec{k},\zeta) q_i v_i(\vec{k},\zeta)\hbar\omega(\vec{k},\zeta) = -iC^{(1)}\Gamma^\epsilon(\vec{k},\zeta;\vec{q},z). \tag{72}$$

Here the collision term comprehends the in-plane contribution

$$\Gamma^\epsilon(\vec{k},i;\vec{q},z) = \langle\langle b_{\vec{k}-\vec{q}}^{i\dagger}\, b_{\vec{k}}^{i}; \epsilon(-\vec{q})\rangle\rangle_z \sqrt{N}, \tag{73}$$

and in analogy with Eq. (63) we define a nonequilibrium in-plane phonon density distribution $\nu(\vec{k},i;\vec{q},z)$ by

$$n'(\vec{k},i)\nu(\vec{k},i;q,z) = -\Gamma_j^s(\vec{k},i;\vec{q},z)F_j(\vec{q},z) - \Gamma^\epsilon(\vec{k},i;q,z)\frac{\Theta(\vec{q},z)}{T}, \tag{74}$$

with $n'(\vec{k},i) = n(\vec{k},i)[1 + n(\vec{k},i)]/k_BT$. Finally, we transform in Eqs. (67) and (72) from normal coordinates $Q(\vec{q},i)$ to center-of-mass displacement operators $s_i(\vec{q})$ by using again Eq. (15). We multiply the resulting equations for $\Gamma_l^s(\vec{k},i;\vec{q},z)$ and $\Gamma^\epsilon(\vec{k},i;q,z)$ by $F_l(\vec{q},z)$ and $\Theta(\vec{q},z)/T$, respectively, add the results, use Eqs. (63) and (74), and obtain the kinetic equation

$$[z - q_i v_i(\vec{k},\zeta)]\nu(\vec{k},\zeta;\vec{q},z) + i\hbar z q_i h_{ij}(\vec{k},\zeta)s_j(\vec{q},z) + q_i v_i(\vec{k},\zeta)\hbar\omega(\vec{k},\zeta)\frac{\Theta(\vec{q},z)}{T} = -iC^{(1)}\nu(\vec{k},\zeta;\vec{q},z). \quad (75)$$

Using the definition of the collision operator $C^{(1)}$, Eqs. (68)–(70), we obtain

$$C^{(1)}\nu(\vec{k},\zeta;\vec{q},z) = \frac{1}{n'(\vec{k},\zeta)}\sum_{\vec{p}\vec{h},i}\left\{W\begin{pmatrix}\zeta & \zeta & i\\ \vec{h} & \vec{k} & \vec{p}\end{pmatrix}[\nu(\vec{k},\zeta) - \nu(\vec{h},\zeta) + \nu(\vec{p},i)] + W\begin{pmatrix}i & \zeta & \zeta\\ \vec{p} & \vec{h} & \vec{k}\end{pmatrix}[\nu(\vec{k},\zeta) - \nu(\vec{p},i) + \nu(\vec{h},\zeta)] + W\begin{pmatrix}\zeta & i & \zeta\\ \vec{k} & \vec{p} & \vec{h}\end{pmatrix} \times [\nu(\vec{k},\zeta) - \nu(\vec{p},i) - \nu(\vec{h},\zeta)]\right\}. \quad (76)$$

Here it is understood that all quantities $\nu(\vec{k},\alpha)$ depend on $\vec{q},z$. The transition probabilities W are given by

$$W\begin{pmatrix}\zeta & \zeta & i\\ \vec{h} & \vec{k} & \vec{p}\end{pmatrix} = 2\pi\hbar\left|\Psi^{(3)}\begin{pmatrix}\zeta & \zeta & i\\ -\vec{h} & \vec{k} & \vec{p}\end{pmatrix}\right|^2 \times\sqrt{k_B T n'(\vec{k},\zeta)n'(\vec{p},i)n'(\vec{h},\zeta)} \times\delta(\omega(\vec{k},\zeta) - \omega(\vec{h},\zeta) + \omega(\vec{p},i)), \quad (77)$$

where $W(\vec{p}i,\vec{h}\zeta,\vec{k}\zeta)$ is obtained from $W(\vec{h}\zeta,\vec{k}\zeta,\vec{p}i)$ by interchanging $(\vec{p},i)$ with $(\vec{h},\zeta)$ and where

$$W\begin{pmatrix}\zeta & i & \zeta\\ \vec{k} & \vec{p} & \vec{h}\end{pmatrix} = 2\pi\hbar\left|\Psi^{(3)}\begin{pmatrix}\zeta & i & \zeta\\ -\vec{k} & \vec{p} & \vec{h}\end{pmatrix}\right|^2 \times\sqrt{k_B T n'(\vec{k},\zeta)n'(\vec{p},i)n'(\vec{h},\zeta)} \times\delta(\omega(\vec{k},\zeta) - \omega(\vec{p},i) - \omega(\vec{h},\zeta)). \quad (78)$$

For a complete description we still need a kinetic equation for the in-plane phonon density fluctuation $\nu(\vec{k},i;\vec{q},z)$ defined by Eq. (74). Proceeding as before, we have derived kinetic equations now for the functions $\Gamma^s_j(\vec{k},i;\vec{q},z)$ and $\Gamma^\epsilon(\vec{k},i;\vec{q},z)$, and taking into account Eq. (74) we get the kinetic equation

$$[z - q_i v_i(\vec{k},j)]\nu(\vec{k},j;\vec{q},z) + q_i v_i(\vec{k},j)\hbar\omega(\vec{k},j)\frac{\Theta(\vec{q},z)}{T} = -iC^{(2)}\nu(\vec{k},j;\vec{q},z). \quad (79)$$

Here the collision term $C^{(2)}\nu(\vec{k},j;\vec{q},z)$ is given by

$$C^{(2)}\nu(\vec{k},j;\vec{q},z) = \frac{1}{n'(\vec{k},j)}\sum_{\vec{p}_1\vec{p}_2}\left\{\frac{1}{2}W\begin{pmatrix}j & \zeta & \zeta\\ \vec{k} & \vec{p}_1 & \vec{p}_2\end{pmatrix} \times[\nu(\vec{k},j) - \nu(\vec{p}_1,\zeta) - \nu(\vec{p}_2,\zeta)] + W\begin{pmatrix}\zeta & j & \zeta\\ \vec{p}_1 & \vec{k} & \vec{p}_2\end{pmatrix} \times[\nu(\vec{k},j) - \nu(\vec{p}_1,\zeta) + \nu(\vec{p}_2,\zeta)]\right\}. \quad (80)$$

As already emphasized in the discussion following Eq. (71), in contradistinction with Eq. (75), there is no coupling in Eq. (79) with the in-plane or out-of-plane displacements.

VI. DISCUSSION AND COMPARISON WITH PHENOMENOLOGICAL THEORY

In Secs. IV and V we have derived the equation of motion (66) for the in-plane phonon displacement response $s_i(\vec{q},z)$ coupled to a kinetic equation (75) for the flexural phonon density fluctuations $\nu(\vec{k},\zeta;\vec{q},z)$. In addition, we have found that the in-plane phonon density is dragged out of equilibrium by collisions with flexural phonons. It is instructive to study these equations in the static response limit, obtained by taking $z = i\epsilon$, $\epsilon \to 0^+$ as prescribed in Sec. III. We recall that here again $\omega(\vec{k},\zeta)$ stands for the renormalized flexural mode frequency as specified in Sec. II. We define static wave-vector-dependent in-plane strains by

$$\epsilon_{ij}(\vec{q}) = iq_j s_i(\vec{q},z=0), \quad (81)$$

and denote the corresponding static stress tensor and the flexural distribution function by $\sigma_{ij}(\vec{q})$ and $\nu(\vec{k},\zeta;\vec{q})$, respectively. Then Eq. (66) reduces to

$$C_{ik,jl}\epsilon_{lj}(\vec{q}) = \sigma_{ik}(\vec{q}) + \frac{\hbar}{N v_{2D}}\sum_{\vec{k}} h_{ki}(\vec{k},\zeta)n'(\vec{k},\zeta)\nu(\vec{k},\zeta;\vec{q}). \quad (82)$$

Likewise, Eqs. (75) and (79) reduce to

$$q_i v_i(\vec{k},\zeta)\left[\hbar\omega(\vec{k},\zeta)\frac{\Theta(\vec{q})}{T} - \nu(\vec{k},\zeta;\vec{q})\right] = -iC^{(1)}\nu(\vec{k},\zeta;\vec{q}) \quad (83)$$

and

$$q_i v_i(\vec{k},j)\left[\hbar\omega(\vec{k},j)\frac{\Theta(\vec{q})}{T} - \nu(\vec{k},j;\vec{q})\right] = -iC^{(2)}\nu(\vec{k},j;\vec{q}). \quad (84)$$

Inspection shows that

$$\nu(\vec{k},\alpha;\vec{q}) = \hbar\omega(\vec{k},\alpha)\frac{\Theta(\vec{q})}{T} \quad (85)$$

is a solution of Eqs. (83) and (84) for $\alpha = \zeta$ and $\alpha = j$, respectively. Here we have taken into account that the collision integrals vanish due to energy conservation. Substitution of $\nu(\vec{k},\zeta;\vec{q})$ in Eq. (82) leads to the equation

$$C_{ik,jl}\epsilon_{lj}(\vec{q}) = \sigma_{ik}(\vec{q}) + \beta_{ik}(\zeta)\Theta(\vec{q}), \quad (86)$$

where

$$\beta_{ik}(\zeta) = \frac{\hbar^2}{N v_{2D}}\sum_{\vec{k}} h_{ki}(\vec{k},\zeta)n'(\vec{k},\zeta)\frac{\omega(\vec{k},\zeta)}{T} \quad (87)$$

is the tensor of thermal tension. Indeed, for a temperature change at fixed strains Eq. (86) gives

$$\frac{\partial \sigma_{ik}(\vec{q})}{\partial \Theta(\vec{q})} = -\beta_{ik}(\zeta). \tag{88}$$

For isothermal processes ($\Theta(\vec{q}) = 0$),

$$C_{ik,jl}\epsilon_{lj}(\vec{q}) = \sigma_{ik}(\vec{q}), \tag{89}$$

and we identify $C_{ik,jl}$ with the isothermal elastic constants. The rhs of Eq. (86) describes the response of the 2D crystal to wave-vector-dependent stress and temperature fields. Hence Eq. (86) is the generalization to the spatially nonuniform case of the thermal equation of state [39], now for a 2D crystal. The uniform situation is recovered by observing that for $\vec{q} \to 0$ one has $\epsilon_{ij}(\vec{q}) \to \sqrt{N}\epsilon_{ij}$, $\sigma_{ij}(\vec{q}) \to \sqrt{N}\sigma_{ij}$, and $\Theta(\vec{q}) \to \sqrt{N}\Theta$, where ϵ_{ij}, σ_{ij}, and Θ are uniform quantities. In the absence of external stresses, the relative change of the area of the unit cell due to a uniform temperature change Θ reads

$$\frac{d}{d\Theta}(\epsilon_{11} + \epsilon_{22}) = \frac{\beta_{11}(\zeta)}{B_{2D}} \equiv \alpha_T(\zeta), \tag{90}$$

where $B_{2D} = \gamma_{12} + \gamma_{66}$ is the 2D bulk modulus and $\alpha_T(\zeta)$ the thermal expansion coefficient. As mentioned in Sec. II, a negative value of the anharmonic coupling $h^{(3)}$ implies $h_{ii}(\vec{k},\zeta) < 0$ and $\beta_{11}(\zeta) < 0$, which leads to a negative contribution to the thermal expansion.

We now consider space- and time-dependent phenomena. In Sec. II we have studied, within the renormalized harmonic approximation, the change of the flexural mode frequency $\tilde{\omega}(\vec{k},\zeta)$ as a function of uniform static strains. Generalizing Eq. (30) to hydrodynamic space and time dependent deformations, we consider the modulation of the flexural mode frequency by an in-plane sound wave described by a displacement field $s_i(\vec{r},t)$ that varies slowly in space and time. The displaced flexural phonon frequency is given by

$$\tilde{\omega}(\vec{k},\zeta;\vec{r},t) = \omega(\vec{k},\zeta) + \delta\tilde{\omega}(\vec{k},\zeta;\vec{r},t), \tag{91}$$

where

$$\delta\tilde{\omega}(\vec{k},\zeta;\vec{r},t) = -h_{ij}(\vec{k},\zeta)\frac{\partial s_j}{\partial r_i}(\vec{r},t). \tag{92}$$

The corresponding local equilibrium density distribution reads

$$\tilde{n}(\vec{k},\zeta;\vec{r},t) = \frac{1}{\exp(\hbar\tilde{\omega}(\vec{k},\zeta;\vec{r},t)/k_B T) - 1}. \tag{93}$$

The true nonequilibrium density distribution is written as

$$n(\vec{k},\zeta;\vec{r},t) = \tilde{n}(\vec{k},\zeta;\vec{r},t) + n'(\vec{k},\zeta)\nu(\vec{k},\zeta;\vec{r},t), \tag{94}$$

where the second term on the rhs stands for the deviation from local equilibrium. Following the method of Ref. [35] we have derived for $n(\vec{k},\zeta;\vec{r},t)$ a linearized Peierls-Boltzmann equation with the result

$$\left(\frac{\partial}{\partial t} + v_i(\vec{k},\zeta)\frac{\partial}{\partial r_i}\right)\nu(\vec{k},\zeta;\vec{r},t) + \hbar h_{ij}(\vec{k},\zeta)\frac{\partial^2}{\partial t\partial r_i}s_j(\vec{r},t) - \hbar\omega(\vec{k},\zeta)v_i(\vec{k},\zeta)\frac{\partial}{\partial r_i}\frac{\Theta(\vec{r},t)}{T} = -C^{(1)}\nu(\vec{k},\zeta;\vec{r},t). \tag{95}$$

Here, the linearized collision term is a functional of the deviation from local equilibrium $\nu(\vec{k},\zeta;\vec{r},t)$. The Fourier transform of Eq. (95) then is found to agree with the kinetic Eq. (75) if there we take $z = \omega + i\epsilon$, $\epsilon \to 0^+$.

Likewise, the linearized equation of motion for the displacement field is given by the sound wave equation

$$\left(\frac{\partial^2}{\partial t^2} - \frac{v_{2D}}{m}C_{ik,jl}\frac{\partial^2}{\partial r_k\partial r_l}\right)s_j(\vec{r},t) = \frac{-1}{m}\frac{\partial}{\partial r_j}\left[v_{2D}\sigma_{ij}(\vec{r},t) + \frac{\hbar}{N}\sum_{\vec{k}} h_{ij}(\vec{k},\zeta)n'(\vec{k},\zeta)\nu(\vec{k},\zeta;\vec{r},t)\right]. \tag{96}$$

The corresponding Fourier transform agrees with Eq. (66).

VII. CONCLUDING REMARKS

We have presented a unified analytical theory of flexural phonon kinetics and hydrodynamic lattice deformations in 2D crystals. The central result of our paper is the system of symmetrically coupled dynamic equations (66) and (75), a wave equation for in-plane displacements, and a kinetic equation for the flexural phonon density distribution function. The thermoelastic coupling $h_{ij}(\vec{k},\zeta)$ between both equations is a direct consequence of the anharmonic interaction $\Phi^{(3)}$ that accounts for the scattering of flexural phonons with in-plane acoustic waves. The thermoelastic coupling leads to a change of the flexural mode frequency $\omega(\vec{k},\zeta)$ and is also at the origin of static phenomena like negative thermal expansion. In contradistinction to 3D crystals where the three acoustic modes have a linear dispersion at long wavelength, the anharmonic correction cannot be taken as a small perturbation to the harmonic flexural mode frequency. Hence we have treated the flexural mode in the renormalized harmonic approximation.

The physical meaning of the present theory can be summarized as follows: the modulation of the flexural mode frequency by an in-plane sound wave drives the flexural mode density distribution out of equilibrium and gives rise to a corresponding kinetic equation. Due to the thermoelastic coupling, the perturbed flexural density distribution produces in turn thermal stresses in the sound wave equation. Correspondingly, the in-plane acoustic displacements act as a perturbing field in the kinetic equation of the flexural mode. Since we have not considered cubic anharmonicities where three in-plane phonon interactions are involved, the in-plane phonon density distribution is not changed in lowest order anharmonicity by acoustic in-plane sound waves. However, as a quadratic anharmonic effect the in-plane phonons are dragged out of equilibrium by collisions with the flexural modes, a phenomenon reminiscent of phonon drag due to electron phonon collisions in metals [52].

The present analytical work is complementary to recent papers [25–27] on hydrodynamic phonon transport in 2D crystals where emphasis was put on the numerical solution of the Peierls-Boltzmann equation without coupling to lattice displacements as a moving background. While a thorough discussion of the solution of the coupled dynamic equations and a study of the resonances of the corresponding correlation functions will be presented in a following paper [53], we want to point out already here the possibility to investigate hydrodynamic phenomena by studying the resonances of the displacement-displacement correlations, now in 2D crystals, by optical methods. An extensive discussion of Brillouin light scattering from 3D crystals in the hydrodynamic regime has been given by Griffin [54].

ACKNOWLEDGMENTS

This work was supported by the Flemish Science Foundation (FWO-Vl) and the Methusalem program.

APPENDIX

In this Appendix we give some details about the derivation of the equations of motion of Secs. IV and V. The Hamiltonian of the system up to third-order anharmonicities is given by $H = H_{hR} + \Phi_R^{(3)}$. It is understood that $\omega(\vec{k},\zeta)$ stands for the renormalized flexural mode frequency $\omega_R(\vec{k},\zeta)$. We quote the commutation rules:

$$\big[b_{\vec{q}}^{i},H\big] = \hbar\omega(\vec{q},i)b_{\vec{q}}^{i} + \frac{1}{2}\sqrt{\frac{\hbar}{2\omega(\vec{q},i)}}\sum_{\vec{k}\vec{p}}\Phi^{(3)}\begin{pmatrix} i & \zeta & \zeta\\ -\vec{q} & \vec{k} & \vec{p}\end{pmatrix}Q\begin{pmatrix}\zeta\\ \vec{k}\end{pmatrix}Q\begin{pmatrix}\zeta\\ \vec{p}\end{pmatrix},$$

$$\big[b_{\vec{q}}^{i\dagger},H\big] = -\hbar\omega(\vec{q},i)b_{\vec{q}}^{i\dagger} - \frac{1}{2}\sqrt{\frac{\hbar}{2\omega(\vec{q},i)}}\sum_{\vec{k}\vec{p}}\Phi^{(3)}\begin{pmatrix} i & \zeta & \zeta\\ \vec{q} & \vec{k} & \vec{p}\end{pmatrix}Q\begin{pmatrix}\zeta\\ \vec{k}\end{pmatrix}Q\begin{pmatrix}\zeta\\ \vec{p}\end{pmatrix}. \tag{A1}$$

To get the equation of motion for the correlations $D_{ij}^{Q}(\vec{q},z)$, we start from Eq. (47) with $A = Q(\vec{q},i)$ and $B = Q(-\vec{q},j)$, use the commutation rules (A1), and obtain

$$zD_{ij}^{Q}(\vec{q},z) = \sqrt{\frac{\hbar\omega(\vec{q},i)}{2}}\left\langle\!\!\left\langle (b_{\vec{q}}^{i} - b_{-\vec{q}}^{i\dagger})\,;\,Q\begin{pmatrix} j\\ -\vec{q}\end{pmatrix}\right\rangle\!\!\right\rangle_z. \tag{A2}$$

Applying again Eq. (47) to the GF on the rhs, we obtain the exact result Eq. (49).

The equation of motion for the GFs in Eq. (50) are obtained again by means of Eq. (47) with $A = b_{-\vec{k}}^{\zeta\dagger}\, b_{-\vec{k}+\vec{q}}^{\zeta}$, $A = b_{\vec{k}}^{\zeta}\, b_{\vec{k}-\vec{q}}^{\zeta\dagger}$, etc., and the commutation rules Eq. (A1). We quote:

$$\begin{aligned}
&[z + \omega(\vec{k}-\vec{q},\zeta) - \omega(\vec{k},\zeta)]\left\langle\!\!\left\langle b_{\vec{k}-\vec{q}}^{\zeta\dagger} b_{\vec{k}}^{\zeta}\,;\,Q\begin{pmatrix} j\\ -\vec{q}\end{pmatrix}\right\rangle\!\!\right\rangle_z\\
&= -\sum_{i}\sum_{\vec{p}_1\vec{p}_2}\Bigg[\sqrt{\frac{1}{2\hbar\omega(\vec{k}-\vec{q},\zeta)}}\Phi^{(3)}\begin{pmatrix} i & \zeta & \zeta\\ \vec{p}_1 & \vec{k}-\vec{q} & \vec{p}_2\end{pmatrix}\left\langle\!\!\left\langle Q\begin{pmatrix} i\\ \vec{p}_1\end{pmatrix}Q\begin{pmatrix}\zeta\\ \vec{p}_2\end{pmatrix}b_{\vec{k}}^{\zeta}\,;\,Q\begin{pmatrix} j\\ -\vec{q}\end{pmatrix}\right\rangle\!\!\right\rangle_z\\
&\quad - \sqrt{\frac{1}{2\hbar\omega(\vec{k},\zeta)}}\Phi^{(3)}\begin{pmatrix} i & \zeta & \zeta\\ \vec{p}_1 & \vec{p}_2 & -\vec{k}\end{pmatrix}\left\langle\!\!\left\langle Q\begin{pmatrix} i\\ \vec{p}_1\end{pmatrix}b_{\vec{k}-\vec{q}}^{\zeta\dagger}Q\begin{pmatrix}\zeta\\ \vec{p}_2\end{pmatrix};Q\begin{pmatrix} j\\ -\vec{q}\end{pmatrix}\right\rangle\!\!\right\rangle_z\Bigg].
\end{aligned} \tag{A3}$$

The GFs on the rhs are then approximated by

$$\left\langle\!\!\left\langle Q\begin{pmatrix} i\\ \vec{p}_1\end{pmatrix}Q\begin{pmatrix}\zeta\\ \vec{p}_2\end{pmatrix}b_{\vec{k}}^{\zeta}\,;\,Q\begin{pmatrix} j\\ -\vec{q}\end{pmatrix}\right\rangle\!\!\right\rangle_z = n(\vec{k},\zeta)\sqrt{\frac{\hbar}{2\omega(\vec{k},\zeta)}}\delta_{-\vec{p}_2,\vec{k}}\left\langle\!\!\left\langle Q\begin{pmatrix} i\\ \vec{p}_1\end{pmatrix};Q\begin{pmatrix} j\\ -\vec{q}\end{pmatrix}\right\rangle\!\!\right\rangle_z,$$

$$\left\langle\!\!\left\langle Q\begin{pmatrix} i\\ \vec{p}_1\end{pmatrix}b_{\vec{k}-\vec{q}}^{\zeta\dagger}Q\begin{pmatrix}\zeta\\ \vec{p}_2\end{pmatrix};Q\begin{pmatrix} j\\ -\vec{q}\end{pmatrix}\right\rangle\!\!\right\rangle_z = n(\vec{k}-\vec{q},\zeta)\sqrt{\frac{\hbar}{2\omega(\vec{k}-\vec{q},\zeta)}}\delta_{\vec{p}_2,\vec{k}-\vec{q}}\left\langle\!\!\left\langle Q\begin{pmatrix} i\\ \vec{p}_1\end{pmatrix};Q\begin{pmatrix} j\\ -\vec{q}\end{pmatrix}\right\rangle\!\!\right\rangle_z. \tag{A4}$$

For small $\vec{q}$ the anharmonic force constants entail wave-vector conservation and hence $\vec{p}_1 = \vec{q}$. Then with Eqs. (A4), expression (A3) reduces to Eqs. (53) and (54). In a similar way we calculate the other GFs on the rhs of Eq. (50). With these approximations, Eq. (49) becomes a Dyson equation with self-energy Eq. (52). In order to proceed beyond the approximation Eq. (52) for $G_j^{Q}(\vec{k},\zeta;\vec{q},z)$, we write the GFs on the rhs of Eq. (A3) as sums of uncorrelated contributions (A4) and their cumulant parts. The latter, denoted by $\{...\}_z$, are defined by

$$\left\{Q\begin{pmatrix} i\\ \vec{p}_1\end{pmatrix}Q\begin{pmatrix}\zeta\\ \vec{p}_2\end{pmatrix}b_{\vec{k}}^{\zeta}\right\}_z \equiv \left\langle\!\!\left\langle Q\begin{pmatrix} i\\ \vec{p}_1\end{pmatrix}Q\begin{pmatrix}\zeta\\ \vec{p}_2\end{pmatrix}b_{\vec{k}}^{\zeta}\,;\,Q\begin{pmatrix} j\\ -\vec{q}\end{pmatrix}\right\rangle\!\!\right\rangle_z - n(\vec{k},\zeta)\sqrt{\frac{\hbar}{2\omega(\vec{k},\zeta)}}\,\delta_{-\vec{p}_2,\vec{k}}\,\delta_{\vec{p}_1,\vec{q}}\,D_{ij}^{Q}(\vec{q},z),$$

$$\left\{Q\begin{pmatrix} i\\ \vec{p}_1\end{pmatrix}b_{\vec{k}-\vec{q}}^{\zeta\dagger}\,Q\begin{pmatrix}\zeta\\ \vec{p}_2\end{pmatrix}\right\}_z \equiv \left\langle\!\!\left\langle Q\begin{pmatrix} i\\ \vec{p}_1\end{pmatrix}b_{\vec{k}-\vec{q}}^{\zeta\dagger}\,Q\begin{pmatrix}\zeta\\ \vec{p}_2\end{pmatrix};Q\begin{pmatrix} j\\ -\vec{q}\end{pmatrix}\right\rangle\!\!\right\rangle_z - n(\vec{k}-\vec{q},\zeta)\sqrt{\frac{\hbar}{2\omega(\vec{k}-\vec{q},\zeta)}}\delta_{\vec{p}_2,\vec{k}-\vec{q}}\,\delta_{\vec{p}_1,\vec{q}}\,D_{ij}^{Q}(\vec{q},z). \tag{A5}$$

With definition (56) for $\Gamma_j^Q(\vec{k},\zeta;\vec{q},z)$, Eq. (A3) is rewritten for small $\vec{q}$ as

$$[z - q_i v_i(\vec{k},\zeta)]\Gamma_j^Q(\vec{k},\zeta;\vec{q},z) + zq_i n'(\vec{k},\zeta)h_{ik}(\vec{k},\zeta)\frac{i\hbar}{\sqrt{m}}D_{kj}^Q(\vec{q},z)$$
$$= -\frac{1}{\sqrt{2\hbar\omega(\vec{k},\zeta)}}\sum_i\sum_{\vec{p}_1\vec{p}_2}\left[\Phi^{(3)}\begin{pmatrix} i & \zeta & \zeta \\ \vec{p}_1 & \vec{k}-\vec{q} & \vec{p}_2\end{pmatrix}\left\{Q\begin{pmatrix} i \\ \vec{p}_1\end{pmatrix}Q\begin{pmatrix} \zeta \\ \vec{p}_2\end{pmatrix}b_{\vec{k}}^{\zeta}\right\}_z - \Phi^{(3)}\begin{pmatrix} i & \zeta & \zeta \\ \vec{p}_1 & \vec{p}_2 & -\vec{k}\end{pmatrix}\left\{Q\begin{pmatrix} i \\ \vec{p}_1\end{pmatrix}b_{\vec{k}-\vec{q}}^{\zeta\dagger}Q\begin{pmatrix} \zeta \\ \vec{p}_2\end{pmatrix}\right\}_z\right], \tag{A6}$$

where $v_i(\vec{k},\zeta) = \partial\omega(\vec{k},\zeta)/\partial k_i$. Next we disentangle the cumulants using Eq. (10):

$$\left\{Q\begin{pmatrix} i \\ \vec{p}_1\end{pmatrix}Q\begin{pmatrix} \zeta \\ \vec{p}_2\end{pmatrix}b_{\vec{k}}^{\zeta}\right\}_z = \frac{\hbar}{2\sqrt{\omega(\vec{p}_1,i)\omega(\vec{p}_2,\zeta)}}\left[\{b_{\vec{k}_1}^i b_{\vec{p}_2}^{\zeta} b_{\vec{k}}^{\zeta}\}_z + \{b_{-\vec{p}_1}^{i\dagger} b_{\vec{p}_2}^{\zeta} b_{\vec{k}}^{\zeta}\}_z + \{b_{\vec{p}_1}^{i} b_{-\vec{p}_2}^{\zeta\dagger} b_{\vec{k}}^{\zeta}\}_z + \{b_{-\vec{p}_1}^{i\dagger} b_{-\vec{p}_2}^{\zeta\dagger} b_{\vec{k}}^{\zeta}\}_z\right]. \tag{A7}$$

The cumulants on the rhs are again defined by subtraction of the uncorrelated part from the GF, e.g.,

$$\{b_{\vec{p}_1}^{i} b_{-\vec{p}_2}^{\zeta\dagger} b_{\vec{k}}^{\zeta}\}_z = \left\langle\!\!\left\langle b_{\vec{p}_1}^{i}\, b_{-\vec{p}_2}^{\zeta\dagger}\, b_{\vec{k}}^{\zeta}\,; Q\begin{pmatrix} j \\ -\vec{q}\end{pmatrix}\right\rangle\!\!\right\rangle_z - n(\vec{k},\zeta)\delta_{-\vec{p}_2,\vec{k}}\left\langle\!\!\left\langle b_{\vec{p}_1}^{i}\,; Q\begin{pmatrix} j \\ -\vec{q}\end{pmatrix}\right\rangle\!\!\right\rangle_z, \tag{A8}$$

etc., for the other terms in (A7). In a similar way one treats the second term on the rhs of (A6), thereby obtaining cumulants such as $\{b_{\vec{p}_1}^{i} b_{\vec{k}-\vec{q}}^{\zeta\dagger} b_{\vec{p}_2}^{\zeta}\}_z$, etc. These cumulants satisfy in turn equations of motion viz.

$$[z - \omega(\vec{p}_1,i) + \omega(\vec{p}_2,\zeta) - \omega(\vec{k},\zeta)]\{b_{\vec{p}_1}^{i} b_{-\vec{p}_2}^{\zeta\dagger} b_{\vec{k}}^{\zeta}\}_z = \frac{1}{\hbar}\left[\left\langle\!\!\left\langle [b_{\vec{p}_1}^{i} b_{-\vec{p}_2}^{\zeta\dagger} b_{\vec{k}}^{\zeta},\Phi^{(3)}]\,; Q\begin{pmatrix} j \\ -\vec{q}\end{pmatrix}\right\rangle\!\!\right\rangle_z \right.$$
$$\left. - n(\vec{k},\zeta)\delta_{-\vec{p}_2,\vec{k}}\left\langle\!\!\left\langle [b_{\vec{p}_1}^{i},\Phi^{(3)}];\, Q\begin{pmatrix} j \\ -\vec{q}\end{pmatrix}\right\rangle\!\!\right\rangle_z\right]. \tag{A9}$$

Evaluation of the commutators gives higher-order GFs that we have to approximate by their uncorrelated parts. In addition, we take into account only the most singular functions, i.e., $\Gamma_j^Q(\vec{k},\zeta;\vec{q},z)$ and $\Gamma_j^Q(\vec{k},i;\vec{q},z)$, thereby obtaining, e.g.,

$$[z-\omega(\vec{p}_1,i) + \omega(\vec{p}_2,\zeta) - \omega(\vec{k},\zeta)]\{b_{\vec{p}_1}^{i} b_{-\vec{p}_2}^{\zeta\dagger} b_{\vec{k}}^{\zeta}\}_z = \sqrt{\hbar}\,\Psi^{(3)}\begin{pmatrix} i & \zeta & \zeta \\ -\vec{p}_1 & -\vec{p}_2 & -\vec{k}\end{pmatrix}\Xi^{(1)}(\vec{p}_1,i,\vec{p}_2,\zeta), \tag{A10}$$

with the definitions

$$\Psi^{(3)}\begin{pmatrix} i & \zeta & \zeta \\ -\vec{p}_1 & -\vec{p}_2 & -\vec{k}\end{pmatrix} = \frac{\Phi^{(3)}\begin{pmatrix} i & \zeta & \zeta \\ -\vec{p}_1 & -\vec{p}_2 & -\vec{k}\end{pmatrix}}{\sqrt{8\omega(\vec{p}_1,i)\omega(\vec{p}_2,\zeta)\omega(\vec{k},\zeta)}}, \tag{A11}$$

and

$$\Xi^{(1)}(\vec{p}_1,i,\vec{p}_2,\zeta) = [n(\vec{p}_1,i) - n(\vec{p}_2,\zeta)]\Gamma_j^Q(\vec{k},\zeta;\vec{q},z) - [1 + n(\vec{p}_1,i) + n(\vec{k},\zeta)]\Gamma_j^Q(\vec{p}_2,\zeta;\vec{q},z)$$
$$+ [n(\vec{k},\zeta) - n(\vec{p}_2,\zeta)]\Gamma_j^Q(\vec{k},i;\vec{q},z). \tag{A12}$$

Similar expressions are obtained for the second and fourth cumulant on the rhs of Eq. (A7). The cumulant $\{b_{\vec{p}_1}^{i} b_{\vec{p}_2}^{\zeta} b_{\vec{k}}^{\zeta}\}_z$ can be neglected since it does not yield singular contributions. In an analogous way we proceed with the cumulants obtained from the second term on the rhs of Eq. (A6). For instance, we obtain with $\vec{q} \Rightarrow 0$,

$$[z + \omega(\vec{k},\zeta) - \omega(\vec{p}_2,\zeta) + \omega(\vec{p}_1,i)]\{b_{-\vec{p}_1}^{i\dagger} b_{\vec{k}}^{\zeta\dagger} b_{\vec{p}_2}^{\zeta}\}_z = -\sqrt{\hbar}\tilde{\Phi}^{(3)}\begin{pmatrix} i & \zeta & \zeta \\ -\vec{p}_1 & \vec{k} & -\vec{p}_2\end{pmatrix}\Xi^{(1)}(\vec{p}_1,i,\vec{p}_2,\zeta). \tag{A13}$$

As we have shown, expressions (A10) and (A13) are complementary. Substituting both back into Eq. (A8) and using for $z = i\epsilon$, $\epsilon \to 0^+$ the identity

$$\frac{1}{X + i\epsilon} - \frac{1}{X - i\epsilon} = -2\pi i\delta(X), \tag{A14}$$

where $X = \omega(\vec{k},\zeta) - \omega(\vec{p}_2,\zeta) + \omega(\vec{p}_1,i)$, we obtain finally the contribution $R_j^{Q(1)}$, Eq. (67), to the collision term $C^{(1)}\Gamma_j^Q(\vec{k},\zeta;\vec{q},z)$. In a similar way we treat the remaining cumulants in (A6) and their complementary contributions, thereby obtaining contributions $R_j^{Q(2)}$ and $R_j^{Q(3)}$ to the collision term.

[1] A. K. Geim and K. S. Novoselov, The rise of graphene, Nat. Mater. **6**, 183 (2007).
[2] A. H. Castro Neto, F. Guinea, N. M. R. Peres, K. S. Novoselov, and A. K. Geim, The electronic properties of graphene, Rev. Mod. Phys. **81**, 109 (2009).
[3] C. Lee, X. Wei, J. W. Kysar, and J. Hone, Measurement of the elastic properties and intrinsic strength of monolayer graphene, Science **321**, 385 (2008).
[4] A. A. Balandin, S. Ghosh, W. Bao, I. Calizo, D. Teweldebrhan, F. Miao, and C. N. Lau, Superior thermal conductivity of single-layer graphene, Nano Lett. **8**, 902 (2008).
[5] D. Yoon, Y.-W. Son, and H. Cheong, Negative thermal expansion coefficient of graphene measured by Raman spectroscopy, Nano Lett. **11**, 3227 (2011).
[6] W. Bao, F. Miao, Z. Chen, H. Zhang, W. Jang, C. Dames, and C. N. Lau, Controlled ripple texturing of suspended graphene and ultrathin graphite membranes, Nat. Nanotechnol. **4**, 562 (2009).
[7] R. Saito, G. Dresselhaus, and M. S. Dresselhaus, *Physical Properties of Carbon Nanotubes* (Imperial College Press, London, 1998).
[8] I. M. Lifshitz, Thermal properties of chain and layered structures at low temperatures, Zh. Eksp. Teor. Fiz. **22**, 475 (1952).
[9] N. Mounet and N. Marzari, First-principles determination of the structural, vibrational and thermodynamic properties of diamond, graphite, and derivatives, Phys. Rev. B **71**, 205214 (2005).
[10] K. V. Zakharchenko, M. I. Katsnelson, and A. Fasolino, Finite Temperature Lattice Properties of Graphene Beyond the Quasiharmonic Approximation, Phys. Rev. Lett. **102**, 046808 (2009).
[11] B. Amorim, R. Roldán, E. Cappelluti, A. Fasolino, F. Guinea, and M. I. Katsnelson, Thermodynamics of quantum crystalline membranes, Phys. Rev. B **89**, 224307 (2014).
[12] P. M. Chaikin and T. C. Lubensky, *Principles of Condensed Matter Physics* (Cambridge University Press, Cambridge, UK, 2000).
[13] D. Nelson, T. Piran, and S. Weinberg, *Statistical Mechanics of Membranes and Surfaces*, 2nd ed. (World Scientific, Singapore, 2004).
[14] A. L. C. da Silva, L. Cndido, J. N. Teixeira Rabelo, G.-Q. Hai, and F. M. Peeters, Anharmonic effects on thermodynamic properties of a graphene monolayer, Europhys. Lett. **107**, 56004 (2014).
[15] K. H. Michel, S. Costamagna, and F. M. Peeters, Theory of anharmonic phonons in two-dimensional crystals, Phys. Rev. B **91**, 134302 (2015).
[16] K. H. Michel, S. Costamagna, and F. M. Peeters, Theory of thermal expansion in 2d crystals, Phys. Status Solidi B **252**, 2433 (2015).
[17] N. Bonini, J. Garg, and N. Marzari, Acoustic phonon lifetimes and thermal transport in free-standing and strained graphene, Nano Lett. **12**, 2673 (2012).
[18] L. Paulatto, F. Mauri, and M. Lazzeri, Anharmonic properties from a generalized third-order *ab initio* approach: Theory and applications to graphite and graphene, Phys. Rev. B **87**, 214303 (2013).
[19] J. H. Seol, I. Jo, A. L. Moore, L. Lindsay, Z. H. Aitken, M. T. Pettes, X. Li, Z. Yao, R. Huang, D. Broido, N. Mingo, R. S. Ruoff, and L. Shi, Two-dimensional phonon transport in supported graphene, Science **328**, 213 (2010).
[20] D. L. Nika, A. S. Askerov, and A. A. Balandin, Anomalous size dependence of the thermal conductivity of graphene ribbons, Nano Lett. **12**, 3238 (2012).
[21] D. L. Nika and A. A. Balandin, Two-dimensional phonon transport in graphene, J. Phys.: Condens. Matter **24**, 233203 (2012).
[22] Z. Wang, R. Xie, C. T. Bui, D. Liu, X. Ni, B. Li, and J. T. L. Thong, Thermal transport in suspended and supported few-layer graphene, Nano Lett. **11**, 113 (2011).
[23] L. Lindsay, D. A. Broido, and Natalio Mingo, Flexural phonons and thermal transport in graphene, Phys. Rev. B **82**, 115427 (2010).
[24] D. Singh, J. Y. Murthy, and T. S. Fisher, On the accuracy of classical and long wavelength approximations for phonon transport in graphene, J. Appl. Phys. **110**, 113510 (2011).
[25] S. Lee, D. Broido, K. Esfarjani, and G. Chen, Hydrodynamic phonon transport in suspended graphene, Nat. Commun. **6**, 6290 (2015).
[26] A. Cepellotti, G. Fugallo, L. Paulatto, M. Lazzeri, F. Mauri, and N. Marzari, Phonon hydrodynamics in two-dimensional materials, Nat. Commun. **6**, 6400 (2015).
[27] A. Cepellotti and N. Marzari, Thermal Transport in Crystals As a Kinetic Theory of Relaxons, Phys. Rev. X **6**, 041013 (2016).
[28] R. N. Gurzhi, Thermal conductivity of dielectrics and ferrodielectrics at low temperatures, Sov. Phys. JETP **19**, 490 (1964).
[29] E. W. Prohofsky and J. A. Krumhansl, Second-sound propagation in dielectric solids, Phys. Rev. **133**, A1403 (1964).
[30] R. A. Guyer and J. A. Krumhansl, Solution of the linearized phonon Boltzmann equation, Phys. Rev. **148**, 766 (1966).
[31] L. P. Mezhov-Deglin, Measurement of the thermal conductivity of crystalline He^4, Sov. Phys. JETP **22**, 47 (1966).
[32] C. C. Ackerman, B. Bertman, H. A. Fairbank, and R. A. Guyer, Second Sound in Solid Helium, Phys. Rev. Lett. **16**, 789 (1966).
[33] L. D. Landau and G. Placzek, Struktur der unverschobenen streulinie, Phys. Z. Sowjet **5**, 172 (1934); English translation, Structure of the undisplaced scattering line, in *Collected Papers of L. D. Landau* (Pergamon Press, New York, 1965).
[34] P. C. Kwok and P. C. Martin, Unified approach to interacting phonon problems, Phys. Rev. **142**, 495 (1966).
[35] W. Götze and K. H. Michel, First and second sound in crystals, Phys. Rev. **156**, 963 (1967).
[36] L. J. Sham, Equilibrium approach to second sound in solids, Phys. Rev. **156**, 494 (1967).
[37] D. N. Zubarev, Double-time Green functions in statistical physics, Usp. Fiz. Nauk **71**, 71 (1960).
[38] W. Götze and K. H. Michel, Two-fluid transport equations for lattices, Phys. Rev. **157**, 738 (1967).
[39] G. Leibfried and W. Ludwig, Theory of anharmonic effects in crystals, in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic Press, New York, 1961), Vol. 12.
[40] A. A. Maradudin and G. K. Horton, *Dynamical Properties of Solids* (North-Holland Publishing Company, Amsterdam, 1974), Vol. 1.
[41] R. E. Peierls, *Quantum Theory of Solids*, No. 23 (Oxford University Press, Oxford, UK, 1955).
[42] A. Fasolino, J. H. Los, and M. I. Katsnelson, Intrinsic ripples in graphene, Nat. Mater. **6**, 858 (2007).

[43] E. Mariani and F. von Oppen, Flexural Phonons in Free-Standing Graphene, Phys. Rev. Lett. **100**, 076801 (2008).

[44] G. Mie, Zur kinetischen Theorie der einatomigen Körper, Ann. Phys. (Berlin, Ger.) **316**, 657 (1903).

[45] E. Grüneisen, First and second sound in crystals, Ann. Phys. (Berlin, Ger.) **331**, 393 (1908).

[46] A. Akhiezer, On the absorption of sound in solids, J. Phys. USSR **1**, 277 (1939).

[47] T. O. Woodruff and H. Ehrenreich, Absorption of sound in insulators, Phys. Rev. **123**, 1553 (1961).

[48] H. E. Bömmel and K. Dransfeld, Excitation and attenuation of hypersonic waves in quartz, Phys. Rev. **117**, 1245 (1960).

[49] R. Kubo, Statistical-mechanical theory of irreversible processes, I. General theory and simple applications to magnetic and conduction problems, J. Phys. Soc. Jpn. **12**, 570 (1957).

[50] L. P. Kadanoff and P. C. Martin, Hydrodynamic equations and correlation functions, Ann. Phys. (NY) **24**, 419 (1963).

[51] W. Götze and K. H. Michel, Self-consistent phonons, in *Dynamical Properties of Solids*, edited by G. K. Horton and A. A. Maradudin (North-Holland Publishing Company, Amsterdam, 1974), pp. 499–540.

[52] J. M. Ziman, *Electrons and Phonons: The Theory of Transport Phenomena in Solids*, International Series of Monographs on Physics (Oxford University Press, Oxford, 1960).

[53] P. Scuracchio, K. H. Michel, and F. M. Peeters, Hydrodynamics and second sound in two-dimensional crystals (unpublished).

[54] A. Griffin, Brillouin light scattering from crystals in the hydrodynamic region, Rev. Mod. Phys. **40**, 167 (1968).