Hydrodynamic structure factor of quasicrystals

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A theory of the hydrodynamic structure factor for quasicrystals is developed and exploited. Based on the hydrodynamic equations for icosahedral quasicrystals we introduce the terms of dynamic correlation and response. The phononic and phasonic diffuse part of the dynamic structure factor are examined in detail in frequency and time domain. We present a complete set of solutions for the hydrodynamic equations. Out of the diffusive modes we separately study the phasonic diffusion, the anisotropy of the phasonic diffusion constants, and the general solution for phason wall diffusion. All results include phonon-phason coupling.

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I. INTRODUCTION

Quasicrystals are solids with long-range order, but of noncrystallographic, for example, fivefold or icosahedral symmetry. As immediate consequence they must be incommensurate. The Fourier transform of their mass density is a series in which there are more basic harmonics n than dimensions d of space. As d linear combinations of the phases of the basic harmonics describe (phononic) translations $\boldsymbol{u}$ in real space, the remaining $n-d$ combinations give rise to a novel degree of freedom, denoted "phase" or "phason."[1–4] An elegant way to tackle the phase is to lift the quasicrystalline structure to a periodic structure in an n-dimensional space $\mathbb{R}^n=\mathbb{E}^{\parallel}\oplus\mathbb{E}^{\perp}$, where it corresponds to a translation $\boldsymbol{w}$ in the orthogonal complement $\mathbb{E}^{\perp}$ of physical space $\mathbb{E}^{\parallel}$. It manifests itself as structural rearrangements by atomic flips among split positions. Together with the gradient $\nabla\boldsymbol{u}$ the gradient of the phase displacement $\nabla\boldsymbol{w}$ can be incorporated into a generalized phonon-phason elasticity theory.[4–6] Time-dependent long-wavelength excitations are treated with a generalized hydrodynamic theory, of which there are several versions. Here we apply the theory of Lubensky *et al.*[7]

Phason modes are involved in many dynamic processes of quasicrystals, e.g., phase transitions,[8] diffusion,[9,10] dislocation motion, and crack propagation.[11] Direct observations of phason oscillations were made by Edagawa *et al.*[12] Phason fluctuations become most evident in quasielastic neutron-scattering experiments. They display asymmetric shoulders of the Bragg peaks that Jarić *et al.*[13] had predicted by performing calculations of the *static* structure factor, and they allow to measure absolute values of the phason elastic constants.

Here we will generalize the theory of Jarić *et al.* to determine the phonon and phason hydrodynamic structure factor. The phason part has not yet been measured as it is somewhat distant in frequency from the window accessible by neutrons. But as more and more quasicrystalline structures outside the traditional metal class are discovered or synthesized, e.g., in polymers,[14] liquid crystals,[15] or colloids,[16] experiments on the phason hydrodynamic structure factor might become feasible in the future. They would give access to hydrodynamic material constants as viscosities and kinetic coefficients. Here however, we restrict to metals exclusively. A short account on some features of the hydrodynamic structure factor, which we were able to reproduce, was given in by Ishii in 1998.[17] We concentrate on single-component icosahedral quasicrystals.

Phason diffusion constants were measured in the decay of phason walls that follow dislocations, and in speckle patterns.[18–20] The latter depend directly on the phason modes in reciprocal-space representation, of which we also will give a detailed account.

The paper is organized as follows: in Sec. II we will derive the hydrodynamic structure factor from the continuum representation of a quasicrystal mass density, for which the dynamic susceptibility is required. This response function is obtained from the hydrodynamic equations and is evaluated for icosahedral quasicrystals. The main properties of phonons and phasons will be detected in the diffuse part of the dynamic structure factor. In Sec. III the solutions of the hydrodynamic equations are listed, of which the phasonic diffusion will be separately studied in Sec. IV.

II. DYNAMIC STRUCTURE FACTOR

A. Expression for $S(\boldsymbol{k},\omega)$

The time-dependent mass density of a distorted quasicrystal is given by

$$\rho(\boldsymbol{x}^{\parallel},t)=\sum_{\boldsymbol{\kappa}\in\tilde{\mathcal{G}}}\rho_{\boldsymbol{\kappa}}e^{i[\boldsymbol{k}^{\parallel}\cdot\boldsymbol{x}^{\parallel}-\boldsymbol{\kappa}\cdot\boldsymbol{\gamma}(\boldsymbol{x}^{\parallel},t)]},\tag{2.1}$$

where $\boldsymbol{\gamma}$ denotes the phonon-phason displacement vector

$$\boldsymbol{\gamma}(\boldsymbol{x}^{\parallel},t)=\boldsymbol{u}(\boldsymbol{x}^{\parallel},t)\oplus\boldsymbol{w}(\boldsymbol{x}^{\parallel},t)=\begin{pmatrix}\boldsymbol{u}(\boldsymbol{x}^{\parallel},t)\\ \boldsymbol{w}(\boldsymbol{x}^{\parallel},t)\end{pmatrix},\tag{2.2}$$

and $\boldsymbol{\kappa}$ the vectors of the reciprocal hyperlattice $\tilde{\mathcal{G}}$,

$$\boldsymbol{\kappa}=\boldsymbol{k}^{\parallel}\oplus\boldsymbol{k}^{\perp}=\begin{pmatrix}\boldsymbol{k}^{\parallel}\\ \boldsymbol{k}^{\perp}\end{pmatrix}.\tag{2.3}$$

Note, that $\boldsymbol{\gamma}$ depends only on the physical coordinate $\boldsymbol{x}^{\parallel}=:\boldsymbol{x}$. The density-density correlation function is defined as

$$C_{\rho\rho}(\boldsymbol{x},t):=\lim_{V\to\infty}\frac{1}{V}\int_V d^3\tilde{x}\lim_{T\to\infty}\frac{1}{T}\int_{-T/2}^{T/2}d\tilde{t}\langle\rho(\tilde{\boldsymbol{x}},\tilde{t})\rho^*(\tilde{\boldsymbol{x}}-\boldsymbol{x},\tilde{t}-t)\rangle$$
$$=\sum_{\boldsymbol{\kappa}\in\tilde{\mathcal{G}}}|\rho_{\boldsymbol{\kappa}}|^2e^{i\boldsymbol{k}^{\parallel}\cdot\boldsymbol{x}}\langle e^{-i\boldsymbol{\kappa}\cdot[\boldsymbol{\gamma}(\boldsymbol{x},t)-\boldsymbol{\gamma}(0,0)]}\rangle.\tag{2.4}$$

The thermal average is performed in the canonical ensemble. The dynamic structure factor is the spatial and temporal Fourier transform of this correlation function,

$$S(\boldsymbol{k},\omega) := \int d^3x \int dt\, C_{\rho\rho}(\boldsymbol{x},t) e^{i\omega t - i\boldsymbol{k}\cdot\boldsymbol{x}}. \tag{2.5}$$

In the harmonic case Eq. (2.4) can be expanded,

$$\langle e^A \rangle = e^{1/2\langle A^2\rangle} \approx 1 + \frac{1}{2}\langle A^2\rangle + \cdots, \tag{2.6}$$

and we have to average over the phonon-phason displacements $\gamma_\alpha(\boldsymbol{x},t)\gamma_\beta(\boldsymbol{0},0)$. The result is given by the generalized fluctuation-dissipation theorem

$$\langle \gamma_\alpha(\boldsymbol{x},t)\gamma_\beta(\boldsymbol{x}',t')\rangle = \int \frac{d\omega}{2\pi} e^{-i\omega(t-t')} 2k_B T \frac{\chi''_{\alpha\beta}(\boldsymbol{x},\boldsymbol{x}',\omega)}{\omega}, \tag{2.7}$$

see Chaikin and Lubensky.[21] $\chi''_{\alpha\beta}$ denotes the imaginary part of the dynamic response function $\chi_{\alpha\beta}(\boldsymbol{x},\boldsymbol{x}',\omega)$. For the dynamic structure factor follows,

$$S(\boldsymbol{k},\omega) = \sum_{\boldsymbol{\kappa}\in\tilde{\mathcal{G}}} |\rho_{\boldsymbol{\kappa}}|^2 e^{-2W(\boldsymbol{\kappa})} \times \left\{ (2\pi)^4 \delta(\boldsymbol{k}-\boldsymbol{k}^\parallel)\delta(\omega) + k_B T \boldsymbol{\kappa}^t \frac{2\underline{\boldsymbol{\chi}}''(\boldsymbol{k}-\boldsymbol{k}^\parallel,\omega)}{\omega}\boldsymbol{\kappa} \right\}, \tag{2.8}$$

with the Debye-Waller factor,

$$2W(\boldsymbol{\kappa}) := \kappa_\alpha \langle \gamma_\alpha(\boldsymbol{x},0)\gamma_\beta(\boldsymbol{x},0)\rangle \kappa_\beta = \kappa_\alpha \int \frac{d^3q}{(2\pi)^3} k_B T \chi_{\alpha\beta}(\boldsymbol{q})\kappa_\beta, \tag{2.9}$$

where we use the Einstein summation convention. The last equality follows directly from Eq. (2.7) and the thermodynamic sum rule,[21,22]

$$\chi_{\alpha\beta}(\boldsymbol{x},\boldsymbol{x}') = \lim_{\omega\to 0} \chi_{\alpha\beta}(\boldsymbol{x},\boldsymbol{x}',\omega) = \int \frac{d\omega}{\pi} \frac{\chi''_{\alpha\beta}(\boldsymbol{x},\boldsymbol{x}',\omega)}{\omega}, \tag{2.10}$$

where $\chi_{\alpha\beta}(\boldsymbol{x},\boldsymbol{x}')$ is the static susceptibility. Hence the static structure factor $S(\boldsymbol{k})$ results as an integral over Eq. (2.8),

$$S(\boldsymbol{k}) = \int \frac{d\omega}{2\pi} S(\boldsymbol{k},\omega) = \sum_{\boldsymbol{\kappa}\in\tilde{\mathcal{G}}} |\rho_{\boldsymbol{\kappa}}|^2 e^{-2W(\boldsymbol{\kappa})} \{ (2\pi)^3 \delta(\boldsymbol{k}-\boldsymbol{k}^\parallel) + k_B T \boldsymbol{\kappa}^t \underline{\boldsymbol{\chi}}(\boldsymbol{k}-\boldsymbol{k}^\parallel)\boldsymbol{\kappa} \}. \tag{2.11}$$

B. Dynamic response function

The main quantity in the expression of the dynamic structure factor in Eq. (2.8) is the dynamic response function $\underline{\boldsymbol{\chi}}(\boldsymbol{x},\boldsymbol{x}',\omega)$, which depends only on the difference $\boldsymbol{x}-\boldsymbol{x}'$ due to translational invariance. Its Fourier transform $\underline{\tilde{\boldsymbol{\chi}}}(\boldsymbol{x}-\boldsymbol{x}',t-t')$ describes how the displacements $\boldsymbol{\gamma}(\boldsymbol{x},t) = (\boldsymbol{u},\boldsymbol{w})^t$ vary in the presence of external volume forces $\boldsymbol{b}(\boldsymbol{x}',t') = (\boldsymbol{b}^\parallel,\boldsymbol{b}^\perp)^t$,

$$\boldsymbol{\gamma}(\boldsymbol{x},t) = \int dt' \int d^3x' \underline{\tilde{\boldsymbol{\chi}}}(\boldsymbol{x}-\boldsymbol{x}',t-t')\boldsymbol{b}(\boldsymbol{x}',t'), \tag{2.12}$$

or after spatial and temporal Fourier transformation,

$$\boldsymbol{\gamma}(\boldsymbol{q},\omega) = \underline{\boldsymbol{\chi}}(\boldsymbol{q},\omega)\boldsymbol{b}(\boldsymbol{q},\omega). \tag{2.13}$$

It therefore serves as Green's function. Such a linear relation between forces and displacements can be obtained via the linearized hydrodynamic equations of the icosahedral quasicrystal,[7]

$$-i\omega\delta\rho(\boldsymbol{q},\omega) = -i\boldsymbol{q}\cdot\boldsymbol{g}, \tag{2.14a}$$

$$-i\omega\boldsymbol{g}(\boldsymbol{q},\omega) = -\underline{\boldsymbol{\eta}}(\boldsymbol{q})\boldsymbol{g} - i(A-B)\frac{\boldsymbol{q}}{\rho_0}\delta\rho - \underline{\tilde{\mathbf{N}}}(\boldsymbol{q})\boldsymbol{u} - \underline{\mathbf{C}}(\boldsymbol{q})\boldsymbol{w} + \boldsymbol{b}^\parallel(\boldsymbol{q},\omega), \tag{2.14b}$$

$$-i\omega\boldsymbol{u}(\boldsymbol{q},\omega) = \frac{\boldsymbol{g}}{\rho_0} - \Gamma_u\left[-iB\frac{\boldsymbol{q}}{\rho_0}\delta\rho + \underline{\mathbf{N}}(\boldsymbol{q})\boldsymbol{u} + \underline{\mathbf{C}}(\boldsymbol{q})\boldsymbol{w}\right] + \Gamma_u\boldsymbol{b}^\parallel(\boldsymbol{q},\omega), \tag{2.14c}$$

$$-i\omega\boldsymbol{w}(\boldsymbol{q},\omega) = -\Gamma_w[\underline{\mathbf{M}}(\boldsymbol{q})\boldsymbol{w} + \underline{\mathbf{C}}^t(\boldsymbol{q})\boldsymbol{u}] + \Gamma_w\boldsymbol{b}^\perp(\boldsymbol{q},\omega). \tag{2.14d}$$

In addition to the phonon and phason displacements $\boldsymbol{u}(\boldsymbol{q},\omega)$ and $\boldsymbol{w}(\boldsymbol{q},\omega)$, the hydrodynamic variables of this system of equations are the momentum density $\boldsymbol{g}(\boldsymbol{q},\omega)$ and the change in mass density, given by $\delta\rho(\boldsymbol{q},\omega)$ with the quiescent density ρ_0. For icosahedral symmetry we obtain five elastic constants. The phononic block $\underline{\mathbf{N}}(\boldsymbol{q})$ depends on the two Lamé constants λ and μ. The matrix $\underline{\mathbf{C}}(\boldsymbol{q})$ describes the coupling between phonons and phasons with the corresponding coupling constant K_3 while K_1, K_2 are two pure phasonic elastic constants of the phasonic matrix $\underline{\mathbf{M}}(\boldsymbol{q})$. A^{-1} is the compressibility while B couples the density to quasilattice dilations. $\underline{\boldsymbol{\eta}}(\boldsymbol{q})$ indicates the tensor of viscosity. The two kinetic coefficients Γ_u and Γ_w are the diffusion mobilities for vacancies and phasons. For the definition of these matrices and the elastic constants see Appendix A. We arrive at Eq. (2.13) by eliminating the momentum density $\mathbf{g}(\mathbf{q},\omega)$ in Eq. (2.14c). The reduced system takes the form

$$\boldsymbol{\gamma}(\boldsymbol{q},\omega) = \underbrace{[i\omega^3\underline{\mathbf{A}} - \omega^2\underline{\mathbf{B}}(\boldsymbol{q}) - i\omega\underline{\boldsymbol{\Gamma}}(\boldsymbol{q}) - \underline{\boldsymbol{\Delta}}(\boldsymbol{q})]^{-1}[-\omega^2\Gamma_u\underline{\mathbf{A}} - i\omega\underline{\mathbf{E}}(\boldsymbol{q}) - \underline{\mathbf{Z}}(\boldsymbol{q})]}_{\equiv\underline{\boldsymbol{\chi}}(\boldsymbol{q},\omega)}\boldsymbol{b}(\boldsymbol{q},\omega), \tag{2.15}$$

with the 6×6 matrices $\underline{\mathbf{A}}$, $\underline{\mathbf{B}}(\boldsymbol{q})$, $\underline{\boldsymbol{\Gamma}}(\boldsymbol{q})$, $\underline{\boldsymbol{\Delta}}(\boldsymbol{q})$, $\underline{\mathbf{E}}(\boldsymbol{q})$, and $\underline{\mathbf{Z}}(\boldsymbol{q})$ which are listed in Appendix B.

C. Phononic diffuse part

The first term of the dynamic structure factor in Eq. (2.8) corresponds to the usual Bragg reflections, while the second one describes the diffuse broadening, resulting from the phonon-phason fluctuations. We want to study the shape of these diffuse shoulders and their dependence on the great number of parameters separately for phonons and phasons before we change from frequency to time domain.

Around the Bragg peak $\boldsymbol{k}^{\parallel}$ the phononic diffuse part is proportional to

$$\mathcal{D}_{\text{pho}}(\boldsymbol{q},\boldsymbol{k}^{\parallel},\omega) \coloneqq \boldsymbol{k}^{\parallel t}\frac{2\underline{\boldsymbol{\chi}}''_{uu}(\boldsymbol{q},\omega)}{\omega}\boldsymbol{k}^{\parallel} \tag{2.16}$$

with $\boldsymbol{q}\coloneqq\boldsymbol{k}-\boldsymbol{k}^{\parallel}$ and the phononic block of the dynamic response function $\underline{\boldsymbol{\chi}}_{uu}(\boldsymbol{q},\omega)$ given by Eq. (B7a). In a first step we examine the decoupled case. For $K_3=0$ Eq. (2.16) reduces to

$$\begin{aligned}\mathcal{D}^0_{\text{pho}}(\boldsymbol{q},\boldsymbol{k}^{\parallel},\omega) &\coloneqq \boldsymbol{k}^{\parallel t}\frac{2\underline{\boldsymbol{\chi}}^{0\prime\prime}_{uu}(\boldsymbol{q},\omega)}{\omega}\boldsymbol{k}^{\parallel}\\ &= \left[\frac{2\chi^{0\prime\prime}_L(q,\omega)}{\omega}-\frac{2\chi^{0\prime\prime}_T(q,\omega)}{\omega}\right]\frac{(\boldsymbol{k}^{\parallel t}\cdot\boldsymbol{q})^2}{q^2}\\ &\quad+\frac{2\chi^{0\prime\prime}_T(q,\omega)}{\omega}\boldsymbol{k}^{\parallel 2},\end{aligned} \tag{2.17}$$

where we have used the response function without coupling $\underline{\boldsymbol{\chi}}^0_{uu}(\boldsymbol{q},\omega)$ from Eq. (B8). χ^0_L is the longitudinal part of the tensor $\underline{\boldsymbol{\chi}}^0_{uu}$ with respect to $\boldsymbol{q}$, χ^0_T the transversal. Both quantities are defined in Appendix B and depend only on $q=|\boldsymbol{q}|$. We intend to plot Eq. (2.17) as function of $\boldsymbol{q}$ centered around $\boldsymbol{k}^{\parallel}$. The two terms describe a superposition of a dipolelike shape and a sphere. This sum depends on the nine parameters ρ_0, μ, $\Lambda\coloneqq 2\mu+\lambda$, ω, Γ_u, η_L, η_T, A, and B of the prefactors. For icosahedral AlPdMn we take for these constants the following values,

$$\rho_0 = 5080\frac{\text{kg}}{\text{m}^3},\quad \lambda = 85\ \text{GPa},\quad \mu = 65\ \text{GPa},$$

$$\Gamma_u=\Gamma_w = 1\times 10^{-25}\frac{\text{m}^3\ \text{s}}{\text{kg}},\quad \eta_L=\eta_T = 1\frac{\text{cm}^2}{\text{s}}\quad\text{and}$$

$$A = B = 1\ \text{GPa}. \tag{2.18}$$

The mass density ρ_0 of AlPdMn is known.[23] The Lamé constants λ and μ have been determined by Amazit *et al.*[23] via sound velocity measurements. From the phasonic diffusion constant, measured by Francoual *et al.*,[20] follows the value of the kinetic coefficient Γ_w. Their relation will be come evident in the next section. Since the two diffusion mobilities describe the same hopping mechanism for vacancies and phasons, we assume Γ_u to be of the same order as Γ_w. For the kinetic viscosities η_L and η_T we use the value given by

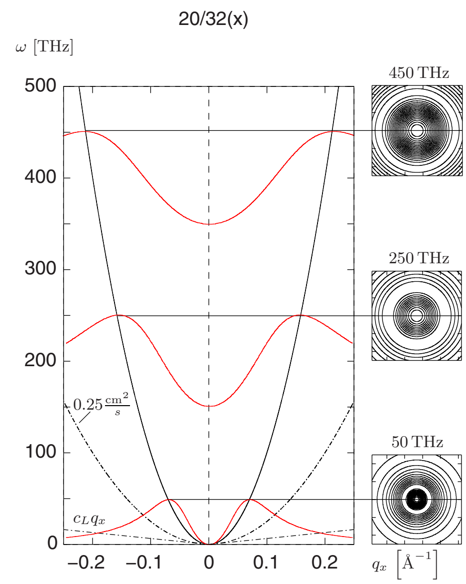

FIG. 1. (Color online) The intensity distribution of the phononic diffuse part for the 20/32(x) reflection along the twofold direction for three different frequencies. The minima are located at $q_x=0$ while the maxima lie on the curve given by Eq. (2.20). For smaller values of the kinetic viscosity, such as $\eta_L=0.25\ \text{cm}^2/\text{s}$, $q_{x,\max}$ approaches the linear-dispersion relation $c_L q_x$.

Lubensky *et al.*[7] while for the yet undetermined constants A and B we make the above assumption. Inserting these values in Eq. (2.17), the phononic isointensity contours in the q_xq_y plane result in concentric circles with the center at $\boldsymbol{k}^{\parallel}$, as the second term in Eq. (2.17) dominates, being two orders larger than the first one. This is illustrated on the right side of Fig. 1 for the 20/32(x) reflection, labeled by its N/M indices,[24] and for three different frequencies ω. If we consider the intensity distribution along this twofold direction, setting $q_y=0$, Eq. (2.17) leads to the wavy curves in Fig. 1, which show a local minimum at $q_x=0$ of value

$$\mathcal{D}^0_{\text{pho}}(\boldsymbol{0},\boldsymbol{k}^{\parallel},\omega) = \frac{2\Gamma_u}{\omega^2}k_x^{\parallel 2}. \tag{2.19}$$

Also two global maxima exist at the positions

$$q_{x,\max} = \pm\frac{\omega}{(\omega^2 d^{0\,2}_L + c_L^4)^{1/4}}. \tag{2.20}$$

This expression is a generalized dispersion relation containing the damping constant

$$d^0_L \coloneqq \eta_L + \frac{\Gamma_u}{\rho_0 c_L^2}(\Lambda - B)^2 \tag{2.21}$$

with $\rho_0 c_L^2=\Lambda+A-2B$. c_L denotes the longitudinal sound velocity. Note that Eq. (2.20) is an approximative solution since we omitted higher-order terms of Γ_u in Eq. (B8). For $d^0_L=0$ Eq. (2.20) is equal to the well-known linear-dispersion relation $q_{x,\max}=\pm\omega/c_L$. In the overdamped case and for high frequencies $q_{x,\max}$ simplifies to $q_{x,\max}=\pm\sqrt{\omega/d^0_L}$. The frequency dependence of $q_{x,\max}$ and the resulting intensities for different frequencies are also shown in Fig. 1. This behavior of the phononic diffuse part is valid for all values of ω, Γ_u, η_L, η_T, A, and B (the rest ones we took as fixed) and for all other reflections. Whether maxima and minima can be resolved separately from each other depends on the damping parameters and primarily on Γ_u. For $\Gamma_u > 1 \times 10^{-16}$ m^3 s/kg the fine structure is lost and maxima and minimum merge to one big maximum. The shape of the isointensity contours also changes and looks like the one of the static structure factor.[25] Therefore making Γ_u larger causes a zooming out.

We now switch on K_3 and at the same time simplify the expression of $\underline{\chi}_{uu}(\boldsymbol{q},\omega)$ in Eq. (2.16). As already mentioned in Sec. I the displayed isointensity contours are out of the measurable frequency window for neutron scattering. Hence we consider the time-dependent structure factor $S(\boldsymbol{k},t)$. Neglecting diffusive and higher-order terms of $\boldsymbol{q}$, Eq. (B7a) splits into a transverse and longitudinal part

$$\underline{\chi}_{uu}(\boldsymbol{q},\omega) = \underline{\chi}^T_{uu}(\boldsymbol{q},\omega) + \underline{\chi}^L_{uu}(\boldsymbol{q},\omega), \tag{2.22}$$

similar to the case of zero coupling. The transverse response function takes the form

$$\underline{\chi}^T_{uu}(\boldsymbol{q},\omega) = \underline{\hat{\chi}}^T_{uu}(\boldsymbol{q},\omega)\underline{\mathbf{P}} = \frac{1}{\rho_0}\left\{-\omega^2 - i\omega\left[\eta_T + \Gamma_u\mu + \frac{\Gamma_w}{\mu q^4}\underline{\mathbf{PCC}^t}\right]q^2 + c_T^2 q^2\right\}^{-1}\underline{\mathbf{P}}, \tag{2.23}$$

or written in spectral representation:

$$\underline{\chi}^T_{uu}(\boldsymbol{q},\omega) = \sum_{\alpha=1}^{3}\frac{1/\rho_0}{-\omega^2 - i\omega d_{T,\alpha}q^2 + c_T^2 q^2}(\hat{\boldsymbol{v}}_\alpha \otimes \hat{\boldsymbol{v}}_\alpha)\underline{\mathbf{P}}, \tag{2.24}$$

where we define

$$d_{T,\alpha} \coloneqq \eta_T + \Gamma_u\mu + \frac{\Gamma_w}{\mu}\frac{\lambda_\alpha}{q^4}, \quad \alpha = 1,2,3, \tag{2.25}$$

with eigenvalues $\lambda_\alpha(\boldsymbol{q})$ (two nonzero ones) and eigenvectors $\hat{\boldsymbol{v}}_\alpha(\boldsymbol{q})$ of the matrix $\underline{\mathbf{P}}\,\underline{\mathbf{C}}\,\underline{\mathbf{C}}^t$. The matrix $\underline{\mathbf{P}} \coloneqq \underline{\mathbf{1}} - \hat{\boldsymbol{q}}\otimes\hat{\boldsymbol{q}}$ projects a vector transverse to the direction of $\hat{\boldsymbol{q}}$. $d_{T,\alpha}$ describes a constant damping for the transverse modes while $c_T \coloneqq \sqrt{\mu/\rho_0}$ denotes the transverse velocity of sound. For the longitudinal response function we obtain

$$\underline{\chi}^L_{uu}(\boldsymbol{q},\omega) = \chi^L_{uu}(q,\omega)\underline{\mathbf{Q}} = \frac{1/\rho_0}{-\omega^2 - i\omega d_L q^2 + c_L^2 q^2}\underline{\mathbf{Q}} \tag{2.26}$$

with the damping constant

$$d_L \coloneqq \eta_L + \frac{\Gamma_u}{\rho_0 c_L^2}(\Lambda - B)^2 + \frac{\Gamma_w}{\rho_0 c_L^2}\frac{\mathrm{Tr}[\underline{\mathbf{QCC}^t}]}{q^4}. \tag{2.27}$$

This is the full version of Eq. (2.21) containing now an additional coupling term, which is about four orders smaller than the second one. $\underline{\mathbf{Q}} \coloneqq \hat{\boldsymbol{q}}\otimes\hat{\boldsymbol{q}}$ is the longitudinal projection operator. The dynamic diffuse part for phonons results as

$$\begin{aligned}\mathcal{D}_{\mathrm{pho}}(\boldsymbol{q},\boldsymbol{k}^{\parallel},\omega) &\coloneqq \boldsymbol{k}^{\parallel t}\frac{2\underline{\chi}^{T\prime\prime}_{uu}(\boldsymbol{q},\omega)}{\omega}\boldsymbol{k}^{\parallel} + \boldsymbol{k}^{\parallel t}\frac{2\underline{\chi}^{L\prime\prime}_{uu}(\boldsymbol{q},\omega)}{\omega}\boldsymbol{k}^{\parallel}\\ &= \sum_{\alpha=1}^{3}\frac{d_{T,\alpha}q^2/\rho_0}{(c_T^2q^2-\omega^2)^2 + \omega^2 d_{T,\alpha}^2 q^4}\boldsymbol{k}^{\parallel t}(\hat{\boldsymbol{v}}_\alpha\otimes\hat{\boldsymbol{v}}_\alpha)\underline{\mathbf{P}}\boldsymbol{k}^{\parallel}\\ &\quad + \frac{d_L q^2/\rho_0}{(c_L^2q^2-\omega^2)^2 + \omega^2 d_L^2 q^4}(\boldsymbol{k}^{\parallel t}\cdot\hat{\boldsymbol{q}})^2. \end{aligned}\tag{2.28}$$

Since the zeros of the denominators of $\underline{\chi}^{T\prime\prime}_{uu}/\omega$ and $\underline{\chi}^{L\prime\prime}_{uu}/\omega$ are the propagative dispersion relations, computed first by Lubensky *et al.*,[7] the inverse temporal Fourier transformation can be performed via residue theorem

$$\begin{aligned}\mathcal{D}_{\mathrm{pho}}(\boldsymbol{q},\boldsymbol{k}^{\parallel},t) &= \sum_{\alpha=1}^{3}\frac{e^{-(1/2)d_{T,\alpha}q^2|t|}}{\rho_0 c_T^3 q^3}\Big[c_T q\cos(c_T q|t|)\\ &\quad + \frac{1}{2}d_{T,\alpha}q^2\sin(c_T q|t|)\Big](\boldsymbol{k}^{\parallel t}\cdot\hat{\boldsymbol{v}}_\alpha)[(\hat{\boldsymbol{v}}^t_\alpha\cdot\boldsymbol{k}^{\parallel})\\ &\quad - (\hat{\boldsymbol{v}}^t_\alpha\cdot\hat{\boldsymbol{q}})(\hat{\boldsymbol{q}}^t\cdot\boldsymbol{k}^{\parallel})]\\ &\quad + \frac{e^{-(1/2)d_L q^2|t|}}{\rho_0 c_L^3 q^3}\Big[c_L q\cos(c_L q|t|)\\ &\quad + \frac{1}{2}d_L q^2\sin(c_L q|t|)\Big](\boldsymbol{k}^{\parallel t}\cdot\hat{\boldsymbol{q}})^2. \end{aligned}\tag{2.29}$$

According to the thermodynamic sum rule, Eq. (2.10), for $t=0$ we obtain the static diffuse part $\mathcal{D}_{\mathrm{pho}}(\boldsymbol{q},\boldsymbol{k}^{\parallel})$ with the two static susceptibilities $\underline{\hat{\chi}}^T_{uu}(\boldsymbol{q},\omega=0) = 1/\rho_0 c_T^2 q^2$ and $\chi^L_{uu}(q,\omega=0) = 1/\rho_0 c_L^2 q^2$. The time dependence of $\mathcal{D}_{\mathrm{pho}}(\boldsymbol{q},\boldsymbol{k}^{\parallel},t)$ takes the form of a damped oscillation. As we can see by the expressions for $d_{T,\alpha}$ and d_L the phason coupling only causes a decay of the propagative phonons in addition to the kinetic viscosities and the kinetic coefficients.

D. Phasonic diffuse part

The dynamic phasonic diffuse part for a single reciprocal-lattice vector $\boldsymbol{k}^{\perp}$ follows without Debye-Waller factor as

$$\mathcal{D}_{\mathrm{pha}}(\boldsymbol{q},\boldsymbol{k}^{\perp},\omega) \coloneqq \boldsymbol{k}^{\perp t}\frac{2\underline{\chi}''_{ww}(\boldsymbol{q},\omega)}{\omega}\boldsymbol{k}^{\perp}. \tag{2.30}$$

The dynamic response function $\underline{\chi}_{ww}(\boldsymbol{q},\omega)$ from Eq. (B7c) is equal to $\underline{\chi}_{ww}(\boldsymbol{q},\omega) = \Gamma_w\underline{\mathbf{S}}^{-1}$ with the stiffness matrix $\underline{\mathbf{S}}(\boldsymbol{q},\omega)$,

$$\underline{\mathbf{S}}(\boldsymbol{q},\omega) = [-i\omega\underline{\mathbf{1}} + \Gamma_w(\underline{\mathbf{M}} - \chi_L^0\underline{\mathbf{C}}^t\underline{\mathbf{QC}} - \chi_T^0\underline{\mathbf{C}}^t\underline{\mathbf{PC}})]. \tag{2.31}$$

As in the previous section χ_L^0 and χ_T^0 are the longitudinal and transverse part of the tensor $\underline{\chi}^0_{uu}$ with respect to $\boldsymbol{q}$, see Eq. (B9). For zero coupling Eq. (2.31) reduces to

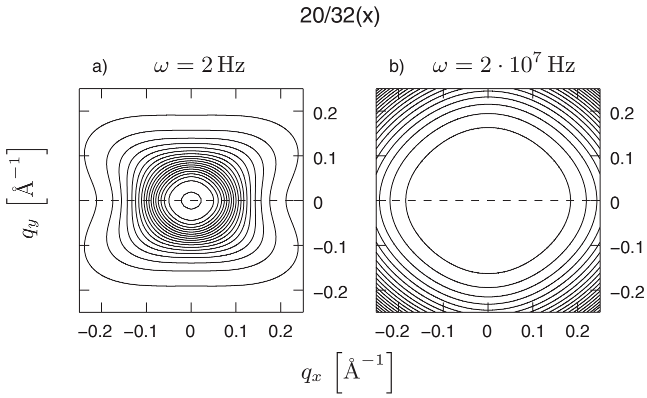

FIG. 2. The zooming effect of the frequency ω illustrated on the phasonic isointensity contours of the 20/32(x) twofold reflection. Since the form of the contours for small values is the same anisotropic as in the static case, for higher ones they take an almost circular shape.

$$\underline{\mathbf{S}}^0(\boldsymbol{q},\omega) = -i\omega\underline{\mathbf{1}} + \Gamma_w\underline{\mathbf{M}}(\boldsymbol{q}), \tag{2.32}$$

and we obtain for $\mathcal{D}^0_{\text{pha}}(\boldsymbol{q},\boldsymbol{k}^\perp,\omega)$,

$$\mathcal{D}^0_{\text{pha}}(\boldsymbol{q},\boldsymbol{k}^\perp,\omega) = \boldsymbol{k}^{\perp t}\frac{2\Gamma_w}{\omega^2+\Gamma_w^2\underline{\mathbf{M}}(\boldsymbol{q})^2}\boldsymbol{k}^\perp = \sum_{i=1}^{3}\frac{2\Gamma_w}{\omega^2+\Gamma_w^2M_i^2q^4}(\boldsymbol{k}^\perp\cdot\hat{\boldsymbol{e}}_i)^2. \tag{2.33}$$

$M_i(\varphi,\vartheta)q^2$ are the eigenvalues and $\hat{\boldsymbol{e}}_i(\varphi,\vartheta)$ the eigenvectors of $\underline{\mathbf{M}}(\boldsymbol{q})$. The product $\Gamma_wM_i(K_1,K_2)=:D_i^0$ gives us, as we will see in the next section, the phasonic diffusion coefficients, which depend on the phasonic elastic constants K_1, K_2 and on the direction of $\boldsymbol{q}=q\hat{\boldsymbol{q}}(\varphi,\vartheta)$. From this relation and by measurements of D_i^0 we can get the values of the kinetic coefficients Γ_w and Γ_u of Eq. (2.18). The shape of the phasonic isointensity contours is governed by the four parameters ω, Γ_w, K_1, and K_2. The two phasonic elastic constants K_1 and K_2, whose ratios are actually responsible for the form,[19,26] have been determined by measurements of the static structure factor. For our computations we take the values specified by Létoublon *et al.*:[27] $K_1=16.2$ MPa and $K_2=-8.4$ MPa for room temperature. Computing $\mathcal{D}^0_{\text{pha}}(\boldsymbol{q},\boldsymbol{k}^\perp,\omega)$ for these values we obtain one maximum at $\boldsymbol{q}=\mathbf{0}$ for all frequencies ω and kinetic coefficients Γ_w. The form of the phasonic isointensity contours also stays the same. Rising ω merely causes a zooming into the unaltered shape (see Fig. 2) while increasing Γ_w shows exactly the opposite behavior. To quantify this broadening we consider the maximum value at $\boldsymbol{q}=\mathbf{0}$,

$$\mathcal{D}^0_{\text{pha}}(\mathbf{0},\boldsymbol{k}^\perp,\omega) = \frac{2\Gamma_w}{\omega^2}\boldsymbol{k}^{\perp 2}, \tag{2.34}$$

which is also valid with coupling, and the contour at half maximum

$$q(\varphi) = \sqrt{\frac{\omega}{\Gamma_w}}F(\varphi,K_1,K_2,\boldsymbol{k}^\perp), \quad q=\sqrt{q_x^2+q_y^2}. \tag{2.35}$$

We obtained this result due to the simple frequency dependence of $\mathcal{D}^0_{\text{pha}}$ in Eq. (2.33). $F(\varphi,K_1,K_2,\boldsymbol{k}^\perp)$ is a function of the M_i. For the maximum positions of the phononic diffuse part in the overdamped case we also obtain the root ratio of frequency and damping constant. In Eq. (2.35) this damping parameter is the kinetic coefficient Γ_w. Eqs. (2.19) and (2.34) are formally the same.

For $K_3\neq 0$ we now neglect in the inverse Fourier transform the propagative terms of the expressions for χ_T^0 and χ_L^0, see Eq. (B9). The stiffness matrix $\underline{\mathbf{S}}(\boldsymbol{q},\omega)$ from Eq. (2.31) simplifies to

$$\underline{\mathbf{S}}(\boldsymbol{q},\omega) = -i\omega\underline{\mathbf{1}} + \Gamma_w\underline{\tilde{\mathbf{M}}}(\mathbf{q}), \tag{2.36}$$

where we have defined

$$\underline{\tilde{\mathbf{M}}}(\boldsymbol{q}) := \underline{\mathbf{M}}(\boldsymbol{q}) - \frac{\underline{\mathbf{C}}^t\underline{\mathbf{Q}}\underline{\mathbf{C}}}{\rho_0c_L^2q^2} - \frac{\underline{\mathbf{C}}^t\underline{\mathbf{P}}\underline{\mathbf{C}}}{\rho_0c_T^2q^2} \propto q^2. \tag{2.37}$$

In analogy to Eq. (2.33) the phasonic diffuse part around $\boldsymbol{k}^\parallel$ takes the form

$$\mathcal{D}_{\text{pha}}(\boldsymbol{q},\boldsymbol{k}^\perp,\omega) = \boldsymbol{k}^{\perp t}\frac{2\Gamma_w}{\omega^2+\Gamma_w^2\underline{\tilde{\mathbf{M}}}(\boldsymbol{q})^2}\boldsymbol{k}^\perp = \sum_{i=1}^{3}\frac{2\Gamma_w}{\omega^2+\Gamma_w^2\tilde{M}_i^2q^4}(\mathbf{k}^\perp\cdot\hat{\boldsymbol{e}}_i)^2. \tag{2.38}$$

$\tilde{M}_i(\varphi,\vartheta)q^2$ are now the eigenvalues and $\hat{\boldsymbol{e}}_i(\varphi,\vartheta)$ the eigenvectors of $\underline{\tilde{\mathbf{M}}}(\mathbf{q})$. The product $\Gamma_w\tilde{M}_i=:D_i$ is the corresponding phasonic diffusion coefficient with coupling. Looking at Eq. (2.38), $\mathcal{D}_{\text{pha}}$ shows the same frequency dependence as without coupling. The prefactor in Eq. (2.35) is therefore still valid for $K_3\neq 0$. For the inverse Fourier transformation of the Lorentz curve in Eq. (2.38) we obtain an exponential decay in time

$$\mathcal{D}_{\text{pha}}(\boldsymbol{q},\boldsymbol{k}^\perp,t) = \boldsymbol{k}^{\perp t}\underline{\tilde{\mathbf{M}}}(\boldsymbol{q})^{-1}e^{-\Gamma_w\underline{\tilde{\mathbf{M}}}(\boldsymbol{q})|t|}\boldsymbol{k}^\perp = \sum_{i=1}^{3}\frac{1}{\tilde{M}_iq^2}e^{-\Gamma_w\tilde{M}_iq^2|t|}(\boldsymbol{k}^\perp\cdot\hat{\boldsymbol{e}}_i)^2. \tag{2.39}$$

The matrix $\underline{\tilde{\mathbf{M}}}(\boldsymbol{q})$ is positive definite.

Using this result for $\mathcal{D}_{\text{pha}}(\boldsymbol{q},\boldsymbol{k}^\perp,t)$, we can establish a connection with the speckle patterns, measured by coherent x-ray spectroscopy.[20,28] The intensity-intensity correlation function $I(\boldsymbol{q},t)$ is

$$I(\boldsymbol{q},t) = 1 + \beta g(\boldsymbol{q},t), \tag{2.40}$$

where $g(\boldsymbol{q},t)$ characterizes the time dependence of the fluctuations. β is the degree of coherence of the beam. For phason fluctuations the function $g(\boldsymbol{q},t)$ is proportional to the squared ratio of the dynamic and static phasonic diffuse part

$$g(\boldsymbol{q},t) \propto \left[\frac{\mathcal{D}_{\text{pha}}(\boldsymbol{q},\boldsymbol{k}^{\perp},t)}{\mathcal{D}_{\text{pha}}(\boldsymbol{q},\boldsymbol{k}^{\perp})}\right]^2 = \left[\frac{\sum_{i=1}^{3}\frac{1}{\tilde{M}_i q^2}e^{-\Gamma_w \tilde{M}_i q^2|t|}(\boldsymbol{k}^{\perp}\cdot\hat{\boldsymbol{e}}_i)^2}{\sum_{j=1}^{3}\frac{1}{\tilde{M}_j q^2}(\boldsymbol{k}^{\perp}\cdot\hat{\boldsymbol{e}}_j)^2}\right]^2. \tag{2.41}$$

III. SOLVING THE SYSTEM OF HYDRODYNAMIC EQUATIONS

The hydrodynamic mode structure of the linearized equations of motion has been studied up to now only for the propagating modes.[7] After a brief recapitulation we present solutions also for the diffusive modes.

A. Solution for the propagating modes

Equations (2.14a), (2.14b), and (2.14c) split into a transverse and longitudinal part of $\hat{\boldsymbol{q}}$, denoted by the subscripts T and L. Considering propagating phonons with $\omega \propto q$, diffusive terms, which are of the next order of wave number, can be neglected. Equation (2.14d) becomes in the propagating regime without forces

$$\boldsymbol{w}(\boldsymbol{q},\omega) = \frac{\Gamma_w \underline{\mathbf{C}}^t}{i\omega}\boldsymbol{u}(\boldsymbol{q},\omega). \tag{3.1}$$

With this expression and after some manipulations we get decoupled equations for $\mathbf{u}_T$ and $\boldsymbol{u}_L = u_L\hat{\boldsymbol{q}}$. For the transverse mode we obtain

$$\left\{-\omega^2 - i\omega\left[\eta_T + \Gamma_u\mu + \frac{\Gamma_w}{\mu q^4}\underline{\mathbf{PCC}}^t\right]q^2 + c_T^2 q^2\right\}\boldsymbol{u}_T = \boldsymbol{b}_T^{\parallel}/\rho_0. \tag{3.2}$$

This equation describes a damped oscillation. The homogeneous solution ($\boldsymbol{b}_T^{\parallel}=0$) is

$$\boldsymbol{u}_T^{\text{hom}}(\boldsymbol{q},t) = 2\sum_{\alpha=1}^{3} c_\alpha \hat{\boldsymbol{v}}_\alpha \cos(c_T q t)e^{-(1/2)d_{T,\alpha}q^2 t}, \tag{3.3}$$

where c_α is a constant depending on initial conditions. As already seen in the section before, the oscillation is given by the transverse velocity of sound c_T, the damping by $d_{T,\alpha}$. In the presence of external forces Eq. (3.2) can be written with the transverse response function (2.24),

$$\boldsymbol{u}_T(\boldsymbol{q},\omega) = \underline{\hat{\boldsymbol{\chi}}}_{uu}^T(\boldsymbol{q},\omega)\boldsymbol{b}_T^{\parallel}(\boldsymbol{q},\omega). \tag{3.4}$$

The particular solution follows as the inverse temporal Fourier transformation of this product,

$$\boldsymbol{u}_T^{\text{par}}(\boldsymbol{q},t) = \sum_{\alpha=1}^{3}\int_{-\infty}^{t} dt' \frac{\sin[c_T q(t-t')]}{\rho_0 c_T q} \times e^{-(1/2)d_{T,\alpha}q^2(t-t')}[\hat{\boldsymbol{v}}_\alpha^t \cdot \boldsymbol{b}_T^{\parallel}(\boldsymbol{q},t')]\hat{\boldsymbol{v}}_\alpha. \tag{3.5}$$

The longitudinal mode, too, is given by a damped oscillation

$$[-\omega^2 - i\omega d_L q^2 + c_L^2 q^2]u_L = \frac{b_L^{\parallel}}{\rho_0} \tag{3.6}$$

with the damping constant d_L and the longitudinal sound velocity c_L. The homogeneous solution follows as:

$$u_L^{\text{hom}}(q,t) = e^{-(1/2)d_L q^2 t}[c_1\cos(c_L q t) + c_2\sin(c_L q t)], \tag{3.7}$$

where the constants c_1 and c_2 are determined by initial conditions. For $b_L^{\parallel} \neq 0$, Eq. (3.6), is equal to

$$u_L(q,\omega) = \chi_{uu}^L(q,\omega)b_L^{\parallel}(q,\omega), \tag{3.8}$$

with the longitudinal response function $\chi_{uu}^L(q,\omega)$ from Eq. (2.26). As we have seen before the particular solution in time space takes the form of a convolution

$$u_L^{\text{par}}(q,t) = \sum_{\alpha=1}^{3}\int_{-\infty}^{t} dt' \frac{\sin[c_L q(t-t')]}{\rho_0 c_L q} \times e^{-(1/2)d_L q^2(t-t')}b_L^{\parallel}(q,t'). \tag{3.9}$$

The corresponding phason field $\boldsymbol{w}(\boldsymbol{q},t)$ for the homogeneous solution of $\boldsymbol{u}=\boldsymbol{u}_T+\boldsymbol{u}_L$ is obtained by inverse Fourier transformation of Eq. (3.1),

$$\frac{\partial \boldsymbol{w}}{\partial t}(\boldsymbol{q},t) = -\Gamma_w \underline{\mathbf{C}}^t \boldsymbol{u}(\boldsymbol{q},t). \tag{3.10}$$

B. Solution for the diffusive modes

When solving the system of hydrodynamic equations for the remaining four diffusive modes one has to neglect all propagative and inertial terms. We obtain an expression analog to Eq. (3.1), which now relates $\boldsymbol{u}$ with $\boldsymbol{w}$ in the diffusive regime

$$\boldsymbol{u}(\boldsymbol{q},t) = -\left(\frac{\underline{\mathbf{Q}}}{\rho_0 c_L^2} + \frac{\underline{\mathbf{P}}}{\rho_0 c_T^2}\right)\frac{\underline{\mathbf{C}}}{q^2}\boldsymbol{w}(\boldsymbol{q},t). \tag{3.11}$$

Inserting this equation in Eq. (2.14d) we obtain

$$[-i\omega\underline{\mathbf{1}} + \Gamma_w \underline{\tilde{\mathbf{M}}}(\boldsymbol{q})]\boldsymbol{w}(\boldsymbol{q},\omega) = \Gamma_w \boldsymbol{b}^{\perp}(\boldsymbol{q},\omega) \tag{3.12}$$

with the phasonic matrix $\underline{\tilde{\mathbf{M}}}(\boldsymbol{q})$ from Eq. (2.37). As we will discuss in detail in the next section, Eq. (3.12) describes an anisotropic diffusion equation for phasons. Considering the case $\boldsymbol{b}^{\perp}=\mathbf{0}$, we get

$$\boldsymbol{w}^{\text{hom}}(\boldsymbol{q},t) = \sum_{i=1}^{3} e^{-\Gamma_w \tilde{M}_i q^2 t}[\hat{\boldsymbol{e}}_i^t \cdot \boldsymbol{w}(\boldsymbol{q},0)]\hat{\boldsymbol{e}}_i, \quad t>0, \tag{3.13}$$

where the diffusion coefficients result as $\Gamma_w \tilde{M}_i =: D_i$ and the initial condition is given by $\boldsymbol{w}(\boldsymbol{q},0)$. Expressing Eq. (3.12) with the phasonic dynamic response function

$$\boldsymbol{w}(\boldsymbol{q},\omega) = \underline{\boldsymbol{\chi}}_{ww}(\boldsymbol{q},\omega)\boldsymbol{b}^{\perp}(\boldsymbol{q},\omega), \tag{3.14}$$

the particular solution follows as convolution

$$\boldsymbol{w}^{\text{par}}(\boldsymbol{q},t) = \sum_{i=1}^{3} \int_{-\infty}^{t} dt' e^{-\Gamma_w \tilde{M}_i q^2 (t-t')} [\hat{\boldsymbol{e}}_i^t \cdot \boldsymbol{b}^{\perp}(\boldsymbol{q},t')] \hat{\boldsymbol{e}}_i. \quad (3.15)$$

One diffusive mode still remains, namely, the one for the vacancy diffusion. Eliminating g_L in Eq. (2.14a) we obtain a diffusion equation of the form

$$[-i\omega + D_{\delta\rho} q^2] \delta\rho(\boldsymbol{q},\omega) = 0, \quad (3.16)$$

with the diffusion constant $D_{\delta\rho}$,

$$D_{\delta\rho} = \Gamma_u \frac{\Lambda A - B^2}{\Lambda - B}, \quad (3.17)$$

and the solution

$$\delta\rho(\boldsymbol{q},t) = e^{-D_{\delta\rho} q^2 t} \delta\rho(\boldsymbol{q},0), \quad t > 0. \quad (3.18)$$

IV. PHASON DIFFUSION

In this section we present a special solution of the phasonic diffusion equation. Before we go into detail, we first have to compute the eigenvalues $\tilde{M}_i q^2$ of the phasonic matrix $\underline{\tilde{\mathbf{M}}}(\boldsymbol{q})$. A first investigation was already given by Ishii.[29]

A. Directionality of the diffusion coefficients

As already mentioned in the previous section the diffusion coefficients $D_i(\varphi,\vartheta) \coloneqq \Gamma_w \tilde{M}_i(\varphi,\vartheta)$ depend on the direction $\boldsymbol{q} = q\hat{\boldsymbol{q}}(\varphi,\vartheta)$. In order to simplify matters we study their directionality on the $q_x q_y$-twofold plane and determine the eigenvalues of $\underline{\tilde{\mathbf{M}}}(\boldsymbol{q})$ for $q_z = 0 \Leftrightarrow \vartheta = \pi/2$. The result is shown in Fig. 3. For $K_3 = 0$ the diffusion constants D_i^0 take maximum and minimum values along the threefold, fivefold, and twofold axes. Two of them become degenerate along the threefold and fivefold axis. The corresponding eigenvectors along these directions span a plane, in which the anisotropy is lost. Only for the twofold direction we get three different diffusion coefficients: $D_1^0 \neq D_2^0 \neq D_3^0$. Switching on the coupling, the values D_i^0 diminish and additional intersections and extrema occur for higher K_3. An exceptional position takes the fivefold axis. Along this direction two diffusion constants never change their value and decouple from K_3. For our computations we use the values of K_1 and K_2 given in Sec. II D. From these and the stability criteria,[30,31] the maximum value of the coupling constant follows as $K_{3,\max} = 0.77$ GPa. At this value two modes become soft.

Along the twofold axis $\boldsymbol{q}_y = q_y \hat{\boldsymbol{e}}_y$ the diffusion coefficients D_i and the eigenvectors $\hat{\boldsymbol{e}}_i$ take the form

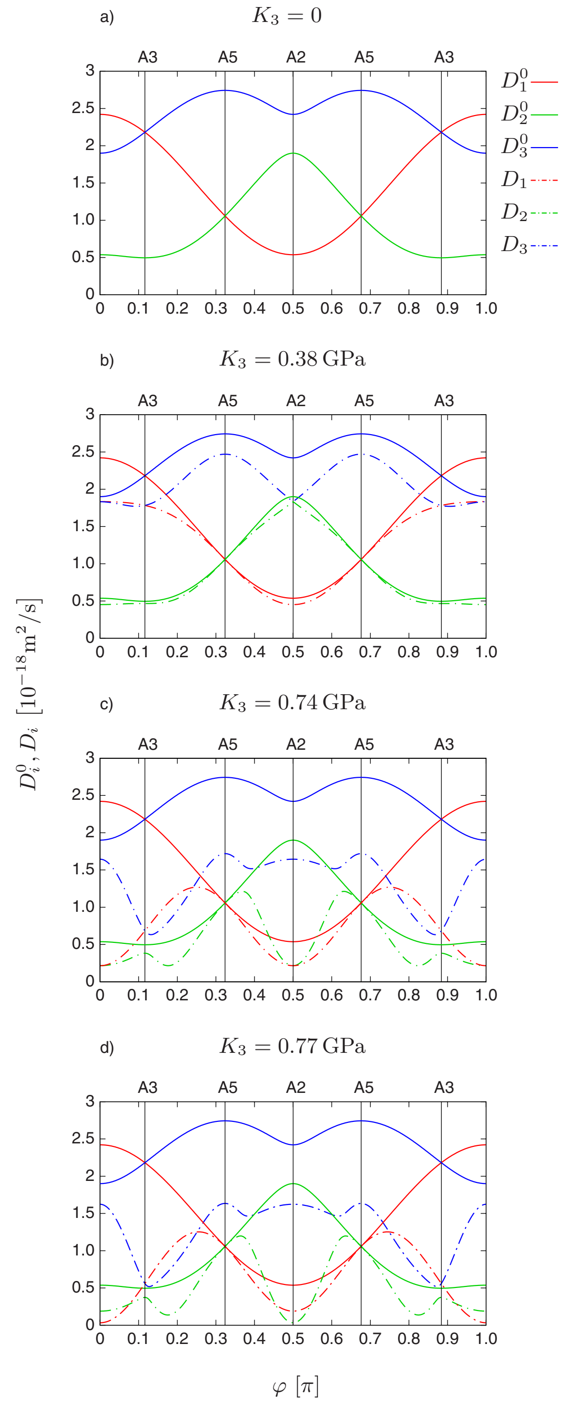

FIG. 3. (Color online) The directionality of the diffusion coefficients without and with coupling, D_i^0 and D_i, as a function of the polar angle φ. An increase in K_3 lowers the coefficients. There are systematic degeneracies along the threefold and fivefold axes, the intersection point of the latter being independent of K_3. At the maximum value of K_3 a softening occurs.

$$D_1 = \Gamma_w \left[K_1 + \left(\tau - \frac{1}{3} \right) K_2 - \frac{K_3^2}{\mu \tau^2} \right], \quad \hat{\boldsymbol{e}}_1 = (0,0,1)^t,$$

$$D_2 = \Gamma_w \left[K_1 - \frac{K_2}{3} - \frac{K_3^2}{\rho_0 c_L^2} \right], \quad \hat{\boldsymbol{e}}_2 = (0,1,0)^t,$$

$$D_3 = \Gamma_w \left[K_1 - \left(\frac{1}{\tau} + \frac{1}{3} \right) K_2 - \tau^2 \frac{K_3^2}{\mu} \right], \quad \hat{\boldsymbol{e}}_3 = (1,0,0)^t, \quad (4.1)$$

with $D_i < D_i^0$. There are accidental degeneracies for $K_3 = 0.38$ GPa and $K_3 = 0.74$ GPa, see Fig. 3.

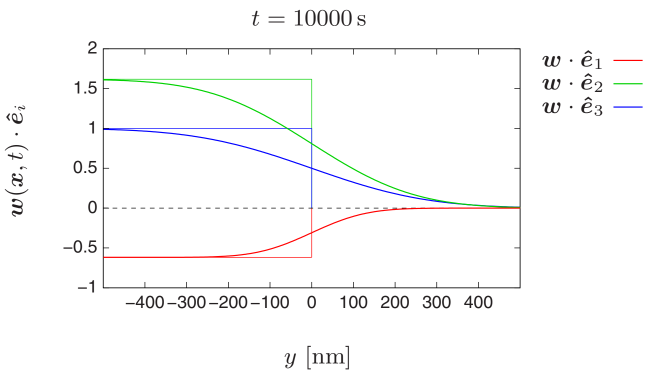

FIG. 4. (Color online) Broadening of the phason walls.

B. Phason wall diffusion

Diffusion constants can be measured by phason wall diffusion experiments.[18] Equation

$$[-i\omega\underline{\mathbf{1}}+\Gamma_w\underline{\tilde{\mathbf{M}}}(\boldsymbol{q})]\boldsymbol{w}(\boldsymbol{q},\omega)=0, \tag{4.2}$$

already specified in Sec. III B describes an anisotropic diffusion equation. In direct space it is for $K_3=0$,

$$\frac{\partial w_\alpha}{\partial t}(\boldsymbol{x},t)=\sum_{\beta=4}^{6}\sum_{i,j=1}^{3}C_{\alpha i\beta j}\frac{\partial}{\partial x_i}\frac{\partial}{\partial x_j}w_\beta(\boldsymbol{x},t),\quad \alpha=4,5,6, \tag{4.3}$$

with the stiffness tensor $C_{\alpha i\beta j}$ and $C_{\alpha i\beta j}q_iq_j := M_{\alpha\beta}(\boldsymbol{q})$. For the special initial condition of a phason wall

$$\boldsymbol{w}(\boldsymbol{x},t=0)=\mathbf{b}^{\perp}\{1-\Theta[\hat{\boldsymbol{n}}^t\cdot(\boldsymbol{x}-\boldsymbol{x}_0)]\}, \tag{4.4}$$

where $\mathbf{b}^{\perp}$ denotes the Burgers vector, $\hat{\boldsymbol{n}}$ the plane normal of the wall, and Θ the usual Heaviside step function, we obtain with Eq. (3.13),

$$\boldsymbol{w}(\boldsymbol{x},t)=\frac{1}{2}\sum_{i=1}^{3}[\hat{\boldsymbol{e}}_i^t(\varphi,\vartheta)\cdot\mathbf{b}^{\perp}]\times\left(1-\mathrm{erf}\left\{\frac{\hat{\boldsymbol{n}}^t\cdot(\boldsymbol{x}-\boldsymbol{x}_0)}{2\sqrt{D_i(\varphi,\vartheta)t}}\right\}\right)\hat{\boldsymbol{e}}_i(\varphi,\vartheta)|_{\varphi,\vartheta},\quad t>0, \tag{4.5}$$

where erf is the error function. If the normal vector $\hat{\boldsymbol{n}}$ points along the twofold axis A2: $\hat{\boldsymbol{n}}=\hat{\boldsymbol{e}}_y$ and the Burgers vector is taken as measured by Feuerbacher *et al.*: $\mathbf{b}^{\perp}\propto(1,\tau,1-\tau)^t$, then the wall splits into three propagating walls with different diffusion coefficients. For the plane in the experiment of Ref. 18, $\hat{\boldsymbol{n}}\propto(\tau,-1,\tau+1)^t$, $\hat{\boldsymbol{e}}_1$ and $\hat{\boldsymbol{e}}_2$ are orthogonal to $\mathbf{b}^{\perp}$, hence only one wall was visible (see Fig. 4).

For this direction the effect of K_3 on the diffusion coefficients has been discussed in the previous section. For $K_3=0$ the D_i in Eq. (4.1) are linearly dependent on K_1 and K_2. The maximal difference between them occurs for the maximum value of $|K_2/K_1|$. This is given by the stability criteria:[30,31] $|K_2/K_1|_{\max}=3/4$.

V. CONCLUSION

In conclusion we have derived the hydrodynamic structure factor from the time-dependent mass density of a distorted quasicrystal. The main quantity in the expression of $S(\boldsymbol{k},\omega)$ is the imaginary part of the dynamic susceptibility. For icosahedral quasicrystals we have obtained this dynamic response function, which relates linearly external volume forces to displacements, from the corresponding hydrodynamic equations.

We have investigated the diffuse part of the hydrodynamic structure factor for a single reciprocal-lattice vector and without Debye-Waller factor for phonons and phasons. The essential result for the phononic diffuse part are two maxima along a dispersion relation modified by damping parameters. The phonon-phason coupling constant K_3 is one of these parameters in addition to the kinetic viscosities and kinetic coefficients. Whether the two maxima can be separately observed primarily depends on Γ_u. The displayed phononic isointensity contours, which were concentric circles around the reciprocal-lattice vector in physical space, are out of the measurable frequency window for neutron scattering. Hence we considered the time-dependent phononic diffuse part, which takes the form of a damped oscillation. The diffusive character of the phasons can be detected in their diffuse part of the dynamic structure factor. Frequency ω and kinetic coefficient Γ_w cause an inverse zooming effect of the phasonic isointensity contours. For small frequency values they are anisotropic, becoming almost circular for higher ones. Since the phasonic diffuse part is a Lorentzian in frequency, we obtained an exponential decay in time for the inverse Fourier transformation. We used this result to establish a connection to the intensity-intensity correlation function $I(\boldsymbol{q},t)$, measured by coherent x-ray spectroscopy. $I(\boldsymbol{q},t)$ depends on the function $g(\boldsymbol{q},t)$, which for phason fluctuations is proportional to the squared ratio of dynamic and static phasonic diffuse part.

Analyzing the hydrodynamic mode structure of the linearized equations of motion, we have listed all solutions for the propagating and the diffusive modes. The propagating solutions are damped oscillations depending on the corresponding sound velocity. Coupling to the phasons causes an additional damping term. On the other hand the phasonic diffusion constants take reduced values for $K_3\neq 0$. The particular solutions can be expressed with the corresponding parts of the dynamic response function.

We separately have studied the directionality of the phasonic diffusion coefficients in the q_xq_y-twofold plane. There are systematic degeneracies along the threefold and fivefold axes and accidental ones along the twofold axis for special values of the coupling constant. The intersection point along the fivefold axis is independent of K_3. Solving the phasonic diffusion equation for the special initial condition of a phason wall in general results in a superposition of three error functions, for each diffusion constant one. In the experiment of Ref. 18 only one wall is visible. The selection rule is the component of the Burgers vector along the eigenvectors of the phasonic matrix.

APPENDIX A: THE ELASTIC MATRICES

The definition of the elastic constants in this paper is the same as of Widom *et al.*,[30] which is also used in several experimental papers.[20,26,27] For the phononic matrices $\underline{\mathbf{N}}(\boldsymbol{q})$ and $\underline{\tilde{\mathbf{N}}}(\boldsymbol{q})$ follows

$$\underline{\mathbf{N}}(\boldsymbol{q}) = (\Lambda\underline{\mathbf{Q}} + \mu\underline{\mathbf{P}})q^2, \quad \underline{\tilde{\mathbf{N}}}(\boldsymbol{q}) = [(\Lambda - B)\underline{\mathbf{Q}} + \mu\underline{\mathbf{P}}]q^2, \tag{A1}$$

with the longitudinal and transverse projection operator

$$\underline{\mathbf{Q}} := \hat{\boldsymbol{q}} \otimes \hat{\boldsymbol{q}} \quad \text{and} \quad \underline{\mathbf{P}} = \underline{\mathbf{1}} - \hat{\boldsymbol{q}} \otimes \hat{\boldsymbol{q}}, \tag{A2}$$

given already in Sec. II C. λ and μ are the usual Lamé constants and Λ defined as $\Lambda := 2\mu + \lambda$. The tensor of viscosity $\underline{\boldsymbol{\eta}}(\boldsymbol{q})$ splits also in a transverse and longitudinal part

$$\underline{\boldsymbol{\eta}}(\boldsymbol{q}) = (\eta_L\underline{\mathbf{Q}} + \eta_T\underline{\mathbf{P}})q^2, \tag{A3}$$

where η_T and η_L are the correspond kinetic viscosities. Finally, phasonic matrix $\underline{\mathbf{M}}(\boldsymbol{q})$ and coupling matrix $\underline{\mathbf{C}}(\boldsymbol{q})$ take the form

$$\underline{\mathbf{M}}(\boldsymbol{q}) = K_1 q^2 \underline{\mathbf{1}} + K_2 \begin{pmatrix} -\frac{1}{3}q^2 - \frac{1}{\tau}q_y^2 + \tau q_z^2 & 2q_xq_y & 2q_xq_z \\ 2q_xq_y & -\frac{1}{3}q^2 - \frac{1}{\tau}q_z^2 + \tau q_x^2 & 2q_yq_z \\ 2q_xq_z & 2q_yq_z & -\frac{1}{3}q^2 - \frac{1}{\tau}q_x^2 + \tau q_y^2 \end{pmatrix}, \tag{A4}$$

$$\underline{\mathbf{C}}(\boldsymbol{q}) = K_3 \begin{pmatrix} q_x^2 - \tau q_y^2 + \frac{1}{\tau}q_z^2 & \frac{2}{\tau}q_xq_y & -2\tau q_xq_z \\ -2\tau q_xq_y & q_y^2 - \tau q_z^2 + \frac{1}{\tau}q_x^2 & \frac{2}{\tau}q_yq_z \\ \frac{2}{\tau}q_xq_z & -2\tau q_yq_z & q_z^2 - \tau q_x^2 + \frac{1}{\tau}q_y^2 \end{pmatrix}, \quad q^2 := |\boldsymbol{q}|^2 = q_x^2 + q_y^2 + q_z^2. \tag{A5}$$

Note, that these are the same matrices as by Jarić and Nelson[13] with the replacements for the elastic constants there,

$$m_1 = 2\sqrt{6}\left(\mu + \frac{\lambda}{4}\right), \quad m_2 = \frac{\sqrt{30}}{2}\lambda, \quad m_3 = 3K_1,$$

$$m_4 = 2\sqrt{5}K_2, \quad m_5 = \sqrt{60}K_3. \tag{A6}$$

APPENDIX B: THE DYNAMIC RESPONSE FUNCTION

Consider now the dynamic response function

$$\underline{\boldsymbol{\chi}}(\boldsymbol{q},\omega) = [i\omega^3\underline{\mathbf{A}} - \omega^2\underline{\mathbf{B}}(\boldsymbol{q}) - i\omega\underline{\boldsymbol{\Gamma}}(\boldsymbol{q}) - \underline{\boldsymbol{\Delta}}(\boldsymbol{q})]^{-1} \times [-\omega^2\Gamma_u\underline{\mathbf{A}} - i\omega\underline{\mathbf{E}}(\boldsymbol{q}) - \underline{\mathbf{Z}}(\boldsymbol{q})], \tag{B1}$$

the six matrices $\underline{\mathbf{A}}$, $\underline{\mathbf{B}}(\boldsymbol{q})$, $\underline{\boldsymbol{\Gamma}}(\boldsymbol{q})$, $\underline{\boldsymbol{\Delta}}(\boldsymbol{q})$, $\underline{\mathbf{E}}(\boldsymbol{q})$, and $\underline{\mathbf{Z}}(\boldsymbol{q})$ can be identified as

$$\underline{\mathbf{A}} = \begin{pmatrix} \underline{\mathbf{1}} & \underline{\mathbf{0}} \\ \underline{\mathbf{0}} & \underline{\mathbf{0}} \end{pmatrix}, \quad \underline{\mathbf{B}}(\boldsymbol{q}) = \begin{pmatrix} \underline{\boldsymbol{\eta}} + \Gamma_u\underline{\mathbf{N}} & \Gamma_u\underline{\mathbf{C}} \\ \underline{\mathbf{0}} & \underline{\mathbf{0}} \end{pmatrix},$$

$$\underline{\boldsymbol{\Gamma}}(\boldsymbol{q}) = \begin{pmatrix} \frac{\underline{\tilde{\mathbf{N}}}}{\rho_0} + \Gamma_u\underline{\boldsymbol{\eta}}\underline{\mathbf{N}} + \frac{A - B}{\rho_0}q^2\underline{\mathbf{Q}} & \left[\frac{1}{\rho_0} + \Gamma_u\underline{\boldsymbol{\eta}}\right]\underline{\mathbf{C}} \\ \underline{\mathbf{0}} & \underline{\mathbf{1}} \end{pmatrix},$$

$$\underline{\boldsymbol{\Delta}}(\boldsymbol{q}) = \begin{pmatrix} -\Gamma_u[\Lambda A - B^2]\frac{q^4}{\rho_0}\underline{\mathbf{Q}} & -\Gamma_u A\frac{q^2}{\rho_0}\underline{\mathbf{Q}}\underline{\mathbf{C}} \\ -\Gamma_w\underline{\mathbf{C}}^t & -\Gamma_w\underline{\mathbf{M}} \end{pmatrix},$$

$$\underline{\mathbf{E}}(\boldsymbol{q}) = \begin{pmatrix} \frac{1}{\rho_0}\underline{\mathbf{1}} + \Gamma_u\underline{\boldsymbol{\eta}} & \underline{\mathbf{0}} \\ \underline{\mathbf{0}} & \underline{\mathbf{0}} \end{pmatrix} \quad \text{and}$$

$$\underline{\mathbf{Z}}(\boldsymbol{q}) = \begin{pmatrix} -\Gamma_u A\frac{q^2}{\rho_0}\underline{\mathbf{Q}} & \underline{\mathbf{0}} \\ \underline{\mathbf{0}} & -\Gamma_w\underline{\mathbf{1}} \end{pmatrix}. \tag{B2}$$

All of them are 6×6 matrices consisting of 3×3 blocks. We define

$$\underline{\mathbf{U}}(\boldsymbol{q},\omega) := [i\omega^3\underline{\mathbf{A}} - \omega^2\underline{\mathbf{B}}(\boldsymbol{q}) - i\omega\underline{\boldsymbol{\Gamma}}(\boldsymbol{q}) - \underline{\boldsymbol{\Delta}}(\boldsymbol{q})] \quad \text{and}$$

$$\underline{\mathbf{V}}(\boldsymbol{q},\omega) := [-\omega^2\Gamma_u\underline{\mathbf{A}} - i\omega\underline{\mathbf{E}}(\boldsymbol{q}) - \underline{\mathbf{Z}}(\boldsymbol{q})], \tag{B3}$$

so that $\underline{\boldsymbol{\chi}}(\boldsymbol{q},\omega)$ becomes

$$\underline{\boldsymbol{\chi}}(\boldsymbol{q},\omega) = \underline{\mathbf{U}}(\boldsymbol{q},\omega)^{-1}\underline{\mathbf{V}}(\boldsymbol{q},\omega), \tag{B4}$$

and for the 3×3 blocks, respectively,

$$\begin{pmatrix} \underline{\boldsymbol{\chi}}_{uu} & \underline{\boldsymbol{\chi}}_{uw} \\ \underline{\boldsymbol{\chi}}_{wu} & \underline{\boldsymbol{\chi}}_{ww} \end{pmatrix} = \begin{pmatrix} \underline{\mathbf{U}}_{uu} & \underline{\mathbf{U}}_{uw} \\ \underline{\mathbf{U}}_{wu} & \underline{\mathbf{U}}_{ww} \end{pmatrix}^{-1} \begin{pmatrix} \underline{\mathbf{V}}_{uu} & \underline{\mathbf{0}} \\ \underline{\mathbf{0}} & \underline{\mathbf{V}}_{ww} \end{pmatrix}. \tag{B5}$$

Carrying the inversion of $\underline{\mathbf{U}}(\boldsymbol{q},\omega)$ out,

$$\underline{\mathbf{U}}^{-1} = \begin{pmatrix} \underline{\mathbf{U}}_{uu}^{-1} + \underline{\mathbf{U}}_{uu}^{-1}\underline{\mathbf{U}}_{uw}\underline{\mathbf{S}}^{-1}\underline{\mathbf{U}}_{wu}\underline{\mathbf{U}}_{uu}^{-1} & -\underline{\mathbf{U}}_{uu}^{-1}\underline{\mathbf{U}}_{uw}\underline{\mathbf{S}}^{-1} \\ -\underline{\mathbf{S}}^{-1}\underline{\mathbf{U}}_{wu}\underline{\mathbf{U}}_{uu}^{-1} & \underline{\mathbf{S}}^{-1} \end{pmatrix}, \tag{B6}$$

the phononic $\underline{\boldsymbol{\chi}}_{uu}(\boldsymbol{q},\omega)$, phasonic $\underline{\boldsymbol{\chi}}_{ww}(\boldsymbol{q},\omega)$, and coupling part $\underline{\boldsymbol{\chi}}_{uw}(\boldsymbol{q},\omega)=\underline{\boldsymbol{\chi}}_{wu}^t(\boldsymbol{q},\omega)$ of the dynamic response function results, with the stiffness tensor $\underline{\mathbf{S}} := \underline{\mathbf{U}}_{ww} - \underline{\mathbf{U}}_{wu}\underline{\mathbf{U}}_{uu}^{-1}\underline{\mathbf{U}}_{uw}$, as

$$\underline{\boldsymbol{\chi}}_{uu}(\boldsymbol{q},\omega) = \underline{\mathbf{U}}_{uu}^{-1}\underline{\mathbf{V}}_{uu} + \underline{\mathbf{U}}_{uu}^{-1}\underline{\mathbf{U}}_{uw}\underline{\mathbf{S}}^{-1}\underline{\mathbf{U}}_{wu}\underline{\mathbf{U}}_{uu}^{-1}\underline{\mathbf{V}}_{uu} = \underline{\boldsymbol{\chi}}_{uu}^0 - \underline{\boldsymbol{\chi}}_{uw}\underline{\mathbf{C}}^t\underline{\boldsymbol{\chi}}_{uu}^0, \tag{B7a}$$

$$\underline{\boldsymbol{\chi}}_{uw}(\boldsymbol{q},\omega) = -\underline{\mathbf{U}}_{uu}^{-1}\underline{\mathbf{U}}_{uw}\underline{\mathbf{S}}^{-1}\underline{\mathbf{V}}_{ww} = -\underline{\boldsymbol{\chi}}_{uu}^0\underline{\mathbf{C}}\underline{\mathbf{S}}^{-1}\Gamma_w, \tag{B7b}$$

$$\underline{\boldsymbol{\chi}}_{ww}(\boldsymbol{q},\omega) = \underline{\mathbf{S}}^{-1}\underline{\mathbf{V}}_{ww} = \Gamma_w\underline{\mathbf{S}}^{-1}, \tag{B7c}$$

where $\underline{\mathbf{S}}=[-i\omega\underline{\mathbf{1}}+\Gamma_w(\underline{\mathbf{M}}-\chi_L^0\underline{\mathbf{C}}^t\underline{\mathbf{Q}}\underline{\mathbf{C}}-\chi_T^0\underline{\mathbf{C}}^t\underline{\mathbf{P}}\underline{\mathbf{C}})]$. $\underline{\boldsymbol{\chi}}_{uu}^0$ denotes the phononic response function in case of no coupling ($K_3=0$),

$$\underline{\boldsymbol{\chi}}_{uu}^0(\boldsymbol{q},\omega) = \chi_L^0(q,\omega)\underline{\mathbf{Q}} + \chi_T^0(q,\omega)\underline{\mathbf{P}}. \tag{B8}$$

The longitudinal and transverse part take the form

$$\chi_L^0(q,\omega) = \frac{-\omega^2\Gamma_u - i\omega\left(\frac{1}{\rho_0} + \eta_L\Gamma_u q^2\right) + \Gamma_u\frac{A}{\rho_0}q^2}{i\omega^3 - \omega^2(\eta_L + \Gamma_u\Lambda)q^2 - i\omega(c_L^2q^2 + \eta_L\Gamma_u\Lambda q^4) + \Gamma_u\frac{\Lambda A - B^2}{\rho_0}q^4}, \tag{B9a}$$

$$\chi_T^0(q,\omega) = \frac{-i\omega\Gamma_u + \frac{1}{\rho_0} + \eta_T\Gamma_u q^2}{-\omega^2 - i\omega(\eta_T + \Gamma_u\mu)q^2 + c_T^2q^2 + \eta_T\Gamma_u\mu q^4}. \tag{B9b}$$

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