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Spontaneous decay of long-wavelength surface acoustic phonons

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The damping constant of long-wavelength acoustic surface phonons due to their spontaneous decay via cubic anharmonicity is calculated. Since the Rayleigh branch is the phonon branch of lowest frequency in the crystal, collinear- or quasicollinear-decay processes play an important role. Power laws for the dependence of the damping constant on the frequency are derived for the dispersionless and dispersive cases which, in contrast to the high-temperature regime, differ from the Herring scaling. Numerical estimates for the damping constants are also given.

I. INTRODUCTION

The anharmonicity of the lattice potential provides an intrinsic attenuation mechanism for the propagation of surface phonons on ideal crystal surfaces. For long-wavelength acoustic surface phonons, several theoretical[1–4] and experimental[5–9] studies have been performed on the temperature and frequency dependence of their damping constants resulting from cubic anharmonicity. In the collisionless regime, where the frequency ω of the surface acoustic phonon under consideration is larger than the inverse lifetimes of the thermal phonons it interacts with via the cubic-anharmonic coupling constants, its damping constant may be calculated in perturbation theory from the so-called bubble-diagram approximation[10] [Fig. 1(a)] for the surface-phonon self-energy. Within this approximation, Maradudin and Mills[1] found, in a calculation based on a lattice-dynamical model, that for $\hbar\omega \ll k_B T$ the damping constant of a Rayleigh mode is governed by the interaction with thermal phonons of the lowest bulk branch and is proportional to ωT^4. This frequency and temperature dependence of the damping constant of a Rayleigh wave is identical to that of the damping constant of long-wavelength transverse-acoustic (TA) bulk phonons from the Landau-Rumer process.[11] It has been confirmed experimentally by Salzmann *et al.*[5] and Budreau and Carr[6] for quartz, while Daniel and de Klerk[7] have found the T^4 dependence in a certain temperature interval, but they do not confirm the linear frequency dependence. Measurements of the attenuation of surface acoustic waves have also been performed by Slobodnik *et al.*[8] in $LiNbO_3$, and Slobodnik and Budreau[9] in

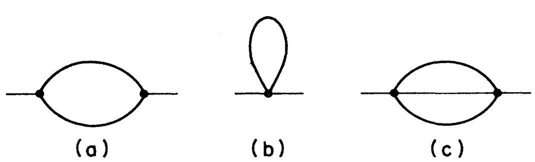

FIG. 1. Self-energy diagrams considered for the damping constant and/or frequency dispersion of Rayleigh modes.

$Bi_{12}GeO_{20}$; however, largely in regimes where the scattering processes considered in Ref. 1 do not seem to represent the dominant attenuation mechanism. Therefore, they cannot be expected to show the ωT^4 dependence of the damping constant. The results of Maradudin and Mills have been rederived and extended in the framework of nonlinear, elastic, continuum theory by King and Sheard,[2] and also by Sakuma and Nakayama.[4] While the earlier calculations did not account for the presence of the surface in the displacements associated with the thermal bulk phonons, the latter authors employ the normal modes of a semi-infinite, isotropic, elastic medium. Taking only the total reflection[12] or mixed[13] modes into account for the thermal phonons, they find, with decreasing temperature, strong deviations[3] from the ωT^4 law, which, however, still follow the Herring scaling[14]

$$\Gamma_{\lambda \mathbf{q}}(\lambda T)=\lambda^5 \Gamma_{\mathbf{q}}(T) , \tag{1.1}$$

where $\Gamma_{\mathbf{q}}(T)$ is the damping constant of the Rayleigh mode with wave vector $\mathbf{q}$ at temperature T. It is, in fact, easily seen from the general expression for the damping constant of a Rayleigh mode obtained from the bubble diagram with bare propagators in the dispersionless approximation that the Herring scaling applies to it even for anisotropic crystals. In certain cases, in particular for collinear processes, this approximation may, however, break down, as will be shown below. More recently, Tamura[15] has performed a calculation of the intrinsic damping constant of Rayleigh waves for an isotropic, semi-infinite, elastic continuum in the regime $\hbar\omega > k_B T$, where he considers the scattering of transverse bulk phonons of shear horizontal polarization as the dominant damping mechanism. From these processes, he obtains a lifetime of the order of magnitude of seconds for Rayleigh-wave frequencies of 100 GHz at 0.4 K. Since his result is based on the bubble diagram with bare propagators in the dispersionless approximation, it obeys the Herring scaling.

With the present investigation, we attempt to close a gap left open in the series of studies hitherto carried out on the attenuation of surface acoustic phonons, namely the regime of zero temperature or temperatures so small compared to $\hbar\omega/k_B$ that all thermal processes are negligible. Here, it is only the spontaneous decay that leads to a damping of phonons in an ideal crystal. To our knowledge, even the order of magnitude of the intrinsic lifetime of Rayleigh modes for a given frequency is unknown in this regime up to now. It is also not known at which temperature thermal processes become unimportant, as the temperature is decreased. In the following, it will be shown that for strongly anisotropic crystals under the experimental conditions considered in the calculation of Ref. 15, namely Rayleigh waves of 100 GHz at 0.4 K, the spontaneous decay can outweigh the thermal processes in certain directions of propagation.

The lifetime of longitudinal-acoustic (LA) bulk phonons at zero temperature in a frequency regime where the dispersion can be largely neglected follows the Herring scaling, i.e., $\Gamma_{\mathbf{q}} \sim q^5$ (Refs. 16 and 17). This also applies to the transverse-acoustic phonons,[18,19] except in certain cases for the collinear decay. The collinear processes form an inherent problem of the dispersionless theory[20] in that they lead to divergences in lowest-order perturbation theory. Recently, it has been shown[19,21] that for propagation directions for which the principal curvatures of the corresponding sheet of the slowness surface have different signs, the bubble diagram does not yield a finite result in the limits of vanishing dispersion and infinite lifetime of the decay products. Consequently, the Herring scaling is expected to be no longer valid, if the noncollinear decay is forbidden by energy and momentum conservation.

The anomalous behavior found in the case of transverse bulk phonons is expected to be even more pronounced in the case of surface phonons for the following reason. Since the Rayleigh branch is the lowest branch in the phonon spectrum, the phase space for the decay products of a Rayleigh mode is largely restricted to propagation directions close to that of the decaying mode in nearly isotropic crystals. Furthermore, the two-dimensional character of the surface phonons as decay products should be reflected in a frequency dependence of the damping constant significantly different from that of bulk phonons and in a strong dependence of the damping constant on the propagation direction. We will find in the present work that this is indeed the case.

The present paper is organized in the following way. In Sec. II, we present the basic expressions needed for the calculation of the damping constant of long-wavelength acoustic surface phonons from the bubble diagram within continuum elasticity theory. The continuum approach has, apart from its easier tractability if compared to a lattice-dynamical calculation, the advantage that the knowledge of only a small set of parameters is required, namely the mass density and the second- and third-order elastic moduli, which are directly accessible experimentally. In Sec. III, we then derive an expression for the damping constant of Rayleigh modes from their collinear decay in the dispersionless approximation. A power law for its frequency dependence is derived which does not follow the Herring scaling, and numerical estimates of its absolute value are given for several substances. After a general discussion on the dispersion of Rayleigh waves, its significant influence on the collinear and quasi-collinear decay is considered in Sec. IV. In Sec. V, we address the influence of higher-order phonon processes and in particular discuss the role of the collinear decay in the four-phonon processes symbolized by the diagram Fig. 1(c).

II. GENERAL THEORY

The damping constant $\Gamma_{\mathbf{q}S}$ for the amplitude of a surface phonon due to the spontaneous decay via cubic anharmonicity may be calculated on the basis of the following formula:

$$\Gamma_{\mathbf{q}S}=\frac{L^2}{8\pi^2}\sum_{J,J'}\int d^2q'|V_3(-\mathbf{q}S,\mathbf{q}'J,\mathbf{q}-\mathbf{q}'J')|^2\frac{\Gamma_{\mathbf{q}'J}+\Gamma_{\mathbf{q}-\mathbf{q}'J'}}{(\omega_{\mathbf{q}S}-\omega_{\mathbf{q}'J}-\omega_{\mathbf{q}-\mathbf{q}'J'})^2+(\Gamma_{\mathbf{q}'J}+\Gamma_{\mathbf{q}-\mathbf{q}'J'})^2}\ , \tag{2.1}$$

where $\mathbf{q}$ and $\mathbf{q}'$ are two-dimensional wave vectors parallel to the surface. The indices J and J' denote the phonon branches of the decay products and also, in the case of bulk phonons, the vertical components of their wave vectors. The surface phonons, the damping of which we are studying at this stage, are the Rayleigh modes. Since the Rayleigh modes are the phonons of lowest frequency for a given $\mathbf{q}$, significant contributions to the integral in (2.1) result primarily from other Rayleigh modes. We may therefore drop the indices S, J, and J'.

The derivation of (2.1), which corresponds to the bubble diagram with "solid lines" in interacting-phonon theory, is entirely analogous to the corresponding formula for bulk phonons given in Ref. 19. To evaluate the quantities in (2.1) for a semi-infinite crystal, we use continuum elasticity theory and introduce the displacement field $\mathbf{u}(\mathbf{x})$. On expanding $\mathbf{u}(\mathbf{x})$ into normal modes, we decompose

$$\mathbf{u}(\mathbf{x})=\mathbf{u}^{(B)}(\mathbf{x})+\mathbf{u}^{(R)}(\mathbf{x})\ , \tag{2.2}$$

where

$$u_\alpha^{(R)}(\mathbf{x})=\left[\frac{\hbar}{2\rho L^2}\right]^{1/2}\sum_{\mathbf{q}}e^{i\mathbf{q}\cdot\mathbf{x}_\parallel}\left[\frac{q}{\omega_{\mathbf{q}}}\right]^{1/2}\times\sum_r[b_\alpha(\hat{\mathbf{q}}r)e^{qa(\hat{\mathbf{q}}r)z}]A(\mathbf{q}) \tag{2.3}$$

and $\mathbf{u}^{(B)}(\mathbf{x})$ contains the pure bulk and mixed modes and other possibly existing surface acoustic modes of higher frequency, e.g., the shear horizontal mode discussed in

Refs. 22 and 23. $\hat{\mathbf{q}}=\mathbf{q}/q$ denotes the propagation direction in the surface which is identified with the x-y plane, and

$$A(\mathbf{q})=a_{\mathbf{q}}+a^{\dagger}_{-\mathbf{q}} , \tag{2.4}$$

where $a^{\dagger}_{\mathbf{q}}$ and $a_{\mathbf{q}}$ are the creation and annihilation operators. Within the x-y plane, we impose periodic boundary conditions with length of periodicity L. The normalization condition requires

$$\int_{-\infty}^{0} dz \sum_{\alpha} \left| \sum_{r} b_{\alpha}(\hat{\mathbf{q}}r)e^{qa(\hat{\mathbf{q}}r)z} \right|^2 = 1/q . \tag{2.5}$$

On inserting the expansion (2.3) for $\mathbf{u}^{(R)}(\mathbf{x})$ into the nonlinear part of the potential energy of a semi-infinite, elastic medium

$$\begin{aligned}\delta\Phi^{(s)}&=\tfrac{1}{6}\sum_{\alpha,\beta,\mu,\nu,\zeta,\xi} S_{\alpha\beta\mu\nu\zeta\xi}\int d^3x \left[\frac{\partial}{\partial x_\beta}u_\alpha^{(R)}(\mathbf{x})\right]\left[\frac{\partial}{\partial x_\nu}u_\mu^{(R)}(\mathbf{x})\right]\left[\frac{\partial}{\partial x_\xi}u_\zeta^{(R)}(\mathbf{x})\right]\\ &=\frac{\hbar}{6}\sum_{\mathbf{q},\mathbf{q}'} V_3(-\mathbf{q},\mathbf{q}',\mathbf{q}-\mathbf{q}')A(-\mathbf{q})A(\mathbf{q}')A(\mathbf{q}-\mathbf{q}') ,\end{aligned} \tag{2.6}$$

the following general expression for the nonlinear coupling constants in (2.1) is obtained:

$$\begin{aligned}V_3(-\mathbf{q},\mathbf{q}',\mathbf{q}-\mathbf{q}')=&\left[\frac{\hbar}{8\rho^3L^2}\frac{qq'|\mathbf{q}-\mathbf{q}'|}{\omega_{\mathbf{q}}\omega_{\mathbf{q}'}\omega_{\mathbf{q}-\mathbf{q}'}}\right]^{1/2}\\ &\times\sum_{\alpha,\beta,\mu,\nu,\zeta,\xi} S_{\alpha\beta\mu\nu\zeta\xi}\sum_{r,r',r''} b_\alpha(-\hat{\mathbf{q}}r)b_\mu(\hat{\mathbf{q}}'r')b_\zeta(\mathbf{q}-\mathbf{q}'r'')C_\beta(\mathbf{q}'r)C_\nu(\mathbf{q}'r')C_\xi(\mathbf{q}-\mathbf{q}'r'')\\ &\times[qa(\hat{\mathbf{q}}r)+q'a(\hat{\mathbf{q}}'r')+|\mathbf{q}-\mathbf{q}'|a(\mathbf{q}-\mathbf{q}'r'')]^{-1} ,\end{aligned} \tag{2.7}$$

where

$$C_\alpha(\mathbf{q}r)=\begin{cases} iq_\alpha & \text{if } \alpha\neq z\\ qa(\hat{\mathbf{q}}r) & \text{if } \alpha=z .\end{cases} \tag{2.8}$$

The connection between the expansion coefficients $S_{\alpha\beta\mu\nu\zeta\xi}$ of the potential-energy density with respect to the infinitesimal strain parameters and the second- and third-order elastic constants can be found in Ref. 24.

III. THE COLLINEAR DECAY

Within elasticity theory, Rayleigh waves are nondispersive, i.e.,

$$\omega_{\mathbf{q}}=v_{\hat{\mathbf{q}}}q . \tag{3.1}$$

If (3.1) is used in (2.1), the approximation (3.2),

$$\frac{\Gamma_{\mathbf{q}'}+\Gamma_{\mathbf{q}-\mathbf{q}'}}{(\omega_{\mathbf{q}}-\omega_{\mathbf{q}'}-\omega_{\mathbf{q}-\mathbf{q}'})^2+(\Gamma_{\mathbf{q}'}+\Gamma_{\mathbf{q}-\mathbf{q}'})^2}\approx\pi\delta(\omega_{\mathbf{q}}-\omega_{\mathbf{q}'}-\omega_{\mathbf{q}-\mathbf{q}'}) , \tag{3.2}$$

which would lead to Fermi's golden rule expression and correspond to the bubble diagram with bare propagators, is not applicable, independent of how small the damping constants of the decay products are. As pointed out in Refs. 19 and 21 for the case of bulk phonons, a necessary condition for the validity of this approximation is that after transforming to a set of integration variables which contains the variable,

$$x=\omega_{\mathbf{q}}-\omega_{\mathbf{q}'}-\omega_{\mathbf{q}-\mathbf{q}'} , \tag{3.3}$$

the integrand has to vary slowly within the width $\Gamma_{\mathbf{q}'}+\Gamma_{\mathbf{q}-\mathbf{q}'}$. This condition cannot be fulfilled in our case, since the Jacobian of such a transformation diverges when $\hat{\mathbf{q}}'$ approaches $\hat{\mathbf{q}}$. Explicitly,

$$d^2q'=J\,dx\,d\phi , \tag{3.4}$$

where

$$J^{-1}=q'^{-1}\left|\frac{\partial}{\partial q'}(\omega_{\mathbf{q}}-\omega_{\mathbf{q}'}-\omega_{\mathbf{q}-\mathbf{q}'})\right| , \tag{3.5}$$

$$\omega_{\mathbf{q}}-\omega_{\mathbf{q}'}-\omega_{\mathbf{q}-\mathbf{q}'}=\frac{1}{2}\frac{qq'}{q'-q}\Omega(\hat{\mathbf{q}})\phi^2+O(\phi^3) \tag{3.6}$$

for $q'<q$. Here, ϕ is the angle between $\hat{\mathbf{q}}$ and $\hat{\mathbf{q}}'$. We therefore have to deal with the full expression on the right-hand side of (2.1). In order to evaluate this expression, we first expand (3.6). The coefficient $\Omega(\hat{\mathbf{q}})$ is given by

$$\Omega(\hat{\mathbf{q}})=\left[1+\frac{\partial^2}{\partial\phi^2}\right]v_{\hat{\mathbf{q}}} \tag{3.7}$$

and is connected with the curvature $\kappa(\hat{\mathbf{q}})$ of the slowness curve of the Rayleigh waves via

$$\kappa(\hat{\mathbf{q}})=-\Omega(\hat{\mathbf{q}})\left[1+\left[v_{\hat{\mathbf{q}}}^{-1}\frac{\partial}{\partial\phi}v_{\hat{\mathbf{q}}}\right]^2\right]^{-3/2} . \tag{3.8}$$

Inserting (3.6) into (2.1) and confining the integration over the angle ϕ to a small interval, which yields the dominant contributions, leads to the approximate equation

$$\Gamma_{\mathbf{q}}\approx\frac{L^2}{8\pi^2}\int_0^q dq'q'|V_3(-\mathbf{q},q'\hat{\mathbf{q}},(q-q')\hat{\mathbf{q}})|^2\times\int_{-\epsilon}^{\epsilon}d\phi\frac{\gamma_{\hat{\mathbf{q}}}(q,q')}{f_{\hat{\mathbf{q}}}^2(q,q')\phi^4+\gamma_{\hat{\mathbf{q}}}^2(q,q')} , \tag{3.9}$$

where

$$\gamma_{\hat{\mathbf{q}}}(q,q')=\Gamma_{q'\hat{\mathbf{q}}}+\Gamma_{(q-q')\hat{\mathbf{q}}} , \tag{3.10}$$

$$f_{\hat{\mathbf{q}}}(q,q')=\frac{1}{2}\frac{qq'}{q-q'}\Omega(\hat{\mathbf{q}}) . \tag{3.11}$$

To estimate (3.9), we note that the main contributions to the double integral do not result from the small-q' region but rather from $q'\approx\frac{1}{2}q$, because V_3 varies as q' and so the numerator is proportional to q'^3. We will now assume that γ/f is so small that we may replace the boundaries of the ϕ integration by $\pm\infty$. Later on it will become clear that this approximation is, in fact, valid apart from special directions, where the curvature of the slowness curve is very small. We may then write

$$\int_{-\epsilon}^{\epsilon} d\phi \frac{\gamma}{f^2\phi^4+\gamma^2}\approx\frac{\pi}{\sqrt{2}}(f\gamma)^{1/2} . \tag{3.12}$$

Assuming, furthermore, that the collinear decay is the dominant damping process, which is always the case for a nearly isotropic elastic medium, we obtain the following self-consistency equation for $\Gamma_{\hat{\mathbf{q}}}(q):=\Gamma_{\mathbf{q}}$:

$$\Gamma_{\hat{\mathbf{q}}}(q)=\frac{L^2q^{11/2}}{8\pi|\Omega(\hat{\mathbf{q}})|^{1/2}}\int_0^1 dk[\Gamma_{\hat{\mathbf{q}}}(kq)+\Gamma_{\hat{\mathbf{q}}}((1-k)q)]^{-1/2}k^{1/2}(1-k)^{1/2}|V_3(-\hat{\mathbf{q}},k\hat{\mathbf{q}},(1-k)\hat{\mathbf{q}})|^2 . \tag{3.13}$$

Here, we have made use of the form (2.7) of the cubic coupling coefficients. Equation (3.13) can be solved with the power-law ansatz:

$$\Gamma_{\hat{\mathbf{q}}}(q)=q^{\eta}\Gamma_{\hat{\mathbf{q}}}(1) \tag{3.14}$$

and

$$\eta=\tfrac{11}{3} , \tag{3.15}$$

$$\Gamma_{\hat{\mathbf{q}}}^{3/2}(1)=\frac{L^2}{8\pi|\Omega(\hat{\mathbf{q}})|^{1/2}}\int_0^1 dk\frac{k^{1/2}(1-k)^{1/2}}{[k^{11/3}+(1-k)^{11/3}]^{1/2}}|V_3(-\hat{\mathbf{q}},k\hat{\mathbf{q}},(1-k)\hat{\mathbf{q}})|^2 . \tag{3.16}$$

Numerical estimates of the quantity $\Gamma_{\hat{\mathbf{q}}}(1)$ for various substances are given in Table I. In obtaining these numbers, we have used the isotropic approximation for the velocities and the displacement field of the Rayleigh modes by using

$$C_l^2=C_{11}/\rho , \tag{3.17a}$$

$$C_t^2=C_{44}/\rho , \tag{3.17b}$$

in the corresponding formulas for these quantities.[23] The elements of the tensor S are chosen to correspond to the situation of a Rayleigh wave propagating in the [100] direction on a (001) surface. The isotropic approximation for the Rayleigh waves should be valid in the case of BaF_2. For the other substances, it can only yield a rough estimate. It should be noticed that the contribution of the collinear spontaneous decay to the damping constant for Rayleigh waves of 100 GHz in silicon obtained in the dispersionless approximation is about 3 orders of magnitude larger than the corresponding value obtained by Tamura[15] from the three-phonon scattering processes at 0.4 K. For a comparison, we quote approximate values for the damping constants of acoustic bulk phonons in BaF_2 at the same frequency from the results of Ref. 19:

$$\Gamma_{\mathrm{LA}}(\nu=100\ \mathrm{GHz})\lesssim 18\ \mathrm{s}^{-1} , \tag{3.18a}$$

$$\Gamma_{\mathrm{TA}}(\nu=100\ \mathrm{GHz})\lesssim 8\ \mathrm{s}^{-1} . \tag{3.18b}$$

Furthermore, from Table I, it can be seen that $\Gamma_{\mathbf{q}}$ is indeed so small compared to $f_{\hat{\mathbf{q}}}(q,q')$, which usually is of the order of magnitude of the Rayleigh wave frequency, that our assumption leading to the approximation (3.12)

TABLE I. Damping constants of Rayleigh modes due to their spontaneous decay, neglecting dispersion. The data correspond to the [100] direction of a (001) surface; isotropic approximation for the Rayleigh-mode velocities and displacement fields. The input parameters are the same as in Ref. 19 except for Cu. Here, the averaged values of Ref. 35 are used for the second- and third-order elastic constants and 8920 kg m^{-3} for the density.

Substance	$\Gamma_R/q^{11/3}$ (10^{-28} m$^{11/3}$ s^{-1})	$\Gamma_R(\nu=0.1$ THz) (10^3 s^{-1})	v_R (10^3 m s^{-1})
BaF_2	3.9	4.5	2.112
NaF	6.3	2.1	2.967
CaF_2	1.5	0.45	3.063
SrF_2	0.75	0.44	2.548
KCl	4.6	12.6	1.677
Si (4 K)	14.3	0.63	5.176
Ge (77 K)	16.4	4.8	3.095
Cu	26.8	14.7	2.596

is justified *a posteriori*. It might not be fulfilled in certain special directions, where the curvature of the slowness curve is very small. In these cases, higher-order terms in the expansion (3.6) have to be taken into account, and the exponent η is changed.

A further assumption required for the results (3.14)–(3.16) to be valid was that the collinear decay is the dominant damping process. We have, however, to be aware of the fact that in the case of strong anisotropy also noncollinear decay comes into play. To calculate its contribution to the damping constant, the approximation (3.2) may be used for sufficiently large angles between $\hat{\mathbf{q}}$ and the propagation directions of the decay products. It can then be seen from a Herring construction[14] that the momentum and strict energy conservation conditions can only be fulfilled simultaneously, if the slowness curve is partly concave. In very anisotropic crystals, the Rayleigh branch can approach the continuum of the transverse bulk modes very closely and can have a phase velocity higher than that of transverse-acoustic bulk phonons in other propagation directions.[25] In such cases, the decay products of a Rayleigh mode may partly consist of bulk phonons without contradicting the general theorem of Lax, Hu, and Narayanamurti.[26] Furthermore, it has been demonstrated for the (001) surface of nickel by Farnell[25] that the Rayleigh mode can, as a function of propagation direction, continuously transform into a bulk mode. At the special direction, where the penetration depth of the surface mode becomes infinite, a localized mode appears in the phonon spectrum with a frequency considerably higher than the lowest bulk transverse phonon with the same wave-vector component in the surface. The damping of this mode at zero temperature may be dominated by the decay into two bulk modes because of the large phase space offered to the bulk modes as decay products, since the momentum-conservation condition does not refer to the vertical components of the wave vectors.

Independent of whether bulk phonons are involved as decay products or not, the contribution of the noncollinear decay of Rayleigh modes (among which we here also count the special localized mode mentioned above) obeys the Herring scaling in the dispersionless approximation, if (3.2) is valid, i.e., $\delta\Gamma_{\mathbf{q}} \sim q^5$.

IV. THE EFFECT OF DISPERSION

As in the case of bulk modes, the dispersion of the phonon frequencies can have important consequences for the collinear and quasicollinear decay. The dispersion can be built into our continuum-theoretical approach, if we replace (3.1) by

$$\omega_{\mathbf{q}} = q v_{\hat{\mathbf{q}}} \left[q - \frac{\mu_{\hat{\mathbf{q}}}}{v_{\hat{\mathbf{q}}}} a_0 q + O(q^2) \right] , \tag{4.1}$$

where a_0 is the lattice constant. In contrast to the case of long-wavelength acoustic bulk phonons, the lowest-order correction in $a_0 q$ to (3.1) is linear in $a_0 q$ for Rayleigh modes. This has been shown by several authors[27,28] for simple lattice-dynamical models. A slightly generalized derivation is given in the Appendix. In Fig. 2, the quantity $\mu_{\hat{\mathbf{q}}}/v_{\hat{\mathbf{q}}}$ is shown as a function of propagation direction

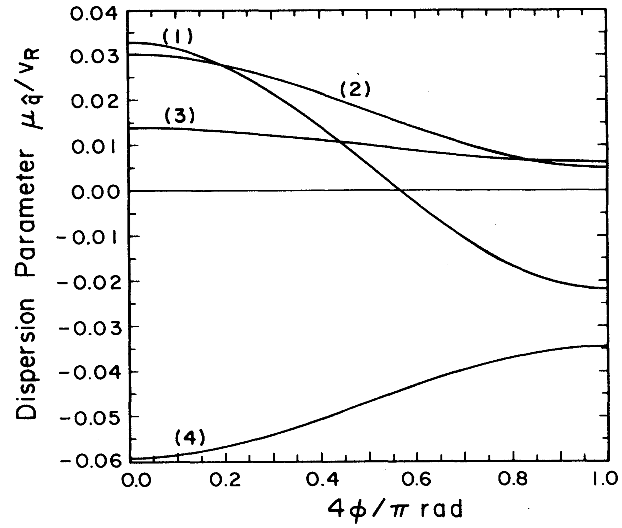

FIG. 2. Dispersion parameter of Rayleigh waves as a function of propagation direction $\hat{\mathbf{q}} = (\cos\phi, \sin\phi, 0)$ calculated from the model in Refs. 23, 28, and 29 for $4C_t^2/3C_l^2 = 0.2(1)$, 0.4(2), 0.6(3), 0.8(4).

$\hat{\mathbf{q}}$, calculated for a free (001) surface of a simple cubic crystal from the model of Gazis, Herman, and Wallis,[29] using the continuum version of the equations of motion and boundary conditions to first order in a_0 given by Maradudin.[23,28] The parameters are chosen such that the elastic constants fulfill the condition of isotropy. Figure 2 shows that the dispersion parameter $\mu_{\hat{\mathbf{q}}}$ can, nevertheless, strongly vary with the direction of propagation. Depending on the ratio of the transverse to longitudinal sound velocity, it can be positive or negative, and for special directions, it may even vanish.

The dispersion of Rayleigh waves discussed so far results from the harmonic approximation to the crystal potential as a consequence of the discreteness of the crystal lattice. We have, however, to be aware of the fact that the anharmonicity can also produce dispersion of the Rayleigh-wave frequencies via the real part of the self-energy, $\Delta_{\mathbf{q}}(\omega)$, taken at the Rayleigh-wave frequency. As has been done by Eckstein and Varga[30] for ^{4}He, one may calculate the frequency correction $\Delta_{\mathbf{q}}(\omega_{\mathbf{q}})$ from the bubble diagram starting with nondispersive bare frequencies and coupling coefficients to obtain to lowest order in q at $T=0$:

$$\Delta_{\mathbf{q}}(\omega_{\mathbf{q}}) = \Delta_{\hat{\mathbf{q}}} q^2 , \tag{4.2}$$

with

$$\Delta_{\hat{\mathbf{q}}} = \frac{-L^2}{8\pi^2} \int^{Q}\!\!\int d^2q' d^2q'' \times \sum_{J',J''} |\tilde{V}_3(\hat{\mathbf{q}}, \mathbf{q}'J', \mathbf{q}''J'')|^2 \times \delta(\mathbf{q}'+\mathbf{q}'') \frac{2}{\omega_{\mathbf{q}'J'} + \omega_{\mathbf{q}''J''}} \tag{4.3}$$

and

$$\tilde{V}_3(\hat{\mathbf{q}},\mathbf{q}'J',\mathbf{q}''J'')=\lim_{q\to 0} q^{-1}V_3(-\mathbf{q},\mathbf{q}'J',\mathbf{q}-\mathbf{q}'J'') \ . \quad (4.4)$$

We thus obtain the result that to lowest order the anharmonicity does not affect the Rayleigh-wave velocity but it does affect its dispersion. The new length scale needed for this is provided by the cutoff Q. The same result (4.2) is obtained from the loop diagram, Fig. 1(b), the effect of which is usually assumed to be of the same order of magnitude as that of the bubble diagram. While the bubble diagram only produces normal dispersion, because of (4.3), the loop diagram may also give rise to anomalous dispersion.

In our perturbative calculation of the damping constants, we use the physical, i.e., the measured Rayleigh-wave frequencies as input parameters. In formulating the perturbation theory in terms of physical quantities, we have to assume that the frequency corrections produced by the anharmonicity are compensated by counterterms in the Hamiltonian.

The basic effect of the dispersion on the (quasi-) collinear decay consists of the introduction of a zeroth-order term in the expansion (3.6) of the form $2\mu_{\hat{\mathbf{q}}}a_0q'(q'-q)$. If this term is sufficiently large compared to $\gamma_{\hat{\mathbf{q}}}$, the approximation (3.2) may be used in (2.1), i.e., low-order perturbation theory is again applicable. The dispersion also modifies the coefficient of the ϕ^2 term in (3.6) and may introduce a term linear in ϕ, but these modifications may be neglected to lowest order in the dispersion. For the quasicollinear situation, the argument of the δ function in (3.2) then is of the following approximate form:

$$\omega_{\mathbf{q}}-\omega_{\mathbf{q}'}-\omega_{\mathbf{q}-\mathbf{q}'} \approx -\left[2\mu_{\hat{\mathbf{q}}}a_0q'|q'-q|+\frac{1}{2}\frac{qq'}{|q'-q|}\Omega(\hat{\mathbf{q}})\phi^2\right] \ . \quad (4.5)$$

The angular bracket can become zero only in the following two cases.

(1) $\mu_{\hat{\mathbf{q}}}>0$ and $\Omega(\hat{\mathbf{q}})<0$, i.e., normal dispersion and $\hat{\mathbf{q}}$ belongs to a concave region of the slowness curve.

(2) $\mu_{\hat{\mathbf{q}}}<0$ and $\Omega(\hat{\mathbf{q}})>0$, i.e., anomalous dispersion and $\hat{\mathbf{q}}$ belongs to a convex region of the slowness curve.

The contribution to the damping constant for these two cases is approximately given by

$$\delta\Gamma_{\mathbf{q}}=q^{9/2}\frac{L^2}{8\pi}|a_0\mu_{\hat{\mathbf{q}}}\Omega(\hat{\mathbf{q}})|^{-1/2} \times\int_0^1 dk|V_3(-\hat{\mathbf{q}},k\hat{\mathbf{q}},(1-k)\hat{\mathbf{q}})|^2 \ . \quad (4.6)$$

In the two complementary cases, quasicollinear decay is forbidden. The damping constant shows a power-law dependence on q with an exponent slightly higher than that found for the nondispersive case, but still lower than five, which would correspond to the Herring scaling. The expression (4.6) for the damping constant is reciprocally proportional to the square root of the curvature of the slowness curve for the respective propagation direction. If this curvature becomes very small, higher-order terms in the expansion of the argument of the δ function in (3.2) with respect to the angle ϕ have to be taken into account, as in the dispersionless case, and the exponent in the power-law dependence of $\Gamma_{\mathbf{q}}$ on q will change. For small dispersion, there is a transitional region, where the approximation (3.2) is not valid and the dispersion cannot be neglected. In this regime, a simple expression for the damping constant like (3.14) and (4.6) in the two boundary cases cannot be obtained.

From the slowness curves of Rayleigh waves shown in Ref. 31, it can be seen that, in the case of copper, at the [001] direction on the (110) surface and the [11$\bar{2}$] direction on the (111) surface, there are pronounced concave regions. For these two examples, we have performed order-of-magnitude estimates for the contribution to the damping constant resulting from (4.6) and listed them in Table II(a). To obtain these values, we have estimated the Rayleigh-wave velocity $v_{\hat{\mathbf{q}}}$ and dispersion parameter $\mu_{\hat{\mathbf{q}}}$ from the results of microscopic calculations of surface phonon dispersion curves, Ref. 32 in the first and Ref. 33 in the second case. The quantity $\Omega(\hat{\mathbf{q}})$ has been obtained from the curvatures of the corresponding slowness curves given in Ref. 31. For the evaluation of the quantity

TABLE II. Estimates for the contribution to the damping constant of Rayleigh modes due to (a) quasicollinear decay and (b) noncollinear decay.

(a)

	$v_{\hat{\mathbf{q}}}$ (10^3 m s^{-1})	$\mu_{\hat{\mathbf{q}}}a_0$ (10^{-8} m^2 s^{-1})	$\kappa(\hat{\mathbf{q}})$ (10^3 m s^{-1})	$\delta\Gamma_R/q^{9/2}$ (10^{-36} m$^{9/2}$ s^{-1})	$\delta\Gamma_R(\nu=0.1$ THz) (10^2 s^{-1})
I[a]	2.1	3.2	2.3	1.4	2.0
II[b]	2.4	7.1	5.6	0.25	0.2

(b)

	$\delta\Gamma_R/q^5$ (10^{-41} m^5 s^{-1})	$\delta\Gamma_R(\nu=0.1$ THz) (10^2 s^{-1})
I[a]	2.4	6.4
II[b]	0.8	1.5

[a]I: [001] direction on the (110) surface of Cu.
[b]II: [11$\bar{2}$] direction on the (111) surface of Cu.

$$I(\hat{\mathbf{q}}):=\frac{L^2 v_{\hat{\mathbf{q}}}^3}{8\pi}\int_0^1 dk\,|V_3(-\hat{\mathbf{q}},k\hat{\mathbf{q}},(1-k)\hat{\mathbf{q}})|^2 \tag{4.7}$$

the isotropic approximation has been used for the displacement field of the Rayleigh waves in the same way as in Sec. III. Although the z dependence of the displacement field, e.g., in the case of the [001] direction in the (110) surface of Cu is governed by two complex constants $a(\hat{\mathbf{q}}r)$ with $a(\hat{\mathbf{q}}1)=a^*(\hat{\mathbf{q}}2)$ (Ref. 34) instead of two real $a(\hat{\mathbf{q}}r)$, the isotropic approximation should still yield a realistic order-of-magnitude estimate.[35]

Since the directions chosen for the two examples are situated in concave regions of the slowness curves, we also have to account for noncollinear-decay processes, illustrated in the Herring plot, in Fig. 3. Here, the angles between the propagation directions of the decaying mode and the decay products are usually large enough for (3.2) to be valid, and for an estimate, we also may neglect the dispersion. The expression for the contribution of these processes to the damping constant may be cast into the following form:

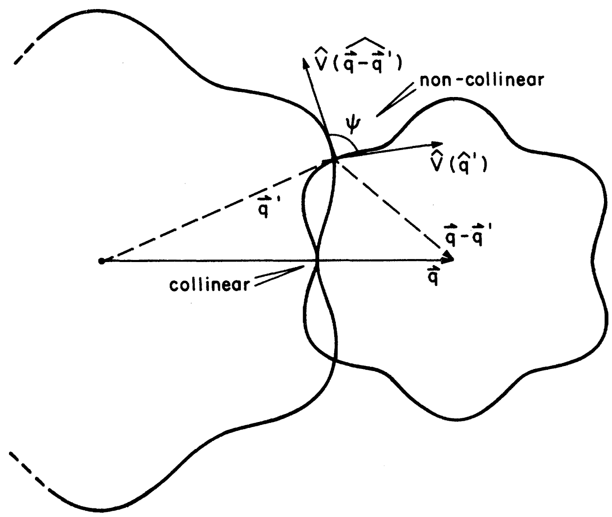

FIG. 3. Herring construction to illustrate the noncollinear decay of Rayleigh modes in the $[11\bar{2}]$ direction on a (111) surface of Cu. The slowness curves have been taken from Ref. 31.

$$\delta\Gamma_{\mathbf{q}}=q^5\frac{L^2}{8\pi}\int_0^{v_{\hat{\mathbf{q}}}} d\omega\,|V_3(-\hat{\mathbf{q}},\mathbf{k},\hat{\mathbf{q}}-\mathbf{k})|^2\left[|\mathbf{V}(\hat{\mathbf{k}})|\left|\mathbf{V}\left(\frac{\hat{\mathbf{q}}-\mathbf{k}}{|\hat{\mathbf{q}}-\mathbf{k}|}\right)\right||\sin\psi|\right]^{-1}, \tag{4.8}$$

where $\omega=kv_{\hat{\mathbf{k}}}$, $\mathbf{V}(\hat{\mathbf{k}})$ is the group velocity, and ψ is the angle between $\mathbf{V}(\hat{\mathbf{k}})$ and $\mathbf{V}((\hat{\mathbf{q}}-\mathbf{k})/|\hat{\mathbf{q}}-\mathbf{k}|)$. The angle ϕ between $\hat{\mathbf{q}}$ and $\mathbf{k}$ as a function of ω may be found by a Herring construction. For a rough order-of-magnitude estimate, we proceed in the following way: The group velocities are approximated by $v_{\hat{\mathbf{q}}}$, $V_3(-\hat{\mathbf{q}},\mathbf{k},\hat{\mathbf{q}}-\mathbf{k})$ is replaced by the collinear matrix element and for $|\sin\psi|^{-1}$, an average value is inserted to yield

$$\delta\Gamma_{\mathbf{q}}=q^5 2v_{\hat{\mathbf{q}}}^{-4}I(\hat{\mathbf{q}})\langle|\sin\psi|^{-1}\rangle\ . \tag{4.9}$$

The data of Table II(b) are obtained from (4.9) with 2 inserted for $\langle|\sin\psi|^{-1}\rangle$. From the values for the partial damping constants in Table II, we may conclude that both types of processes have to be taken into account for frequencies around 100 GHz. For decreasing frequencies the quasicollinear decay should become more and more important.

V. HIGHER-ORDER PROCESSES

In the presence of dispersion, there are always propagation directions where the spontaneous decay via cubic anharmonicity should be absent or its contribution to the damping constant extremely small. To assess the damping of the Rayleigh modes due to anharmonicity in these directions, the analysis of higher-order processes is required, which is, in general, a difficult task. We focus here on the four phonon processes symbolized by the self-energy diagram Fig. 1(c), being aware of the fact that there are further diagrams involving the third-order coupling constants, which yield contributions to the damping constants of the same order of magnitude as the one we consider here.

It has been shown[36] that the contribution from the noncollinear processes of the diagram Fig. 1(c) to the damping constant of bulk phonons follows a scaling law of the form

$$\delta\Gamma_{\lambda\mathbf{q}}(\lambda T)=\lambda^9\delta\Gamma_{\mathbf{q}}(T) \tag{5.1}$$

in the dispersionless approximation. A simple power counting in the general expression for $\delta\Gamma_{\mathbf{q}}(T)$ shows that this is also valid for the damping constant of Rayleigh modes. Although the existence of a second $\mathbf{q}$ integration causes the energy and momentum conservation condition to be less restrictive if compared to the bubble diagram, at $T=0$, the spontaneous decay into three phonons of lower frequency can only take place via collinear processes for phonons of the lowest phonon branch in isotropic or almost isotropic crystals. We will therefore discuss this process in more detail. The expression we have to analyze is of the following form:

$$\delta\Gamma_{\mathbf{q}}=\frac{L^{2D}}{2(2\pi)^{2D}}\int d^Dq'\int d^Dq''|V_4(-\mathbf{q},\mathbf{q}',\mathbf{q}'',\mathbf{q}-\mathbf{q}'-\mathbf{q}'')|^2\frac{\Gamma_{\mathbf{q}'}+\Gamma_{\mathbf{q}''}+\Gamma_{\mathbf{q}-\mathbf{q}'-\mathbf{q}''}}{(\omega_{\mathbf{q}}-\omega_{\mathbf{q}'}-\omega_{\mathbf{q}''}-\omega_{\mathbf{q}-\mathbf{q}'-\mathbf{q}''})^2+(\Gamma_{\mathbf{q}'}+\Gamma_{\mathbf{q}''}+\Gamma_{\mathbf{q}-\mathbf{q}'-\mathbf{q}''})^2}\ . \tag{5.2}$$

Here, $D=3$ for bulk phonons and $D=2$ for Rayleigh modes, and $\mathbf{q},\mathbf{q}'$ and $\mathbf{q}''$ are three- or two-dimensional wave vectors, respectively. By expanding $\omega_{\mathbf{q}}-\omega_{\mathbf{q}'}-\omega_{\mathbf{q}''}-\omega_{\mathbf{q}-\mathbf{q}'-\mathbf{q}''}$ with respect to the angles between $\mathbf{q}'$ and $\mathbf{q}$, and $\mathbf{q}''$ and $\mathbf{q}$ in the same way as for the bubble diagram,[19] it can be shown that for bulk phonons, the collinear, three-phonon decay yields a vanishing contribution.

For Rayleigh modes, we expand

$$\omega_{\mathbf{q}}-\omega_{\mathbf{q}'}-\omega_{\mathbf{q}''}-\omega_{\mathbf{q}-\mathbf{q}'-\mathbf{q}''}=\frac{1}{2}\frac{\Omega(\hat{\mathbf{q}})}{q''+q'-q}[q'(q-q'')\phi'^2+q''(q-q')\phi''^2+2q'q''\phi'\phi'']+O(\phi^3)\ , \tag{5.3}$$

where $q'+q''<q$ and ϕ' is the angle between $\mathbf{q}$ and $\mathbf{q}'$ and ϕ'' the angle between $\mathbf{q}$ and $\mathbf{q}''$. Furthermore, we approximate

$$\begin{aligned}\Gamma_{\mathbf{q}'}+\Gamma_{\mathbf{q}''}+\Gamma_{\mathbf{q}-\mathbf{q}'-\mathbf{q}''}&\approx\Gamma_{q'\hat{\mathbf{q}}}+\Gamma_{q''\hat{\mathbf{q}}}+\Gamma_{(q-q'-q'')\hat{\mathbf{q}}}\\&=:\hat{\gamma}_{\hat{\mathbf{q}}}(qq'q'')\ .\end{aligned} \tag{5.4}$$

After the transformation

$$\phi'=r\cos\beta,\quad \phi''=r\sin\beta\ , \tag{5.5}$$

we may integrate over r in (5.2) and then perform the limit $\hat{\gamma}\to 0$ to obtain

$$\delta\Gamma_{\mathbf{q}}=q^9\frac{L^{2D}}{2^6\pi^3}\int_0^{2\pi}d\beta\int_0^1 dk\int_0^{1-k}dk'kk'|V_4(-\hat{\mathbf{q}},k\hat{\mathbf{q}},k'\hat{\mathbf{q}},(1-k-k')\hat{\mathbf{q}})|^2 g_{\hat{\mathbf{q}}}^{-1}(\beta,k,k') \tag{5.6}$$

with

$$g_{\hat{\mathbf{q}}}(\beta,k,k')=\frac{\Omega(\hat{\mathbf{q}})}{k+k'-1}\{kk'[\sin(2\beta)-1]+k(1-\sin^2\beta)+k'\sin^2\beta\}\ . \tag{5.7}$$

The integrals in (5.6) are well behaved, so that we have obtained the following result: The collinear, three-phonon decay of Rayleigh modes in the dispersionless approximation yields a finite contribution to the damping constant, which scales with the same power of the frequency as the noncollinear, three-phonon decay of bulk phonons.

The influence of the frequency dispersion can be assessed in the same way as for the two-phonon decay. It introduces a zero-order term of the form

$$-2\mu_{\hat{\mathbf{q}}}a_0\{q'(q-q'-q'')+q''(q-q'')\} \tag{5.8}$$

in the expansion (5.3). Proceeding in the same way as for the bubble diagram and realizing that the curly brackets in (5.7) and (5.8) are always positive, we are led to the following conclusion: For propagation directions with anomalous dispersion in a convex region of the slowness curve or with normal dispersion in a concave region of the slowness curve, the spontaneous, quasicollinear, three-phonon decay yields a contribution to the damping constant, which is to lowest-order independent of $\mu_{\hat{\mathbf{q}}}$ and twice the right-hand side of (5.6). In the complementary cases, it is forbidden. This means, in particular, that if the quasicollinear two-phonon decay is forbidden, the analogous processes involving three decay products are also not present. The same nonanalytic behavior of the damping constant as a function of the dispersion parameters is also known in the case of two-phonon-decay processes for bulk modes.[37,19]

VI. CONCLUSIONS

The goal of this investigation has been to derive the frequency dependence and to give estimates for the order of magnitude of the intrinsic damping constants of surface acoustic phonons of long wavelength at zero temperature, which has been unknown so far. It has been shown that because of the restricted phase space for the products of the spontaneous-decay processes, the damping constant depends sensitively on the shape of the slowness curve and the frequency dispersion. Within continuum elasticity theory, neglecting the dispersion, a finite result for the damping constant from collinear processes was obtained, which is proportional to $q^{11/3}$ and gives rise to lifetimes of the order of milliseconds for 100-GHz Rayleigh modes in various substances. Normal dispersion, if large enough, strongly modifies this result causing the decay via both three- and the analogous four-phonon processes to be forbidden in convex regions of the slowness curve. This implies that for nearly isotropic crystals under normal circumstances the intrinsic damping constant of Rayleigh modes will be governed by thermal processes of the kind considered by Tamura[15] down to very low temperatures. For propagation directions in concave regions of the slowness curve, however, quasicollinear as well as truly noncollinear processes occur. Their contributions to the damping constant depend on the frequency via power laws with slightly differing exponents. For two geometries in the case of copper, these contributions have been estimated to be both of the order of 10^2 s for 100-GHz modes. In convex regions of the slowness curve, quasicollinear decay can only occur if the dispersion is anomalous.

The results summarized above only refer to the generalized Rayleigh modes. It is, however, well known that in certain geometries further acoustic surface branches of higher frequencies exist, e.g., the shear horizontal branch in the [110] direction on a (001) surface of certain cubic crystals.[22,23,28] The damping constants of these surface acoustic phonons are expected to be dominated by the spontaneous decay into surface phonons of lower branches, in particular Rayleigh modes. For the above-mentioned shear horizontal modes at long wavelengths, these processes yield a damping constant proportional to q^6. To obtain this q dependence, one has to take account of the fact that the penetration depth of these shear horizontal modes is proportional to q^{-2}.

An experimental verification of our theoretical predictions on the frequency dependence of the damping constants at zero temperature will require special methods, since because of their smallness and strong decrease with frequency, they are not accessible to the current techniques used to investigate surface phonons. Furthermore, the damping due to other mechanisms like surface roughness and imperfections has to be controlled in such a way that the intrinsic damping can be distinguished from it. It is hoped that techniques similar to those used to detect the spontaneous decay of bulk phonons can, in modified form, also be applied to the surface.

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APPENDIX: DISPERSION OF RAYLEIGH WAVES

The derivation of the dispersion of the Rayleigh wave frequencies to lowest order in $a_0 q$ given here is based on an equation of motion and boundary conditions for the displacement field of the following form:

$$\rho \ddot{u}_\alpha = \sum_{\beta,\mu,\nu} \left[C_{\alpha\mu\beta\nu} u_{\beta|\mu\nu} + a_0^2 \sum_{\zeta,\xi} \Lambda_{\alpha\beta\mu\nu\zeta\xi} u_{\beta|\mu\nu\zeta\xi} \right] , \tag{A1}$$

$$0 = \sum_{\beta,\nu} \left[C_{\alpha z\beta\nu} u_{\beta|\nu} + \frac{a_0}{2} \sum_{\mu} D_{\alpha z\beta\mu\nu} u_{\beta|\nu\mu} \right] \Bigg|_{z=0} , \tag{A2}$$

where a_0 is the lattice constant. That this system of equations for the displacement field will yield the correct expression for the dispersion in the frequencies of long-wavelength acoustic phonons has been proven only for the (001) surface of a simple cubic crystal with interactions that couple adjacent layers only.[28] In this case, the nonvanishing elements of the tensor D can be expressed by the elastic constants:

$$\begin{aligned}
&D_{xzxxx} = C_{12}, \quad D_{xzxzz} = C_{44}, \quad D_{xzzxz} = D_{xzzzx} = \tfrac{1}{2}(C_{12} + C_{44}) , \\
&D_{yzyyy} = C_{12}, \quad D_{yzyzz} = C_{44}, \quad D_{yzzyz} = D_{yzzzy} = \tfrac{1}{2}(C_{12} + C_{44}) , \\
&D_{zzxxz} = D_{zzxzx} = \tfrac{1}{2}(C_{12} + C_{44}), \quad D_{zzyyz} = D_{zzyzy} = \tfrac{1}{2}(C_{12} + C_{44}) , \\
&D_{zzzxx} = D_{zzzyy} = C_{44}, \quad D_{zzzzz} = C_{11} .
\end{aligned} \tag{A3}$$

It will be seen that the correction to the frequencies is of first order in $a_0 q$. Therefore, the tensor Λ does not enter the expression for the frequency to lowest order.

Let $u_\alpha^{(1)}$ be a solution for Eqs. (A1) and (A2) corresponding to a surface wave with two-dimensional wave vector $\mathbf{q}$, to first order in $a_0 q$ and $u_\alpha^{(0)}$ the corresponding solution in the absence of the tensor D, i.e., an ordinary Rayleigh wave. Insertion of $u_\alpha^{(1)}$ into (A1) multiplying by $u_\alpha^{(0)*}$, summing over α and integrating over $\mathbf{x}$ with periodic boundary conditions in the x-y plane yields

$$-\rho\omega^2 \sum_\alpha \int d^3x\, u_\alpha^{(0)*}(\mathbf{x}) u_\alpha^{(1)}(\mathbf{x}) = \sum_{\alpha,\beta,\mu,\nu} C_{\alpha\mu\beta\nu} \int d^3x\, u_\alpha^{(0)*}(\mathbf{x}) u_{\beta|\mu\nu}^{(1)}(\mathbf{x}) . \tag{A4}$$

We now integrate the right-hand side of (A4) twice by parts using the boundary condition (A2), and make use of the fact that $u_\alpha^{(0)}$ is a solution of the zeroth-order equation with frequency ω_0 satisfying the zeroth-order boundary conditions. Retaining only terms to first order in $a_0 q$, we arrive at

$$\rho(\omega_0^2 - \omega^2) \sum_\alpha \int d^3x\, u_\alpha^{(0)*}(\mathbf{x}) u_\alpha^{(0)}(\mathbf{x}) = -\frac{a_0}{2} \sum_{\alpha,\beta,\mu,\nu} D_{\alpha z\beta\mu\nu} \int dx\, dy\, u_\alpha^{(0)*}(z=0) \times u_{\beta|\mu\nu}^{(0)}(z=0) . \tag{A5}$$

Equation (A5) no longer contains $u_\alpha^{(1)}$. Since $u_\alpha^{(0)}$ has the form (2.3) and is assumed to be normalized, expression (4.1) is obtained for the Rayleigh-wave frequency with

$$\mu_{\hat{\mathbf{q}}} = \frac{-1}{4\rho v_{\hat{\mathbf{q}}}} \sum_{\alpha,\beta,\mu,\nu} D_{\alpha z\beta\mu\nu} \sum_{r,r'} b_\alpha^*(\hat{\mathbf{q}}r') b_\beta(\hat{\mathbf{q}}r) [i\hat{q}_\mu(1-\delta_{\mu z}) + a(\hat{\mathbf{q}}r)\delta_{\mu z}][i\hat{q}_\nu(1-\delta_{\nu z}) + a(\hat{\mathbf{q}}r)\delta_{\nu z}] . \tag{A6}$$

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