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Light scattering in a phonon gas

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Light-scattering spectrum in dielectric crystals is studied with extended thermodynamics (ET) for a phonon gas, covering from hydrodynamic to ballistic (collisionless) regimes. The ET equation is solved to obtain the power spectrum of the energy density in a phonon-gas mixture, which consists of interacting phonon gases of longitudinal and transverse acoustic (LA and TA) phonon modes. In the hydrodynamic regime, where phonon collisions take place frequently, it is found that the light-scattering spectrum consists of two components, which can be interpreted as the two normal modes formed by the two second-sound modes defined in each of the LA and TA phonon gases. Out of the two normal-mode spectra, the low-frequency component arises from thermal fluctuations and gives rise to a narrow quasielastic spectrum, which corresponds to the well-known thermal Rayleigh scattering due to thermal diffusion, i.e., due to overdamped second sound. With sufficient momentum-conserving phonon collisions (normal phonon scattering), the narrow quasielastic component is demonstrated to develop, from the diffusive central peak, into a pair of shifted inelastic peaks due to propagation of underdamped second sound. The other spectrum component gives rise to a much broader quasielastic scattering, whose wing extends out to the Brillouin-scattering lines of the LA and TA phonons (first sounds). The broader quasielastic spectrum has a linewidth equal to the phonon-collision rate, which suggests that this component originates from a nonequilibrium process in the phonon gas. As the ballistic regime is approached, the line shapes and linewidths of the two normal-mode spectra approach each other, and the two components finally coincide in the limit of the ballistic regime, which is in good agreement with the reported behavior for these spectra that were experimentally observed in many crystals. In the ballistic limit, the light-scattering mechanism in the present model is found to become formally equivalent to the previously proposed microscopic framework, i.e., the second-order difference Raman scattering (two-phonon difference light scattering) on a same phonon dispersion. The derived spectral formula is fitted to the spectra previously observed in the experiments for rutile (TiO$_2$) and strontium titanate (SrTiO$_3$). The fits are quite successful in wide ranges of frequency and temperature, i.e., regardless of degree of nonequilibrium, owing to the ET analysis. The relaxation times for the normal and resistive phonon collisions ($\tau_N$ and $\tau_R$) are determined through the analysis. The temperature dependences of $\tau_N$ and $\tau_R$ indicate that the origin of the broad shifted peaks (the “broad doublet”), which were observed in SrTiO$_3$ at around 30 K, is likely due to underdamped second sound at least in a narrow temperature range around 30 K.

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I. INTRODUCTION

Transport phenomenon is a fundamental nonequilibrium aspect in many-body systems, where thermal equilibrium is realized through the interactions among constituent particles such as molecules, electrons, phonons, etc. One of the most important transport phenomena is the energy transport in dielectric solids, where the energy (or “heat”) is carried only by phonons at a velocity of sound speed. Studying the energy transport in dielectric solids brings us understandings of the mechanism of thermal diffusion, wavelike propagation of heat (“second sound”), ballistic heat conduction. In dielectric solids, the conduction of heat is realized through the lattice anharmonicity, which allows for collisions between phonons in a “phonon gas.”

In early version of nonequilibrium physics that deals with dissipation including diffusion phenomena, it was essential to assume that thermal equilibrium is always achieved locally in a “small cell” of the relevant system. Owing to this local-thermal-equilibrium hypothesis, local thermodynamic variables, such as local temperature, local entropy, etc., may be defined in the length scale of the small cells, and it is possible that the entire system is not necessarily in thermal equilibrium as a whole. For local thermal equilibrium to be achieved in the small cells, the mean free path of the energy quanta (such as molecules, phonons, electrons, etc.) in the cells must be sufficiently shorter than the dimension of the small cells; otherwise, we see the nonequilibrium kinetic behavior inside the individual cells. If the mean free path of the quanta is comparable to the observation length scale, the memory effect of successive collisions between the energy quanta should not be neglected. Since the mean free path of phonons in most dielectric solids can range from nm to $\mu$m or even to mm scales as the temperature is reduced, we cannot always assume local thermal equilibrium within a fixed length scale. Therefore, it is possible that local thermal equilibrium is violated due to temperature change. If local thermal equilibrium is violated, we must consider the correlation between the phonon collisions; such process involves frequency and wave-vector dependences of the transport coefficients such as thermal conductivity or diffusivity.

We have recently reported on light-scattering experiments in crystals of rutile, ZnSe, silicon, and SrTiO$_3$. We found that, at relatively high temperatures, the low-frequency part of the light-scattering spectra consisted of two quasielastic components. Out of the two quasielastic components, the narrower component broadened on cooling, and it had a line-
width that could be well reproduced as $D_{\text{def}}^2$ at high temperatures, where $D_b$ is the thermal diffusivity and $q$ is the wave-vector transfer in the experiments. However, the observed linewidth of the narrower quasielastic component did not follow $D_{\text{def}}^2$ at low temperatures, showing that the observation length scale of $q^{-1}$, which was $\approx 100$ nm, was too short compared to the mean free path of phonons, which becomes longer at low temperature because the number of the thermally excited phonons decreases as the temperature is lowered. The other quasielastic component, the broader one, showed opposite temperature dependence for the linewidth, i.e., it narrowed on cooling, and had no $q$ dependence; therefore, the two quasielastic components approached each other on cooling. However, we could not quantitatively analyze such a behavior in the low-temperature region due to the lack of theories that cover the whole temperature range. In SrTiO$_3$, it has been reported by Hehlen et al.\textsuperscript{6} that an extra soundlike peak appears in the Brillouin-scattering spectrum around 30 K, and they tentatively ascribed the extra Brillouin peak to the second sound, the wavelike propagation of heat,\textsuperscript{7} basing on a theoretical work by Gurevich and Tagantsev.\textsuperscript{8} We found that the extra Brillouin spectrum was seemingly developed from the thermal-diffusive (the narrow) quasielastic component. Since thermal diffusion is considered as overdamped second sound,\textsuperscript{10–12} the spectrum of second sound should transform gradually from a quasielastic profile into a pair of shifted peaks as the thermal wave becomes underdamped. However, in order to deal with all of the above aspects of the low-frequency light-scattering spectra (thermal diffusion, second sound, nonequilibrium heat transport, the broader quasielastic component, etc.), it is essential to formulate the nonequilibrium light-scattering theory that can be applied not only when local thermal equilibrium can be assumed but also when it cannot be established.

Recently, a method of nonequilibrium thermodynamics has been developed,\textsuperscript{5} and it has succeeded in reproducing the pressure dependence of the light-scattering spectrum in rarefied gases. Also, thermal diffusion, propagation of second sound, and ballistic phonon propagation in NaF (Ref. 13) has been well simulated from relatively simple formalism with phonon-gas model.\textsuperscript{5,14} Although the theoretical framework that is called “extended thermodynamics (ET)” seems to be still under test, it is becoming a powerful tool for the analysis of experimental data that concerns highly nonequilibrium processes.\textsuperscript{5,6} In this paper, we solve a set of ET equations for a phonon-gas system to obtain the formula for the light-scattering spectra that is applicable from equilibrium to nonequilibrium regimes in dielectrics. In Sec. II, the derivation of the spectral formula is presented. The narrow central peak due to thermal diffusion (thermal Rayleigh mode), the broad Brillouin doublet due to second sound, and the broad central peak (Mountain mode in crystals) are discussed in that section. Also, we discuss how the nonequilibrium energy-transport affects those “equilibrated” spectra when the nonequilibrium is approached. In Sec. III, we fit the derived expression to the experimentally observed spectra for rutile and SrTiO$_3$ crystals. The possibility of second sound in these systems is discussed in Sec. IV, and it is concluded that the reported broad doublet in SrTiO$_3$ (Ref. 8) is likely due to second sound. Finally, we summarize this report in Sec. V.

II. THEORY

A. Extended thermodynamic equations for a mixture of phonon gases

In order to realistically characterize the phonon gas in a dielectric, we consider the anharmonic interactions between different phonon modes, namely, longitudinal and transverse acoustic (LA and TA) modes, rather than considering only a single phonon mode. Following the treatments in the review by Dreyer and Struchtrup,\textsuperscript{5,14} who have carried out ET analyses on boson gases, the ET equations to be solved is given as follows for one-dimensional case:\textsuperscript{14}

$$\frac{\partial e^{(i)}}{\partial t} + c_{0,i}^2 \frac{\partial e^{(i)}}{\partial x} = -2A(e^{(1)}_0 e^{(i)}_0 - e^{(2)}_0 e^{(i)}_0)$$

$$\frac{\partial p^{(i)}}{\partial t} + \frac{1}{3} \frac{\partial e^{(i)}}{\partial x} + \frac{\partial N^{(i)}_{(x)}}{\partial x} = -\frac{1}{\tau_R} p^{(i)}_x - 2B(e^{(2)}_0 p^{(i)}_0 - e^{(3)}_0 p^{(i)}_0)$$

$$\frac{\partial e^{(0)}}{\partial t} + c_{0}^2 \frac{\partial e^{(0)}}{\partial x} = A(e^{(1)}_0 e^{(0)}_0 - e^{(2)}_0 e^{(0)}_0)$$

$$\frac{\partial p^{(0)}}{\partial t} + \frac{1}{3} \frac{\partial e^{(0)}}{\partial x} + \frac{\partial N^{(0)}_{(x)}}{\partial x} = -\frac{1}{\tau_R} p^{(0)}_x + B(e^{(2)}_0 p^{(0)}_0 - e^{(3)}_0 p^{(0)}_0)$$

$$\frac{\partial e^{(n-1)}}{\partial t} + c_{0,n}^2 \frac{\partial e^{(n-1)}}{\partial x} + \frac{\partial e^{(n)}}{\partial x} = -\left(\frac{1}{\tau_{n-1}^R} + \frac{1}{\tau_R} \right) e^{(n-1)}$$

Here, the traceless symmetric parts of a tensor of rank $n$, $T_{ij\cdots j}^{(n)}$, are denoted by $T_{ij1\cdots j}^{(n)}$, and further abbreviation such as $T_{ij1\cdots j}^{(n)} = T_{ij}^{(n)}$ is used.\textsuperscript{14} The moments of the phase density (distribution function) $f$ are defined by the equation:\textsuperscript{14}

$$u_{ij\cdots k} = u_{i1\cdots k} = h \int \left( e^{(n)} \right)^{n-1} k_{ij1} \cdots k_{ijk} dk.$$

For $n=0,1,2,\ldots$, we obtain $e^{(n)}$ and $p^{(n)} = u_{ij}^{(n)} = u_{ij}^{(n)}$, $N^{(n)}_{(x)} = u_{ij}^{(n)}$, and $u_{ij}^{(n)} = u_{ij}^{(n)}$, which are, respectively, the energy density, the momentum density, the momentum flux (the traceless symmetric part of the stress tensor), and the general flux deviator of order $n$.\textsuperscript{5,14} $k_{ij}$ is the phonon mode LA or TA, respectively, $c_{ij}$ is the sound velocity, and $\frac{1}{\tau_R}$ and $\frac{1}{\tau_n}$ are the relaxation times for the resistive ($R$) and normal ($N$) processes of phonon-phonon
scattering, respectively. Note that the $R$ process include the Umklapp and other nonmomentum-conserving collisions between phonons, while the $N$ process does not destroy momentum conservation. $\alpha_n$ is defined by

$$\alpha_n = \frac{(n-1)^2}{4(n-1)^2 - 1}. \quad (3)$$

The coupling constants responsible for the exchanges of energy and momentum between the two sound modes are defined, respectively, by

$$A = \frac{1}{4R^3} + 2\frac{d_0}{R^5} + \frac{1}{4N^3} + 2\frac{d_0}{N^5}, \quad (4)$$
$$B = \frac{1}{4N^5} + 2\frac{d_0}{N^5}. \quad (5)$$

The set of Eqs. (1a) and (2c) consists of $2n$ equations, i.e., $n$ balance equations for the “fields” for each of the modes. Each equation is obtained from the Boltzmann equation multiplied by $u_{n0}$ and then integrated over the whole Brillouin zone, combined with Callaway’s relaxation-time approximation,\textsuperscript{15} which assumes two relaxation times, $\tau_N$ and $\tau_R$. Equation (1a), the first balance equation for the LA mode, is a continuity equation for the energy density with a production term (the right-hand side) due to the interaction with the other modes (two TA modes). The total energy $e = e^{(1)} + 2e^{(2)}$ is considered, but there is an exchange of energy between the modes unless $c_0^3e^{(1)} - c_0^3e^{(0)} = 0$ holds. Due to $N$ processes, there is an exchange of momentum between the modes unless $c_0^5p_x - c_0^5p_x^{(0)} = 0$.

In this ET model, we consider all the phonons in the whole Brillouin zone, i.e., not only the long-wavelength phonons but also those with much shorter wavelength (much larger wave vectors).\textsuperscript{14} It should be pointed out that the phonon lifetime is shorter for phonons with larger wave vectors. Therefore, phonon collisions within one branch as well as those between different branches can be considered in this model. The intramode phonon collision is expected to occur far less frequently in crystals\textsuperscript{10} if one neglects the life time of thermal phonons (i.e., in the ballistic regime), but, in general, one should include not only the intermode but also the intramode phonon collisions in order to account for the collective excitations in a phonon gas. Another remark is that the one-dimensional model assumes that the phonons traveling in all the directions can participate in the collisions.

**B. Light scattering in phonon gas**

Light scattering occurs through the modulation in dielectric constant $\varepsilon$ by the fluctuations in the thermodynamic variables such as the density or temperature in the medium. In Landau-Placzek theory,\textsuperscript{15} the fluctuation in $\varepsilon$ is described by the following relation:

$$\varepsilon = \frac{\partial \varepsilon}{\partial \rho} / T \delta \rho + \frac{\partial \varepsilon}{\partial T} / T \delta T, \quad (6)$$

where $\rho$ and $T$ are the density and temperature, respectively. In phonon picture, this is expressed equivalently as\textsuperscript{18,19}

$$\delta \varepsilon = \varepsilon^{(1)} \Lambda^{(0)} + \sum_q \varepsilon^{(2)}(q_0) \Lambda^{(0)}(q_0) \Lambda^{(0)}(q_0), \quad (7)$$

where $p_1$ and $p_2$ are the first- and second-order Raman coefficients, respectively, and $A$ is the phonon normal coordinate. $p_1$ gives (ordinary) one-phonon scattering (first-order Raman scattering), and $p_2$ gives second-order Raman scattering, where number-density fluctuations in the phonon gas is responsible for the light-scattering mechanism. The power spectrum, $\langle \delta \varepsilon^{(1)} \delta \varepsilon^{(1)} \rangle$, calculated from Eq. (6) and (7) contains three types of terms: (i) scattering from a single phonon; (ii) scattering from a pair of phonons; (iii) the interference between the contributions of (i) and (ii). In ordinary fluids, only the contribution of (i) is usually considered because the second contribution is much smaller than the first due to relatively large thermal-expansion effect, which indirectly connects the temperature change to the dielectric constant.\textsuperscript{17} The Brillouin scattering from elastic sound wave originates from the contribution from the $p_1$ term, i.e., it is the first-order scattering in which a single phonon is relevant.

In contrast to light scattering in ordinary fluids, the second-order term, (ii), plays a significant role in solids, and it gives rise to light scattering directly due to thermal fluctuations, i.e., the dielectric constant is modulated directly (not via thermal-expansion effect) by the fluctuations in number density of phonons.\textsuperscript{18} Since the number density of phonons is proportional to the energy density $[\varepsilon(x, t)]$ in the phonon gas, its power spectrum, $\langle \varepsilon^{(2)} \varepsilon^{(2)} \rangle$, which we will calculate from the ET equations, describes only the contribution from the second term in the right-hand side of Eq. (7). The power spectra, $\langle \varepsilon^{(2)} \varepsilon^{(2)} \rangle$, should not include the first-order scattering because it contains at least four phonon coordinates, and the Brillouin scattering from a sound wave will not appear in our calculated spectrum, which will be presented below. Since we have been interested in the quasielastic light scattering in solids, including thermal Rayleigh scattering or other low-energy scattering involving a collection of phonons, we will focus only on the second-order scattering, which can be described by the power spectrum of the energy density in the phonon gas.

**C. Frequency- and wave-vector-dependent viscosity in phonon gas**

To clarify the system described by the ET equations of Eqs. (1) and (2), we first consider one-branch phonon ET equation. We formally set the coupling constants to be zero, viz., $A = B = 0$, in Eqs. (1) and (2), and the system reduces as the following:

$$\frac{\partial \varepsilon}{\partial t} + \varepsilon \frac{\partial p_x}{\partial x} = 0$$
$$\frac{\partial p_x}{\partial t} + \frac{1}{5} \frac{\partial \varepsilon}{\partial x} + \frac{\partial N_{(xx)}}{\partial x} = - \frac{1}{\tau_R} p_x, \quad (8)$$

where we have dropped the mode suffix. Taking Fourier-Laplace transform, we obtain...
where we have defined the natural frequency of the phonon gas as a frequency- and wave-vector-dependent viscosity in a phonon gas.

From Eq. (8), and taking into account the mutual independence between $e$ and $p$, the correlation function $\langle e^*(q,0)e(q,s) \rangle$ is defined by a recurrence relation as

$$\langle e^*(q,0)e(q,s) \rangle = \frac{1}{s + \frac{1}{\tau} + \gamma_s(q,s)},$$

and can be expressed as a continued-fraction form as

$$\gamma_s(q,s) = \frac{\alpha_s e^2 q^2}{s + \frac{1}{\tau} + \frac{\alpha_s e^2 q^2}{s + \frac{1}{\tau} + \frac{\alpha_s e^2 q^2}{s + \frac{1}{\tau} + \cdots}}},$$

where we have defined $\tau$ by

$$\frac{1}{\tau} = \frac{1}{\tau_R} + \frac{1}{\tau_N}.$$

If we write the complex quantity $\gamma_s(q,s)_{|_{\omega_{\text{ini}}}}$ as

$$\gamma_s(q,s)_{|_{\omega_{\text{ini}}}} = \gamma'_s + i\gamma''_s,$$

where $\gamma'_s$ and $\gamma''_s$ are real quantities, we obtain the scattering spectrum $S(q,\omega)$ as the real part of $\langle e^*(q,0)e(q,s) \rangle_{|_{\omega_{\text{ini}}}}$

$$S(q,\omega) = \text{Re}[\langle e^*(q,0)e(q,s) \rangle_{|_{\omega_{\text{ini}}}}]$$

$$= \text{Re}\left[ \frac{\omega_0^2 - \omega^2 - \omega\gamma'_s}{\omega_0^2 - \omega^2 - \omega\gamma'_s + \omega^2 \left( \frac{1}{\tau_R} + \gamma'_s \right)^2} \right],$$

where the natural frequency of the phonon gas has been introduced as

$$\omega_0 = \frac{1}{\sqrt{3}} \omega_q,$$

where $s$ is the Laplace operator, and the contributions from the higher order moments than $p(q,s)$, namely, from $u_2(q), u_3(q), \ldots$, have been renormalized into $\gamma_s(q,s)$, which is regarded as a frequency- and wave-vector-dependent viscosity in a phonon gas.

If $\tau_R$ is sufficiently short such that $1/\tau_R \gg \gamma_s(q,i)$, then $\gamma_s(q,i)$ can be neglected and the right-hand side of Eq. (13) can be approximated as a Green function for a usual damped harmonic wave, whose resonance frequency and damping rate are given by $\sqrt{\omega_0^2 - 1/4\tau_R^2}$ and $1/\tau_R$, respectively. For weak damping, therefore, there can be a wave of phonon density, i.e., wavelike propagation of temperature, which is known as the “second sound.”

In contrast, if $\tau_R$ is long such that $\gamma_s(q,s)$ cannot be neglected, then, effects of frequency- and wave-vector-dependent viscosity appear as modifications to the resonance frequency and to the line-width of the second sound as can be found in Eq. (15). In the latter case, local thermal equilibrium within a time scale of $1/\omega_0$ can no more be achieved, and this nonequilibrium regime is referred to as “hydrodynamic” or “collision-dominated.” These modifications to the second-sound frequency and linewidth can be thought of as arising from memory effects in dissipative processes in phonon gas, and in that sense, $\gamma_s(q,s)$ is referred to as the “memory function.” In the present case, if $\tau$ is short enough for $\gamma_4, \gamma_5, \ldots$ to be neglected, we see that $\gamma_s(q,\omega)$ describes a Debye-type relaxation. Since the inverse Laplace transform of $\gamma_s(q,s)$ yields that

$$\gamma_s(q,i) = \alpha_s e^2 q^2 e^{-\tau},$$

we see that the memory function can be approximated as an exponential function when $\tau_s \tau < 1$. On the other hand, higher order terms must be incorporated if $\tau$ is long such that $\gamma_{m+1} \tau > 1$, and in general, an infinitely continued fraction must be considered as the memory function in the ballistic regime.

### D. Coupling between two second sounds

In this section, we now consider the coupling between the two second sounds for the LA and TA phonon gases. From the discussions described in the Sec. II C, each of Eqs. (1) and (2) can be viewed as two damped harmonic waves (second sound) with damping due to viscosity. The contributions to the viscosity from the moments of higher order than $p$ can be renormalized into $\gamma_s(q,s)$ in the Fourier-Laplace space. Therefore, instead of considering the whole moment equations in Eqs. (1) and (2), we take only Eqs. (1a), (1b), (2a), and (2b), replacing $1/\tau^{(1)}_R$ with $1/\tau^{(1)}_R + \gamma_s(q,s)$. Now the equation system in the Fourier-Laplace space becomes...
By eliminating \( p^{(0)}(q,s) \) and \( p^{(0)}(q,s) \), we obtain two coupled wave equations for the second sounds for the LA and TA phonon gases:

\[
\begin{align*}
\left[ s^2 + \left( \frac{1}{\tau_R} + \gamma_3^{(0)}(q,s) \right) + 2c_0^3(A + Bc_0^3) \right] s + \frac{1}{3}c_0^2q^2 \\
- 2Ac_0^3 + 0 = 0 \\
- Bc_0^5 - \frac{1}{3}q^2 + 0 = 0 \\
\end{align*}
\]

\[
\begin{pmatrix}
2Ac_0^3 & ic_0^2q \\
\frac{1}{3}q & s + \left[ 1/\tau_R + \gamma_3^{(0)}(q,s) \right] + 2Bc_0^5 \\
- Ac_0^3 & 0 \\
0 & - Bc_0^5 \\
\end{pmatrix}
\begin{pmatrix}
e^{(0)}(q,s) \\
e^{(0)}(q,s) \\
e^{(0)}(q,s) \\
e^{(0)}(q,s) \\
\end{pmatrix} = 0.
\]

(15)

First, the coupling constants \( A \) and \( B \) become, respectively, as

\[
A = \frac{1}{3c^2}\left( \frac{1}{\tau_R} + \frac{1}{\tau_N} \right), \quad B = \frac{1}{3c^2}\tau_N.
\]

(20)

If we assume a plane-wave solution, we find that the determinant of the characteristic matrix on the left-hand side of Eq. (18) for the system can be factorized into two simple quadratics in \( s \):

\[
M_{11}M_{22} - M_{12}M_{21} = \begin{pmatrix}
2 + \frac{1}{\tau_R} + \gamma_3^{(0)}(q,s) \\
2 + \frac{1}{\tau} + \gamma_5^{(0)}(q,s) \\
\end{pmatrix} s + \omega_0^2
\]

(16)

Setting \( s \to i\omega \), we obtain the eigenvalues (the normal-mode frequencies) as

\[
\omega_1 = \pm \sqrt{\omega_0^2 - \frac{1}{4} \left( \frac{1}{\tau_R} + \gamma_3^{(0)}(q,s) \right)^2 + \frac{1}{2} \left( \frac{1}{\tau_R} + \gamma_5^{(0)}(q,s) \right)},
\]

(21)

\[
\omega_2 = \pm \sqrt{\omega_0^2 - \frac{1}{4} \gamma_3^{(0)}(q,s) + \frac{1}{2} \gamma_5^{(0)}(q,s)}.
\]

(22)

It is worth noting that, without the approximation made in Eq. (19), the determinant of the characteristic matrix is not to be factorized into such two simple quadratics in \( s \); it is too complicated to display here. Even in that case, however, the four eigenvalues can be classified into two types qualitatively similar to Eqs. (21) and (22).
In the present system of "two coupled second sounds," the amplitudes of the two oscillations are the local temperatures in the LA and TA phonon gases [see Fig. 1(b)]. Therefore, if we assume the approximation as in Eq. (19), there is no dissipation from one temperature wave to the other in the "in-phase" mode (Mode 1), because there is no temperature gradient between the two temperature waves; one of the two waves does not drag the other wave. Thus, in the "in-phase" normal mode, the viscous coupling between the second sounds for the LA and TA phonon gases is inactive when we assume the approximation as in Eq. (19), and is very weak even in general cases. On the other hand, in the "out-of-phase" normal-mode (Mode 2), the local temperature changes in the two second sounds have opposite signs [see Fig. 1(b)], and hence, there is a temperature gradient between the two waves. Thus, one of the two waves strongly drags the other wave via the viscosity of the medium phonon gas.

If Eq. (19) is assumed, the spectra for the in-phase mode (Mode 1) and out-of-phase mode (Mode 2) are obtained, respectively, as

\[ S_1(q,s) = \frac{1}{s + \frac{1}{3} c_0^2 q^2 + \frac{1}{\tau_R} + \gamma_3(q,s)} \]

and

\[ S_2(q,s) = \frac{1}{s + \frac{1}{\tau} + \frac{1}{3} c_0^2 q^2 + \frac{1}{\tau} + \gamma_3(q,s)} \]

\[ S_1(q,s) \] is identical with the right-hand side of Eq. (9), which is the spectrum for an independent (noninteracting) second sound: its properties have been described in Sec. II C. \( S_2(q,s) \), on the other hand, includes the interaction between the two second-sound waves for the LA and TA phonon gases.

Since \( S_2 \) vanishes if there is no coupling between the two second sounds, \( S_2 \) can be interpreted as arising from energy and momentum dissipation from one second-sound mode to the other via the viscous coupling between the modes. Thus, the spectrum \( S_2 \) due to such a process may be interpreted as the "Mountain mode," which was first introduced by Mountain in the analysis of the light-scattering spectra in molecular fluids with internal degrees of freedom weakly coupled to the density fluctuations (sound wave) via frequency-dependent viscosity. In our system, the second-sound weights if the two oscillators have the same mass and spring constant. In the "out-of-phase" mode, however, the length of the coupling spring (and/or dash pot) between the two weights changes significantly, resulting in the modification to the resonance frequency and/or to the damping rate. Without assuming Eq. (19), the two normal modes are not completely independent, but their properties are essentially similar to those mentioned above.

In the present system of "two coupled second sounds," the amplitudes of the two oscillations are the local temperatures in the LA and TA phonon gases [see Fig. 1(b)]. Therefore, if we assume the approximation as in Eq. (19), there is no dissipation from one temperature wave to the other in the "in-phase" mode (Mode 1), because there is no temperature gradient between the two temperature waves; one of the two waves does not drag the other wave. Thus, in the "in-phase" normal mode, the viscous coupling between the second sounds for the LA and TA phonon gases is inactive when we assume the approximation as in Eq. (19), and is very weak even in general cases. On the other hand, in the "out-of-phase" normal-mode (Mode 2), the local temperature changes in the two second sounds have opposite signs [see Fig. 1(b)], and hence, there is a temperature gradient between the two waves. Thus, one of the two waves strongly drags the other wave via the viscosity of the medium phonon gas.

If Eq. (19) is assumed, the spectra for the in-phase mode (Mode 1) and out-of-phase mode (Mode 2) are obtained, respectively, as

\[ S_1(q,s) = \frac{1}{s + \frac{1}{3} c_0^2 q^2 + \frac{1}{\tau_R} + \gamma_3(q,s)} \]

and

\[ S_2(q,s) = \frac{1}{s + \frac{1}{\tau} + \frac{1}{3} c_0^2 q^2 + \frac{1}{\tau} + \gamma_3(q,s)} \]

\[ S_1(q,s) \] is identical with the right-hand side of Eq. (9), which is the spectrum for an independent (noninteracting) second sound: its properties have been described in Sec. II C. \( S_2(q,s) \), on the other hand, includes the interaction between the two second-sound waves for the LA and TA phonon gases.

Since \( S_2 \) vanishes if there is no coupling between the two second sounds, \( S_2 \) can be interpreted as arising from energy and momentum dissipation from one second-sound mode to the other via the viscous coupling between the modes. Thus, the spectrum \( S_2 \) due to such a process may be interpreted as the "Mountain mode," which was first introduced by Mountain in the analysis of the light-scattering spectra in molecular fluids with internal degrees of freedom weakly coupled to the density fluctuations (sound wave) via frequency-dependent viscosity. In our system, the second-sound weights if the two oscillators have the same mass and spring constant. In the "out-of-phase" mode, however, the length of the coupling spring (and/or dash pot) between the two weights changes significantly, resulting in the modification to the resonance frequency and/or to the damping rate. Without assuming Eq. (19), the two normal modes are not completely independent, but their properties are essentially similar to those mentioned above.
modes of LA and TA phonon gases are coupled, and one mode can behave as a dissipative counterpart for the other mode; they are just like the sound wave and the internal motions of molecules in a fluid. More detailed discussions are to be given in Sec. II G 4.

E. Phonon regime and Knudsen number

For systematic analyses, we set situations that are separated according to the magnitudes of the ratio of mean free path of phonons to the characteristic length scale. Such a ratio is referred to as the “Knudsen number” in the field of hydrodynamics. Since the characteristic length scale is the reciprocal of $q$ (the magnitude of the wave-vector transfer) in scattering experiments, the Knudsen number in a phonon gas can be defined as

$$\text{Kn} = \frac{ql = q\tau}{\tau} = \frac{ql = q\tau}{\tau}. $$

where $l$ is the mean free path of phonons

The overall Knudsen number is, then, given by

$$\text{Kn} = (\text{Kn}_R^{-1} + \text{Kn}_N^{-1})^{-1} = q\tau. $$

When $\text{Kn}_N \ll 1$ or $\text{Kn}_R \ll 1$, the situation is referred to as “hydrodynamic regime” because we can assume local thermal equilibrium within a length scale of $q^{-1}$. In the hydrodynamic regime, we can observe thermal diffusion, or thermal wave (“second sound”). When $\text{Kn}_N \gg 1$ and $\text{Kn}_R \gg 1$, the situation is referred to as “collisionless” or “ballistic” regime, where we cannot utilize macroscopic descriptions because macroscopic quantities such as temperature or entropy cannot be defined locally; in this case, we do not observe macroscopic phenomena such as diffusion or propagation of “temperature” or “entropy.” Other general situations are categorized as “intermediate regime.” Figure 2 shows how the phonon regimes are defined according to combinations of the phonon Knudsen numbers $\text{Kn}_N$ and $\text{Kn}_R$.

In the hydrodynamic regime, i.e., when $\text{Kn}$ is much smaller than unity, the ET-equation set in Eq. (1) reduce to a classical thermodynamic equation such as the heat diffusion equation or the heat wave equation for second sound. Such thermodynamic equations can be obtained even if we set $\gamma_1$ to be zero, which means that we neglect the moments higher than the momentum density. Therefore, the contributions of the higher order moments are considered to be silent if $\text{Kn} \ll 1$. The higher order moments (or the viscosity term $\gamma_2$) begin to play significant roles when the nonequilibrium degree is increased such that $\text{Kn} \gg 1$.

![FIG. 2. Phonon regimes and phonon Knudsen numbers. $\text{Kn}_R$ and $\text{Kn}_N$ are defined in Eqs. (25) and (26), respectively. The hatched region corresponds to the “second-sound regime,” where a wavelike propagation of heat is expected to exist in solids.](image)

F. Asymptotic expression for the spectrum

The continued-fraction expansion is advantageous in that we can approximate the spectrum with asymptotic forms of the component fractions in Eqs. (10), viz., $\lim_{q \to 0} \gamma_2(q,s)$, assuming equal relaxation times for a mode. In that case, one has only to provide a continued fraction to a certain order ($n \sim 30$ has been turned out to be sufficient), then to terminate it with an asymptotic form obtained from the relation

$$\gamma_n(q,s) = \frac{1}{(s + 1/\tau) + \alpha_\infty q^2 \gamma_2(q,s)}, $$

which gives

$$\gamma_n(q,s) = \frac{2}{c^2q^2} \left[ - \left( s + 1/\tau \right) + \frac{\sqrt{(s + 1/\tau)^2 + c^2q^2}}{c^2q^2} \right], $$

where we note that $\alpha_\infty = \lim_{q \to 0} \alpha_\infty = 1/4$. By terminating the continued fractions in Eq. (10) with the asymptotic form at sufficiently deep level, namely, at $n_{\text{max}} = 31, 5, 14$ where $n_{\text{max}}$ is the highest order of moment, Eqs. (23) and (24) can be applied continuously from thermal equilibrium even to far nonequilibrium regime because $\gamma_2(q,s)$ so terminated includes effectively infinite orders of moments.

For $S_2(q,s)$, we can express the right-hand side of Eq. (24) in a more compact form if we infinitely expand the continued fraction in $\gamma_2(q,s)$ (see Appendix B)

$$\text{Im}[S_2(q,s)_{\text{fin}}] = \frac{1}{2cq} \left[ \ln[(\omega + c\tau)^2 \tau^2 + 1] - \text{ln}[(\omega - c\tau)^2 \tau^2 + 1] \right], $$

$$\text{Re}[S_2(q,s)_{\text{fin}}] = \frac{1}{2cq} \left[ \tan^{-1}(\omega + c\tau) \tau - \tan^{-1}(\omega - c\tau) \tau \right]. $$

The scattering spectrum is given from Eq. (29); the right-hand side is a difference of two arctangent functions. The right-hand side of Eq. (28) is a difference of two logarithmic
functions, and Eqs. (28) and (29) are related by the Kramers-Kronig relation. Since the expressions in Eqs. (28) and (29) contain the infinite hierarchy of the frequency- and wave-vector-dependent viscosity, we may use Eqs. (28) and (29) as a response function of Mode 2 for any combination of KnN and KnR.

The spectral function of the lowest order with the asymptotic expressions of Eq. (27) corresponds to that in the case that \( n_{\text{max}} = 2 \) and \( \gamma_3(q, s) = \gamma_\alpha(q, s) = 2[\sqrt{c^2 q^2 - (s + 1/\tau)^2} - (s + 1/\tau)] \). In that case, \( \gamma_3'(q, \omega) \) and \( \gamma_3''(q, \omega) \) in Eq. (12) are given, respectively, by

\[
\gamma_3'(q, \omega) = \frac{8}{15} \left( \frac{z' - 1/\tau}{\tau} \right), \quad \gamma_3''(q, \omega) = \frac{8}{15} (z'' - \omega),
\]

where \( z' \) and \( z'' \) are defined as in \( z = \sqrt{c^2 q^2 + (i\omega + 1/\tau)^2} = z' + iz'' \), with

\[
z' = \text{Re} \left[ \sqrt{c^2 q^2 + (i\omega + 1/\tau)^2} \right] = \frac{1}{\sqrt{2}} \sqrt{\left( \frac{c^2 q^2 - \omega^2 + \frac{1}{\tau^2}}{\tau^2} \right)^2 + \frac{4\omega^2}{\tau^2}} \left( \frac{c^2 q^2 - \omega^2 + \frac{1}{\tau^2}}{\tau^2} \right),
\]

\[
z'' = \text{Im} \left[ \sqrt{c^2 q^2 + (i\omega + 1/\tau)^2} \right] = \text{sign}(\omega) \times \frac{1}{\sqrt{2}} \sqrt{\left( \frac{c^2 q^2 - \omega^2 + \frac{1}{\tau^2}}{\tau^2} \right)^2 + \frac{4\omega^2}{\tau^2}} - \left( \frac{c^2 q^2 - \omega^2 + \frac{1}{\tau^2}}{\tau^2} \right).
\]

If we expand the continued fraction \( \gamma_3(q, s) \) further, i.e., if \( n_{\text{max}} = 3 \) and \( \gamma_4(q, s) = \gamma_\alpha(q, s) \), then \( \gamma_3 \) and \( \gamma_4' \) are given as follows:

\[
\gamma_3'(q, \omega) = \frac{4}{15} c^2 q^2 \times \frac{1/\tau + \frac{18}{17} z'}{\left( \frac{\omega + 18}{17} z'' \right)^2 + \left( 1/\tau + \frac{18}{17} z' \right)^2},
\]

\[
(31)
\]

\[
\gamma_4'(q, \omega) = -\frac{4}{15} c^2 q^2 \times \frac{\omega + \frac{18}{17} z''}{\left( \frac{\omega + 18}{17} z'' \right)^2 + \left( 1/\tau + \frac{18}{17} z' \right)^2}.
\]

Note that \( \gamma_3(q, \omega) \) as given above can be approximated as a Debye-type relaxational mode (a central Lorentzian) if hydrodynamic regime \( (cq\tau \ll 1) \) is assumed

\[
\gamma_3(q, \omega) = \frac{\tau}{1 + i\omega \tau} \quad (n_{\text{max}} = 3, \gamma_4 = \gamma_\alpha).
\]

### G. Hydrodynamic regime

#### 1. Second sound

If KnR \( \ll 1 \) or KnN \( \ll 1 \), one can show from Eq. (30) that, for \( n_{\text{max}} = 2 \) and \( \gamma_3 = \gamma_\alpha \).

\[
S_1(q, \omega) \rightarrow \frac{2c^2 \tau}{15} \frac{\omega^2 - \omega_0^2}{\omega^2 + 4\omega^2 \tau^2}, \quad \text{and} \quad \gamma_3''(q, \omega) \rightarrow 0,
\]

which give

\[
S_1(q, \omega) = \frac{2c^2 \tau}{15} \frac{\omega^2 - \omega_0^2}{\omega^2 + 4\omega^2 \tau^2},
\]

where we have defined

\[
\Gamma_{ss} = \frac{2c^2 \tau}{15} \frac{\omega_0^2}{\omega_0^2 + \frac{1}{\tau_R}} = \frac{2c^2 \tau}{15} \frac{\omega_0^2}{\frac{7}{5} \tau_R}.
\]

\( S_1(q, \omega) \) in Eq. (33) is the spectrum for a damped harmonic oscillator (DHO), and is essentially identical with that obtained earlier by other authors except the differences in the numerical factors in \( \Gamma_{ss} \). \( \omega_0 \) defined in Eq. (14) is the natural frequency of the phonon gas, and it corresponds to the frequency of an undamped “thermal wave.” \( \Gamma_{ss} \) defined in Eq. (34) is the damping rate of the thermal wave. Note that \( \Gamma_{ss} \) depends not only on \( \tau_R \) but also on \( \tau_N \), c, and \( q \), and it has opposite dependences on \( \tau_N \) and on \( \tau_R \), i.e., the shorter \( \tau_N / \tau_R \) gives the smaller (larger) \( \Gamma_{ss} \).

If \( \Gamma_{ss} < \omega_0 \) holds, the thermal wave is underdamped and a resonance occurs at a frequency

\[
\omega_{ss} = \sqrt{\omega_0^2 - \Gamma_{ss}^2} = \omega_0 \left( 1 - \frac{2\tau_N}{5\tau_R} \right),
\]

which is the frequency of “second sound,” and this resonance effect is referred to as the “second-sound resonance.” Rather than the familiar diffusive transport of heat energy, the second sound is a wavelike propagation of heat at a finite propa-
gation speed \( v_{ss} = c/\sqrt{3} \). In order to have such an *equilibrated* thermal wave as a well-defined collective excitation of phonons, we need, not only the weak-damping condition \( \Gamma_{ss} \ll \omega_0 \), but also the hydrodynamic regime, i.e., \( \text{Kn}_N \ll 1 \) or \( \text{Kn}_R \ll 1 \) because there must be sufficient number of thermal phonons within the relevant scales of space and time. The simultaneous requirements can be expressed in an inequality as
\[
\Gamma_{ss} \ll \omega_0 \ll \tau_N^{-1}.
\]
From Eq. (34), this can be alternatively expressed in a well-known form as
\[
\tau_R^{-1} \ll \omega_0 \ll \tau_N^{-1},
\]
which is called the “window condition” for propagation of second sound in solids. The window condition can be expressed also as
\[
\text{Kn}_N \ll 1 \ll \text{Kn}_R,
\]
which is clearly stating that one simultaneously requires frequent \( N \) processes and infrequent \( R \) processes within a length scale of \( q^{-1} \). In Fig. 2, the region labeled as “pure second sound” corresponds to the condition of Eq. (37). In this regime, “pure” second sound is expected, and its spectrum should be a very sharp doublet located at \( \omega_{ss} = \pm cq/\sqrt{3} \).

2. Thermal diffusion

If \( \Gamma_{ss} > \omega_0 \), then the second sound is overdamped and cannot propagate. This is the thermal diffusion effect, which is a much more common phenomenon in thermal transport in most materials. In this case, \( S_1(q, \omega) \) is reduced approximately to a central peak
\[
S_1(q, \omega) \approx \frac{\Gamma_{th}}{\omega^2 + \Gamma_{th}^2},
\]
which is the well-known “thermal Rayleigh component” with a width proportional to \( q^2 \) as
\[
\Gamma_{th} = D_{th} q^2.
\]
\( D_{th} \) is the thermal diffusivity in a well-known kinetic expression as
\[
D_{th} = \frac{1}{3} c^2 \tau_R.
\]
The width can be alternatively expressed as
\[
\Gamma_{th} = \omega_0^2 \tau_R.
\]

3. Properties of \( S_2(q, \omega) \) and total spectrum

Setting \( cq \tau \ll 1 \) in the right-hand side of Eq. (29), we see that
\[
S_2(q, \omega) \approx \frac{1}{2cq} \times 2cq \tau \frac{\partial}{\partial(\omega \tau)} \tan^{-1} \frac{\omega \tau}{\omega^2 + \Gamma_{th}^2},
\]
where
\[
\Gamma_2 = \frac{1}{\tau} = \frac{1}{\tau_N} + \frac{1}{\tau_R}.
\]
Thus, the linewidth of \( S_2(q, \omega) \) corresponds to the mean frequency of the collisions between the phonons. Since \( \tau \) is an averaged relaxation time over the whole thermal phonons in the Brillouin zone, \( \tau \) is considered to have no dependence on frequency and wave vector of thermal phonons. Thus, \( \tau \) should be independent of \( q \). The width \( \Gamma_2 \) is much broader than \( \omega_0 \) and than \( \Gamma_{th} \) because we are assuming the hydrodynamic regime (\( cq \tau_N \ll 1 \) or \( cq \tau_R \ll 1 \)). Thus, the thermal-DHO spectrum, \( S_1(q, \omega) \), should be superposed on this much broader unshifted Lorentzian of \( S_2(q, \omega) \) as
\[
S_{total}(q, \omega) = P_1 \frac{2\omega_0^2 \Gamma_{ss}}{(\omega^2 - \omega_0^2)^2 + 4\omega^2 \Gamma_{ss}^2} + P_2 \frac{\Gamma_2}{\omega^2 + \Gamma_2^2},
\]
where \( P_1 \) and \( P_2 \) are the weighting coefficients for \( S_1(q, \omega) \) and \( S_2(q, \omega) \), respectively.

In thermal diffusion regime, the resistive process is dominant, and \( \Gamma_2 \) is effectively equal to \( 1/\tau_R \). Thus, we have a useful relation between \( \Gamma_{th} \) and \( \Gamma_2 \) as
\[
\Gamma_{th} \approx \frac{1}{3} \gamma_1 q^2 = \frac{\omega_0^2}{cq},
\]
i.e., the product of the linewidths of thermal Rayleigh and Mountain modes is almost temperature independent at high temperatures. Equation (43) also states that
\[
\sqrt{\gamma_1 \gamma_2} = \frac{1}{\sqrt{3}},
\]
where \( \gamma_1 = \Gamma_{th}/cq \) and \( \gamma_2 = \Gamma_2/cq \) are the linewidths normalized by the average Brillouin shift.

4. Interpretation of the spectral components in the hydrodynamic regime

It is important to note that the thermal relaxation (thermal diffusion) and the fast relaxation (collisions between phonons) should never be discussed separately. Rather, we see that the two spectra, \( S_1(q, \omega) \) and \( S_2(q, \omega) \), reflect two limiting aspects in a medium: the slow relaxation corresponds to the fluctuations just in the vicinity of local thermal equilibrium, whereas the fast one to the kinetic processes, i.e., the collisions, which are the nonequilibrium processes essential for thermal equilibrium to be locally established. Therefore, we see the “duality” of a phonon gas; we see both equilibrium and nonequilibrium processes simultaneously in Eq. (42). It should also be emphasized that such a spectral structure of the dual fluctuation can be observed only if the characteristic length (or time) scale in the scattering experiment lies between the two length scales for thermal diffusion and interparticle collision, i.e., a relation,
\[
l \ll q^{-1} \ll c/(D_{th} q^2)
\]
is required for the two spectra to be clearly resolved. Therefore, the selection of a length scale \( q^{-1} \) is critical in studying
the nonequilibrium phonon dynamics in crystals, and we see that light-scattering experiments can offer a good value since 10 ≤ q −1 ≤ 100 nm. Note that the mean free path of phonons can range from ~1 to ~1000 nm according to the temperature, indicating that temperature change can break the inequality [Eq. (45)]. Since an average Brillouin frequency \( \bar{\omega}_B = q c \) can be regarded as a characteristic frequency in light scattering in a phonon gas,7 Eq. (45) can be written in the frequency domain as

\[
D_0 q^2 \ll \bar{\omega}_B \ll 1^-1,
\]

which states that the Brillouin line should appear in the middle frequency range between the two central peaks of \( S_1 \) and \( S_2 \).

Similar discussions should be able to apply to any scattering experiments for any gas model. In such general cases, \( D_0 \) should be replaced with a generalized diffusivity for constituent particles or excitations. In fact, superionic conductors exhibited similar double quasielastic spectrum in light-33 and neutron-34 scattering experiments and the narrow and broad linewidths were tentatively attributed to the ion diffusion rate and the mean hopping rate of the conducting ions, respectively. This is very similar in nature to the present case as was hinted long ago.35

**H. Ballistic regime (collisionless regime)**

If both \( N \) and \( R \) processes become infrequent such that \( \text{Kn}_N, \text{Kn}_R \gg 1 \), the approximate expression of Eq. (42) in the hydrodynamic regime is not valid, and effects of higher order moments set in. If we retain the approximation defined in Eq. (19), \( S_1 \) and \( S_2 \) are given, respectively, from Eqs. (13) and (29) as

\[
S_1(q, \omega) = \frac{\omega_0^2}{\tau_R} \left( \frac{1}{\tau_R} + \gamma_1 \right)
\]

\[
(\omega_0^2 - \omega^2 - \omega \gamma_1^2) + \omega^2 \left( \frac{1}{\tau_R} + \gamma_1 \right)^2,
\]

\[
S_2(q, \omega) = \frac{1}{2c} \left[ \tan^{-1}(\omega + cq) \tau - \tan^{-1}(\omega - cq) \tau \right],
\]

where \( \gamma_1^2(q, \omega) \) and \( \gamma_2^2(q, \omega) \) are the real and imaginary parts of \( \gamma_1^2(q, \omega) \) defined in Eq. (10), respectively. As the ballistic regime is approached, the widths of \( S_1 \) and \( S_2 \) approach each other as shown in Fig. 3, where the Normal process of phonon collision is ignored by setting \( \tau_N \gg \tau_R \). For small values of \( \tau_R \), the spectra of \( S_1 \) and \( S_2 \) correspond to the thermal-diffusion mode and the Mountain mode, respectively, and both spectra are well approximated as Lorentzians, as we have described in Sec. II G. As \( \tau_R \) becomes longer, the narrow quasielastic component, \( S_1 \), broadens whereas the broad component \( S_2 \) narrows. However, the two widths of \( S_1 \) and \( S_2 \) converges to \( cq \), which is the averaged Brillouin frequency \( \bar{\omega}_B \). In fact, in the extreme limit of the ballistic regime, we can set \( \tau \rightarrow \infty \), and we see, from Eqs. (23) and (24) that \( S_1(q, \omega) \) and \( S_2(q, \omega) \) coincide with each other

\[
S_1(q, \omega) = S_2(q, \omega) = \frac{\omega_0^2 \gamma_1^2(q, \omega)}{[\omega_0^2 - \omega^2 - \gamma_1^2(q, \omega) \omega^2 + \omega^2 \gamma_1^2(q, \omega) \omega^2].}
\]

The convergence of \( S_1 \) and \( S_2 \) is shown in Fig. 3 for \( \text{Kn}_R = 100 \) although the line shape of \( S_1 \) is slightly distorted due to a crude approximation, in which we set \( \gamma_1 \rightarrow \gamma_2 \) in tracing the spectra of \( S_1 \).

The rectangular line shape in the ballistic limit can be checked qualitatively as the following. If we replace the continued fraction \( \gamma_3^2(q, s) \) with \( \gamma_\infty(q, s) \) in Eq. (30), we get

\[
\gamma_3^2(q, \omega) \rightarrow \frac{8}{15} cq \sqrt{q^2 - \omega^2}, \quad \gamma_3^2(q, \omega) \rightarrow -\frac{8}{15} \omega.
\]

Substituting the above expressions of \( \gamma_3^2(q, \omega) \) and \( \gamma_5^2(q, \omega) \) into the right-hand side of Eq. (46) yields a spectrum of the form

\[
S_1(q, \omega) = S_2(q, \omega) = \frac{24q^2 q^2 - \omega^2}{-\omega^4 - 6 \omega^2 + 15q^2} \quad (|\omega| \leq cq).
\]

The numerator and denominator of the right-hand side of Eq. (47) are an ellipse and a convex quartic curve, respectively. Since the latter is close to an ellipse in shape, the spectral shape is almost flat for \( -cq \leq \omega \leq cq \), and it has vertical walls at \( \omega = \pm cq \).

If we do not employ the asymptotic form \( \gamma_\infty(q, s) \) and cut the continued fraction at \( n=n_\text{max} \), i.e., if we set \( \gamma_\text{max}^2+1=0 \), the spectrum in the highly ballistic regime exhibits \( n_\text{max} \) sharp peaks5,36 as we show in Fig. 4, where \( n_\text{max} \) and \( \text{Kn}_N \) are varied (\( \text{Kn}_N \rightarrow \infty \) is assumed for simplicity). In general, it can be shown that these peaks are located at \( \omega(i) = cq \cos \phi_\text{max}(i) \), and their heights are \( h(i) \propto \sin \phi_\text{max}(i) \), where \( \phi_\text{max}(i) = i \pi / (n_\text{max} + 1) \), and \( i \) is an integer (for detailed discussions, see Appendix B). The spectral formula in the ballistic limit is, then, given in the form

\[
S_{\text{ball}}(\omega, q) \propto \sum_{i=1}^{n_\text{max}} \sin \phi_\text{max}(i) \left[ \frac{1}{\omega - cq \cos \phi_\text{max}(i)} \right]^2 + \frac{1}{\tau^2},
\]

where Lorentzian line shape is introduced, and a common linewidth, \( 1/\tau \), has been introduced for all the peaks. The behavior of the spectra in the ballistic limit, i.e., \( \text{Kn}_R = 1000 \) shown in Fig. 4 can be fully reproduced by Eq. (48).

The comblike peak distribution seen in Fig. 4 is apparently due to the lack of number of moments. In fact, in Fig. 4, the spectrum for \( n_\text{max} = 30 \) and \( \text{Kn}_R = 100 \) is comblike whereas that for \( n_\text{max} = 300 \) and \( \text{Kn}_N = 100 \) is rectangular in shape. To include infinite number of higher order moments, we only have to replace the summation in Eq. (48) with an integral, and we obtain the spectrum with infinite number of moments (\( n_\text{max} \rightarrow \infty \)).
The right-hand side can be analytically integrated to give

$$S_{\text{bal}}(\omega, q) \propto \frac{1}{q} \frac{1}{\omega - cq} \cos \phi + 1 + \frac{1}{q} \frac{1}{\omega + cq} \cos \phi.$$

(49)

The right-hand side can be analytically integrated to give

$$S_{\text{bal}}(\omega, q) \propto \frac{1}{q} \frac{1}{\omega - cq} \cos \phi + 1 + \frac{1}{q} \frac{1}{\omega + cq} \cos \phi.$$

(50)

which is identical with the right-hand side of Eq. (29). Setting $\tau \to \infty$ in Eq. (50), we actually obtain a rectangular function

$$\lim_{\tau \to \infty} S_{\text{bal}}(q, \omega) = \begin{cases} 1 & (|\omega| < cq) \\ \frac{1}{2} & (|\omega| = cq) \\ 0 & (|\omega| > cq) \end{cases} \quad \text{rect} \left( \frac{\omega}{2cq} \right).$$

The light scattering in the ballistic regime can be viewed also as arising from elastic scattering from individual phonons traveling in all directions. Similar effect is observed in rarefied gasses, in which the individual molecules elastically scatter the incident radiation as “moving mirrors” with the velocity components following the Maxwell-Boltzmann velocity distribution, giving rise to a Doppler-broadened elastic spectrum, i.e., a central Gaussian spectrum.$^5$ In general, the spectrum obtained from individual scatterers in the low-density limit is given by$^5$

$$S(q, \omega) \propto \text{Re} \left[ \int \frac{\tilde{f}(\epsilon)}{s - i\epsilon \cdot q + 1/\tau} \frac{d\epsilon}{2\pi} \right]_{s=\omega},$$

where $\tilde{f}(\epsilon)$ is the velocity distribution for the scatterers; for ideal (massive) gasses $\tilde{f}(\epsilon) \approx e^{-\epsilon^2/2k_BT}$ (Maxwell-Boltzmann
The isotropic phonon gas is given as

\[ \omega = \omega_{\text{scat}} - \omega_{\text{inc}} = \omega_i(k_2) - \omega_i(k_1), \]

and

\[ q = k_{\text{scat}} - k_{\text{inc}} = k_2 - k_1, \]  

(51)

where \( \omega_i(k) \) is the frequency of a phonon in mode \( i \), and \( k_1 \) and \( k_2 \) are the wave vectors of the relevant phonons. Note that \( q \approx k_1, k_2 \) and that \( q \approx \omega_1, \omega_2 \) in light-scattering experiments. The TPDS from different phonon modes, i.e., the case in which \( i \neq j \), requires that two different phonon modes be degenerated at points in the Brillouin zone. Since such points are usually located at the zone boundaries, it is not expected that TPDS from different phonon modes is dominant at low temperatures. Hence, in the ballistic regime, where there are few phonons excited, the phonons participating in the TPDS should belong to the same branch, viz., \( i = j \). Thus, the TPDS should be “intramode” in the ballistic regime.43

Assuming an isotropic phonon dispersion, and letting the angle between \( q \) and \( k_2 \) be \( \varphi \), a single TPDS yields44 a frequency shift of \( \omega(\varphi) = cq \cos \varphi \). If the two phonons have lifetimes, the spectrum from a single-scattering process may be regarded as a Lorentzian with a “pair width”44 of \( 1/\tau \).38 Thus, summing up all the contributions from the phonon pairs in the Brillouin zone, we have45

\[ S_{\text{TPDS}}(\omega, q) = \int_0^\pi \sin \varphi \frac{1/\tau^2}{(\omega - cq \cos \varphi)^2 + 1/\tau^2} d\varphi = \tan^{-1}(\omega + cq)\tau - \tan^{-1}(\omega - cq)\tau. \]

(52)

This is identical with the spectrum in the ballistic limit presented in Eqs. (49) and (50) if we identify \( \varphi \) with \( \phi \). Therefore, the comblike spectrum in the ballistic limit in Fig. 4 may be viewed also as a collection of scattering spectra due to two-phonon difference processes. The form of TPDS as in Eq. (52) was also reported in Ref. 39 with more rigorous discussions.

Although we assumed the ballistic regime so far, let us investigate the spectrum of Eq. (52) in the hydrodynamic regime. As we have shown in Eq. (41), the differential arctangent spectrum as in the right-hand side of Eq. (52) can be approximated as a Lorentzian with a width of \( \Gamma_{2 \pi} = \tau^{-1} \). Since two-phonon approximation is not possible in the hydrodynamic regime in a strict sense, we must consider that higher order interactions between the participating phonon pairs are effectively renormalized in the pair width. Such renormalization may be characterized as a ladder-type diagram for the self-energy in the Green function technique,18,19,32,46 in which “first sound,” “second sound (including thermal diffusion),” and “nonthermodynamic dielectric fluctuations” have been introduced. Among such contributions in the self-energy, the present TPDS corresponds to the “nonequilib-
where a normalizing factor has been introduced such that the sum coincides with $\sum$. Here, we still retain the approximation as in Eq. (19) for simplicity. Even without the approximation, the properties of these spectra.

Summarizing this section, we have seen that $S_{\text{TPDS}}$ is always coincides with $S_1$ regardless of the phonon regime. Therefore, $S_2$ may be regarded as arising from light scattering from two-phonon difference processes on the same phonon dispersion, and we may always write $S_2(q, \omega)$ as

$$S_2(q, \omega) = \frac{1}{2 \pi c q} \left[ \tan^{-1}(\omega + c q) - \tan^{-1}(\omega - c q) \right],$$

where a normalizing factor has been introduced such that the integrated intensity is equal to 1. $S_2(q, \omega)$ in this “differential arctangent” form can cover spectrum shapes continuously from the broad central Lorentzian in the hydrodynamic regime to the rectangular spectrum in the ballistic regime.

J. Summary of spectral variation

To summarize theory section, we present, in Fig. 5, a chart of the light-scattering spectra calculated for a phonon gas, covering possible major combinations of $(K_{\text{NR}}, K_{\text{NN}})$. Here, we still retain the approximation as in Eq. (19) for simplicity. Even without the approximation, the properties of the calculated spectra are essentially the same except that each linewidth of $S_1$ and $S_2$ slightly differs from that in the approximated case. We have set $n_{\text{max}}=5$ and $\gamma_q=\gamma_s$ in calculating these spectra.

FIG. 5. (Color online) A chart of light-scattering spectra from a phonon gas for combinations of $(K_{\text{NR}}, K_{\text{NN}})$. All the spectra are plotted on log-log scales. We set $n_{\text{max}}=5$ and $\gamma_q=\gamma_s$ in calculating these spectra.

of $S_2$ spectra, there are either narrower quasielastic component due to thermal diffusion or inelastic peaks due to second-sound resonance; both of these are $S_1$’s. The coexistence of $S_1$ and $S_2$ indicates the duality of a relaxation processes in a phonon gas. The origin of $S_2$ can be interpreted as the followings: (1) the nonequilibrium counterpart of fluctuations (relaxation processes) in a phonon gas, (2) the Mountain mode due to frequency-dependent viscosity in a crystal, or (3) two-phonon difference light scattering on the same phonon dispersion, each of which statements is physically equivalent to one another.

The left column of the Fig. 5 corresponds to the diffusion limit, for which $K_{\text{NR}} \ll 1$, i.e., the $R$ process is dominant, and the spectrum consists of two unshifted Lorentzians. The linewidths for the narrow and broad quasielastic components are

$$\Gamma_1 = \frac{1}{c^2 q \tau_{\text{R}}}, \quad \Gamma_2 = \tau^{-1} + \tau_{\text{NR}}^{-1} \tau_{\text{NN}},$$

respectively.

The lower right regions in Fig. 5, i.e., $(K_{\text{NR}}, K_{\text{NN}}) = (100, 0.1)$, corresponds to the “pure” second-sound regime because of Eq. (37), and a very sharp peak due to second-sound resonance is found. However, it should be pointed out that the resonance peaks due to second sound are found in a wider range in Fig. 5, viz., $K_{\text{NR}} \ll 1 \ll K_{\text{NN}}$. This means that the window condition for the observation of the second-sound peak in light-scattering experiments may be rather tolerant.

The upper right region in Fig. 5, i.e., the region $K_{\text{NR}}, K_{\text{NN}} \gg 1$, corresponds to the ballistic limit. As the ballistic limit is approached, $S_1$ and $S_2$ approach each other, and in the limit of $\tau \to \infty$, both spectra converge into a rectangular shape with a cut-off frequency at $\omega = cq = \bar{c} q$. For a finite value of $\tau$, the rectangular spectrum has a weak Lorentzian tail. It is interesting to point out that a broad, shifted-peak structure still exists for $(K_{\text{NR}}, K_{\text{NN}}) = (100, 10)$ as we show in Fig. 6. This broad shifted peak is not due to thermalized second sound, but due to an unequilibrated modulation of number density of phonons. The scattering mechanism in this ballistic regime is better stated as TPDS rather than light scattering by collective excitation of phonons such as thermal diffusion or second sound. TPDS can give rise to a shifted-peak structure as that shown in Fig. 6 also when the group velocity of phonons has directional dependence, i.e., when there is elastic anisotropy, as reported by Farhi et al. 38,47 for the low-temperature light-scattering spectrum observed in KTaO$_3$. We note that the directional dependence of

FIG. 6. Shifted-peak structure in the collisionless regime $(K_{\text{NR}}, K_{\text{NN}}) = (100, 10)$. Both axes are on linear scales.
the pair width can also affect the line shape in the ballistic regime; in fact, the shifted-peak structure that is shown in Fig. 6 appears due to the frequency dependence of the linewidths of the “comb teeth” (those shown in Fig. 4) although an isotropic phonon dispersion is assumed.

III. COMPARISON WITH EXPERIMENTAL RESULTS

Quasielastic light-scattering spectra due to phonon-gas fluctuations have been reported in many crystals. In this section, we analyze the experimental spectra with the formula given in Sec. II F. In most of the following part, we use the approximated spectral function as follows:

\[ S(q, \omega) = P_1 S_1(q, \omega) + P_2 S_2(q, \omega), \]

with

\[ S_1(q, \omega) = \frac{2c q}{\pi} \frac{\omega_0^2 [\gamma'_1(q, \omega) + 1/\tau_R]}{[\omega_0^2 - \omega^2 - \omega^2 \gamma'_1(q, \omega)]^2 + \omega^2 [\gamma'_1(q, \omega) + 1/\tau_R]^2}, \]

\[ S_2(q, \omega) = \frac{1}{\pi} [\tan^{-1}(\omega + cq)\tau - \tan^{-1}(\omega - cq)\tau], \]

where \( \gamma'_1(q, \omega) \) and \( \gamma'_2(q, \omega) \) are defined in Eqs. (31) and (32), respectively. The approximation defined in Eq. (19) has been also employed. The integrated intensities of \( S_1 \) and \( S_2 \) are given by \( 2c q P_1 \) and \( 2c q P_2 \), respectively, so that its ratio is given by \( P_1/P_2 \). Although \( P_1/P_2 \) is expected to be constant for a simple phonon-gas model, it will turn out to be strongly temperature dependent in practical systems. Equation (54) should be able to reproduce spectra in any phonon regime from hydrodynamic to ballistic regimes.

In the observed spectra, which will be shown later in this section, sharp Brillouin peaks due to the first-order light scattering from LA and TA acoustic phonons are present. However, it should be noted that, as we mentioned in Sec. II B, the spectral expressions derived from the ET equations essentially describes the second-order light scattering, for which the second terms on the right-hand sides of Eqs. (6) and (7) are responsible. Therefore, our spectral formula is not expected to contain those first-order contributions (Brillouin scattering) which arise from the first terms on the right-hand sides of Eqs. (6) and (7).

We have recently reported the \( K_{\text{KnR}} \) dependence of the normalized linewidths of the two quasielastic components measured in several crystals as reproduced in Fig. 7(a). We can now simulate the \( K_{\text{KnR}} \) dependence of the normalized widths of \( S_1 \) and \( S_2 \), viz., \( K_{1/cq} \) and \( K_{2/cq} \). The result is shown in Fig. 7(b): good agreement between the (a) observation and (b) theory has been found. For sufficiently small values of \( K_{\text{KnR}} \), \( K_{1/cq} \) and \( K_{2/cq} \) are proportional to \( K_{\text{KnR}} \) and \( K_{\text{KnR}}^2 \), respectively, indicating that \( K_1 \) is proportional to \( q^2 \) and that \( K_2 \) is independent of \( q \) in the hydrodynamic regime. Here note that \( K_1 \) is exactly equal to \( K_{\text{KnR}} \), the half width of the diffusive central peak, in the hydrodynamic regime. For small \( K_{\text{KnR}} \), we also see in Figs. 7(a) and 7(b) that Eq. (43) and (44) are reasonably satisfied. For sufficiently large values of \( K_{\text{KnR}} \), both \( K_{1/cq} \) and \( K_{2/cq} \) are independent of \( K_{\text{KnR}} \), indicating that both \( K_1 \) and \( K_2 \) are proportional to \( q \) in the ballistic limit.

In the following sections, we present fits of the obtained theoretical formula to the spectra observed in crystals of rutile (\( TiO_2 \)) (Refs. 7 and 42) and strontium titanate (\( SrTiO_3 \)).

A. Rutile (\( TiO_2 \))

In rutile, we observed temperature dependences of the quasielastic light scattering (QELS) consisting of two components. Figure 8 shows the temperature dependence of the spectrum observed in rutile on doubly logarithmic scales.

At high temperatures, the double-Lorentzian structure as that simulated and shown in Fig. 5 is clearly shown. The nar-
The quasielastic component broadens and the broader one narrows with increasing temperature. The inset shows the spectra at high temperatures (q||[001]). The solid lines are the fit of Eq. (54).

At around 77 K, the linewidths of the two quasielastic components are so close as it appears that there is only one quasielastic component. Below ~50 K, the temperature dependence of the linewidth becomes weaker. Note that the constant background appearing in the low-temperature spectra is largely due to the dark counts of the photomultiplier tube employed in the experiment. In rutile, the quasielastic component does not develop into an inelastic doublet as that observed in SrTiO3 (Refs. 8 and 50) as we will describe in the Sec. III B.

In fitting the spectra in rutile, we adjusted $P_1$, $P_2$, $c$, $\tau_\infty$, and $\gamma_\infty$ as adjustable parameters. For such high-temperature data, we did not determine $\gamma_\infty$ because the fit was not sensitive to $\gamma_\infty$. It is seen that $c$ ranges from 1500 to 3200 m/s, which is slower than the Debye velocity $v_D=6000$ m/s in rutile. This small average velocity seems to be due to the use of the common $c$ and $\gamma$ for all phonon modes, i.e., for LA and TA modes. In fact, the spectra can also be well fitted if we distinguish $c_{(i)\nu}$ and $c_{(f)\nu}$, where $c_{(i)\nu}$ and $c_{(f)\nu}$ were measured directly from the Brillouin frequency in the scattered spectra. The integrated intensities of $S_L$ and $S_T$ are also listed in Table I. The results are similar to those reported previously.42

Note that the intensity ratio $P_1/P_2$ is very small at high temperatures, and it increases on cooling, indicating that the viscosity in the phonon gas (the contributions to $P_2$) is larger at high temperatures and it decreases with decreasing temperature.

We estimated the thermal diffusivity from the relation, $D_\theta=c^2\gamma_\infty\gamma_\infty^{\text{vis}}$, and compared it with the values calculated from

<table>
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<th>$T$ (K)</th>
<th>$\tau_\infty$ (s)</th>
<th>$\gamma_\infty$ (s)</th>
<th>$c$ (m/s)</th>
<th>$P_1$</th>
<th>$P_2$</th>
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the known values of thermal conductivity \( \kappa \) and specific heat \( C_p \) with the relation \( D_{th} = \kappa / pC_p \), where \( p \) is the mass density. The values are listed in Table II. Good agreement was found at relatively high temperatures. Although the agreement is not so good at relatively low temperatures due to the small \( c \), the deviations were approximately one order, which seems to be reasonable for the present simplification of the model.

### B. SrTiO\(_3\)

SrTiO\(_3\) is well known as a kind of perovskite-type crystals. Many of other perovskite-type crystals such as BaTiO\(_3\) is known to undergo displacive-type ferroelectric phase transition when they are cooled down to their Curie temperature because of freezing of an optical phonon mode ("soft mode") that corresponds to the polarizable displacement of the ions.\(^{53}\) Although SrTiO\(_3\) does have such a soft optical phonon mode, the relatively large quantum fluctuation is thought to disturb the freezing of the soft mode in SrTiO\(_3\),\(^ {54}\) forcing it to stay in paraelectric phase, and hence, SrTiO\(_3\) is referred to as a "quantum paraelectric."\(^ {55}\) In SrTiO\(_3\), an anomalous Brillouin doublet was reported by Hehlen et al.,\(^{8}\) and they proposed that the origin of the new Brillouin doublet was the second sound according to the theoretical prediction by Gurevich and Tagantsev.\(^{9}\) In addition, the molecular dynamics studies published earlier by Schneider and Stoll\(^ {11,12}\) had clearly indicated the existence of second sound in a model of displacive-type ferroelectric with an optical phonon mode that softens in the Brillouin-zone center. We have observed that the thermal-Rayleigh central peak changed into the new Brillouin spectrum on cooling,\(^ {7}\) and pointed out the possibility of the existence of the second sound in SrTiO\(_3\), supporting the proposal made by Hehlen et al. More recently, we have reported an ET analysis of the spectra in SrTiO\(_3\),\(^ {50,56}\) and we have shown that the overdamped thermal wave (thermal diffusion) is actually underdamped in a narrow temperature range around 30 K. Figure 9 shows the temperature variation in the low-frequency light-scattering spectrum in SrTiO\(_3\). Although we have employed a more rigorous, but complicated form for the spectral expression [Eq. (3) in Ref. 50], we have confirmed that fitting Eq. (54) for all the investigated temperatures give almost the same results as the analysis we reported in Ref. 50.

Figure 10 shows the temperature dependences of the obtained \( \tau_N \), \( \omega_{ss} \), and \( \Gamma_{ss} \).\(^ {50}\) The expressions for \( \omega_{ss} \) and \( \Gamma_{ss} \) in

![FIG. 9. (Color online) Temperature dependence of the low-frequency light-scattering spectra in SrTiO\(_3\) from 6 to 295 K (log-log plot). The LA and TA denotes longitudinal and transverse acoustic phonons’ Brillouin lines, respectively. The inset is a semilog plot for the spectra below 72 K. Each spectrum is appropriately scaled for visual clarity. The solid lines are fits of Eq. (54).](https://example.com/fig9)

![FIG. 10. (Color online) Temperature dependences of the normal \( (\tau_N) \) and the resistive \( (\Gamma_{ss}) \) phonon relaxation rates in SrTiO\(_3\). Legends are shown in the figure. All the lines between the symbols are guides to the eyes. The dashed horizontal line at \( \sim 2 \times 10^{15} \) rad/s represents a constant \( q\mu_b/\sqrt{3} = \omega_0 \). The shaded area indicates the "window" for second-sound propagation.](https://example.com/fig10)
terms of $\tau_N$, $\tau_R$ are defined in Eqs. (34) and (36), respectively. The values of $\tau_R$ were measured in a separate work\textsuperscript{7} except for the value at 6.4 K, which could not be determined from the employed experimental condition in the work; $\tau_R$ for 6.4 K was estimated from literature values of $\kappa$ and $C_p$ with Eq. (40). The shaded area in Fig. 10 is the “open window” for second-sound propagation, where $\tau_R < \tau_N^{-1}$. If the second-sound frequency, $\omega_{ss}$, enters into that area, then underdamped second sound is expected to propagate. Indeed, the obtained $\omega_{ss}$ lies in the shaded area in the temperature range approximately between 20 and 30 K as can be seen in Fig. 10.

At higher temperatures than $\sim 30$ K, the second sound is overdamped because $\omega_{ss} < \tau_R^{-1}$, resulting in the quasielastic scattering observed for $T \approx 40$ K. At much higher temperatures, the damping is strong enough to regard the overdamped thermal wave as “diffusion of heat,” and the spectrum consists of a narrow thermal Rayleigh mode and the broad Mountain mode. Indeed, as can be seen in Fig. 10, the estimated $D_R$ and fitted $\tau_R$ well reproduce the reported values of $D_R$ and $\tau_R$, respectively. This double QELS structure is equivalent to what was observed in rutile at high temperatures, as shown in the Sec. III A. At lower temperatures than $\sim 15$ K, the second sound cannot be defined any more because $\tau_N^{-1} < \omega_{ss}$ i.e., there are too few phonon collisions during a period of the second-sound oscillation. Even in such a case, however, light-scattering spectrum can exhibit inelastic peaks as we have shown in Fig. 5. Therefore, we should consider that scattering mechanism for the inelastic peaks observed at 6.4 K in SrTiO$_3$ is TPDS on the same phonon branch\textsuperscript{38} rather than second sound.

We can review how the phonon regime changed in SrTiO$_3$ according to the temperature by plotting the phonon Knudsen numbers in the $(\kappa R, \kappa N)$ plane. Figure 11 shows the temperature dependence of the point $(\kappa R, \kappa N)$). It is seen that SrTiO$_3$ under the investigated condition ($q=6.0 \times 10^7$ 1/m) actually goes out of the thermal-diffusion regime at around 50 K, then, enters into the second-sound regime around 30 K, and finally goes out to the ballistic regime at the lowest temperature.

IV. DISCUSSIONS

In the previous section, we confirmed that the two-component QELS (double QELS) arises from the overdamped thermal waves (the narrow QELS) and their interaction phenomenon (the broad component). In SrTiO$_3$, the thermal wave was found to be underdamped below 40 K, and the narrower QELS changed into a broad-doublet spectrum. Our analysis showed that $\omega_{ss}$ was in the narrow “second-sound window,” and the broad doublet in SrTiO$_3$ could be attributed to the second sound in the hydrodynamic regime.

The physical origin of the broad doublet in SrTiO$_3$ has been a long standing controversy.\textsuperscript{5,55-59} Hehlen et al. were the first to report the extra Brillouin doublet in SrTiO$_3$, and they tentatively proposed that the new doublet was due to the second sound according to the theory of Gurevich and Tagantsev.\textsuperscript{9} However, Scott et al.\textsuperscript{39} have published a number of negative reports and they argued that the new doublet should be arising from the light scattering from two-phonon difference processes on two optical phonon branches that accidentally degenerate at $\sim 37$ K. Scott et al. claimed that the light-scattering intensity for the second sound (entropy fluctuations) should be much weaker than predicted although they did not show a quantitative discussion on this aspect. However, our observation and analysis have revealed that the previously reported\textsuperscript{7,40} narrow QELS due to thermal diffusion (entropy fluctuations) survives even at a temperature as low as 50 K. Furthermore, at even lower temperatures, the narrow QELS gradually changes its shape into a shifted doublet with smooth changes in the line-shape-determining parameters, namely, $\tau_N$, $\tau_R$, $\omega_{ss}$, as shown in Fig. 10.

We also emphasize that the simultaneous existence of the broader QELS and the broad doublet is also a sign of the second sound. Our simulation for the spectrum (Fig. 5) indicates that the second-sound doublet should be superposed on the broader QELS (Mountain mode), which was actually observed in the experiments.\textsuperscript{7,8,50,60} The fact that the fitted $\tau_N$, $c$, and the known $\tau_R$ reproduced the entire spectral shape implies the high reliability of our analysis. In fact, the values obtained for $c$ ranged from 3600 m/s at 6.4 K to 5900 m/s at 50 K, which are quite reasonable compared with $v_D = 5400$ m/s, where $v_D$ is the average sound velocity in the Debye’s theory. The value of $\tau_N$ can be roughly evaluated from the Gurevich-Tagantsev relation,\textsuperscript{9}

\[
\tau_U \sim \frac{1}{\tau_N},
\]

where $\tau_U$ and $\epsilon$ are the umklapp-scattering time and the dielectric constant, respectively. Replacing $\tau_U$ with $\tau_R$, which is smaller than $\tau_N$ in general, and substituting the known value for $\epsilon [\sim 10^{-14}$ for $T < 20$ K (Ref. 61)], $1/\tau_N$ is expected to be on the order of $10^{12}$ rad/s or less for $T < 20$ K; this does not contradict our result.

Tsujiimi and Itoh\textsuperscript{62} have published another interpretation for the broad doublet. Their hypothesis is that the broad doublet is arising from the partially softened TA phonon in the small polar regions, which are thought to exist in SrTiO$_3$.\textsuperscript{63,64} They have shown that significant doping of cations killed the broad doublet in SrTiO$_3$, and they argued that the screening by the doped cations disturbed the development of small polar regions and that it in turn killed the softened TA phonons, which could otherwise exist locally in the small polar region. Since such polar regions contain softened TO
phonons of low energy, the normal phonon collisions that are necessary for the second-sound excitation should also be enhanced in such local regions because normal phonons should have low energy. Therefore, the reported disappearance of the broad doublet in doped samples of SrTiO$_3$ cannot rule out the second-sound scenario. Tsuji and co-workers fit a formula consisting of a spectrum for damped harmonic oscillator and a broad central Lorentzian to the observed spectra, but the fitted formula employed at least six unknown parameters, while our analysis had only five: $\tau_N$, $\omega_0$, $P_1$, $P_2$, and the baseline. Furthermore, the widths for the fitted damped harmonic oscillator and QELS in their analysis could be adjusted independently, whereas $S_1$ and $S_2$ in our ET formula involve common line-shape determining parameters, namely, $\tau_N$ and $\omega_R$, i.e., the two components, $S_1$ and $S_2$, have mutually dependent linewidths.

In KTaO$_3$, which is thought as a similar system to SrTiO$_3$, an extra Brillouin doublet very similar to that in SrTiO$_3$ has also been reported below 15 K. The origin of the new doublet in KTaO$_3$ was first proposed to be due to second sound, then later reinvestigated by Farhi et al., who have shown a possibility that such a broad doublet could arise also from two-phonon difference processes on a single phonon branch if there is elastic anisotropy. In our analysis for SrTiO$_3$, the temperature of 6.4 K corresponds to the Brillouin branch if there is elastic anisotropy. In our analysis for SrTiO$_3$, the temperature of 6.4 K corresponds to the ballistic regime as we showed in Fig. 11. Thus, the scattering process for the low-temperature doublet should be viewed as arising from the TPDS on the same phonon branch, rather than as arising from hydrodynamic second sound, as we pointed out in Sec. III. The doublet-like line shape is obtained also in our model if we assume a frequency dependence for the pair width for an isotropic phonon dispersion: this is compatible with the analysis by Farhi et al.

In the present theory, the temperature dependence of the integrated intensities for both $S_1$ and $S_2$ components have not been given. Also, the observed intensity ratio between the two components, namely, $P_1/P_2$, was strongly temperature dependent, which is not predicted in the present theory. These issues concerning the intensities might be resolved, for instance, by properly including one-phonon (Brillouin) scattering, whose intensity is connected with the intensity of $S_1(q, \omega)$ through the modified Landau-Placzek ratio, and it would also enable reproduction of the interference between the Brillouin peaks and the broader QELS as seen in Fig. 9 as the asymmetry of the LA Brillouin lines.

V. SUMMARY

We calculated light-scattering spectra in a phonon gas from ET equations of Dreyer and Struchtrup. Since the ET-equation set is applicable not only in hydrodynamic regime but also in collisionless regime, the obtained spectral formula can be used in any regime, i.e., in a wide range of temperature and magnitude of $q$.

The spectral formula consists of two parts, namely, the thermal-wave term and the interaction term according to the basing ET-equation set, which can be interpreted as a coupled wave equation for the second sounds in the LA and TA phonon gases. Thus, the two spectral components could be interpreted as arising from the two normal modes in the coupled wave-equation system. The thermal-wave term yields either central peak or broad doublet in the scattering spectrum according to whether the thermal wave is overdamped or underdamped. The interaction term always yields a central peak whose linewidth is equal to the rate of collisions between the thermal phonons. Since, in the hydrodynamic regime, the phonon-collision rate is much higher than the rate of thermal diffusion and than the frequency of first sound, the latter quasielastic component has much broader width than that of the former component or the Brillouin shift. This broad QELS can alternatively be interpreted as the so-called “Mountain mode” because the spectrum comes from the interaction between the second sounds via the nonequilibrium viscosity in the phonon gas.

As the ballistic regime is approached, the two spectral components develops into the same form, which is a rectangular spectrum in the limit of the ballistic regime. In the ballistic limit, both spectral components exhibit $q$-linear dependence for the linewidth, which is in complete contrast to the $q^2$ and $q^0$ dependences in the hydrodynamic regime for the narrow and broad QELS,’s, respectively. We showed that the spectrum in the ballistic regime could alternatively be constructed from the two-phonon difference light-scattering processes concerning two phonons on the same dispersion branch, and that the spectral formula obtained from ET and TPDS coincides with each other in the ballistic limit.

We fitted the spectral formula obtained from ET to the spectra experimentally observed in rutile and SrTiO$_3$. We found good agreements between the fitted parameters and the literature values available. In particular, the temperature dependence of $\tau_N$ in SrTiO$_3$ was determined, and it indicated the existence of the second-sound excitation in the narrow temperature range around 30 K.

ACKNOWLEDGMENTS

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APPENDIX A: CONTINUOUS FRACTION EXPANSION OF THE SPECTRAL FUNCTION

Here we seek direct solutions of Eqs. (A1) for fluctuation spectra of $\epsilon^{(0)}$ and $\epsilon^{(1)}$, rather than those of normal modes. We first perform Fourier-Laplace transform for Eqs. (1) and (2). The resulting equations can be written in a matrix form as the following:

$$ M \cdot v = v_0, \quad (A1) $$

where $M$, $v$, and $v_0$ have been defined as

$$ M = \begin{bmatrix} L & A \\ A' & T \end{bmatrix}, \quad v = \begin{bmatrix} v_1 \\ v_2 \end{bmatrix}, \quad v_0 = \begin{bmatrix} v^{(0)}_1 \\ v^{(0)}_2 \end{bmatrix}. $$

Here $L$, $T$, $A$, $A'$, $v_1$, and $v_2$ are block matrices defined as the followings:
where the wave vector \( q \) must be considered as the wavevector transfer in light-scattering experiments, viz., \( q = (2 \pi \eta / \Lambda) \sin(\theta/2) \), where \( \eta \), \( \Lambda \), and \( \theta \) are the refractive index, the light wavelength, and the scattering angle, respectively.

1. Uncoupled case (independent gases)

First we consider LA and TA modes without interaction with the other modes. Setting \( A=B=0 \), we obtain a subsystem of Eq. (A1) as

\[
L \cdot v_i = v_{i0} \quad \text{(A2)}
\]

or

\[
T \cdot v_i = v_{i0} \quad \text{(A3)}
\]

This system can be regarded as independent or pure phonon gases, which do not interact with other modes and no energy exchange exists. In this case, all we have to do is solve each of Eqs. (A2) and (A3) separately. Each of the subsystems is quite similar to that appears in the analysis of ladder-type electrical circuits.66,67 The present system corresponds to a coupled case of two ladder-type circuits, and the solutions can be expressed as combinations of the solutions for the subsystems.

To obtain the spectrum for mode \( \lambda \), we calculate a quantity \( \langle e^{(\lambda)}(q,0)e^{(\lambda)}(q,s) \rangle \). Thus we may set \( v_0 = [e^{(\lambda)}(q,0), 0, 0, \ldots] \) because of the mutual independence of the variables in each phonon gas.5 It is easily found that in the independent subsystems Eqs. (A2) and (A3),

\[
e^{(\lambda)}(q,s) = (L^{-1})_{11} e^{(\lambda)}(q,0) \frac{\det Q^{(\lambda)}}{\det L} e^{(\lambda)}(q,0) \quad \text{(A4)}
\]

\[
e^{(\lambda)}(q,s) = (T^{-1})_{11} e^{(\lambda)}(q,0) \frac{\det Q^{(\lambda)}}{\det T} e^{(\lambda)}(q,0), \quad \text{(A5)}
\]

where \( \det m \) denotes the determinant of a matrix \( m \), and \( \det Q^{(\lambda)} \) and \( \det \tilde{Q}^{(\lambda)} \) are the \( i=\lambda \) components of the cut matrices for submatrices \( L \) and \( T \), respectively. Therefore, \( Q^{(\lambda)} \) and \( \tilde{Q}^{(\lambda)} \) are obtained by deleting the first row and the first column from \( L \) and \( T \), respectively. The determinants in Eq. (A4) satisfy the following recursive relations:

\[
\frac{\det \tilde{Q}^{(\lambda)}}{\det \tilde{L}_{n}} = \frac{\det Q^{(\lambda)} + \alpha c_{(0)}^2 q^2 \det \tilde{Q}^{(\lambda)}_{n-2}}{\det L_{n}} \quad \text{for} \quad (n \geq 2),
\]

where we have written a matrix with \( i \times i \) components as \( m_i \). We write Eq. (A6) in a matrix form as

\[
\begin{bmatrix}
\det Q^{(\lambda)} \\
\det L_{n-1}
\end{bmatrix} = G_{n-1} \begin{bmatrix}
L_{ni} \\
\alpha c_{(0)}^2 q^2
\end{bmatrix}
\]

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This method is known as “Möbius transformation” or linear fractional transformation. We find that the matrix $G_{i-1}$ can be factorized as

$$G_{i-1} = G_{i-2} \left[ \begin{array}{ccc} L_{i-1} & 1 \\ \alpha_{i-1} c_i q^2 & 0 \end{array} \right] = G_{i-3} \left[ \begin{array}{ccc} L_{i-2} & 1 \\ \alpha_{i-2} c_i q^2 & 0 \end{array} \right] = G_i H_2 H_3 \cdots H_{i-1},$$

where

$$H_i = \left[ \begin{array}{ccc} L_{ii} & 1 \\ \alpha_{ii} c_i q^2 & 0 \end{array} \right].$$

and

$$G_i = \left[ \begin{array}{ccc} \det Q_i^{(i)} & \det Q_0^{(i)} \\ \det L_1 & \det L_0 \end{array} \right] = \left[ \begin{array}{ccc} 1 & 0 \\ L_{11} & 1 \end{array} \right].$$

Hence we see that

$$\begin{bmatrix} \det Q_n^{(i)} \\ \det L_n \end{bmatrix} = G_i H_2 H_3 \cdots H_{i-1} \begin{bmatrix} L_{nn} \\ \alpha_{ii} c_i q^2 \end{bmatrix}.$$ 

We note here that the operation of $H_{i-1}$ to a vector $[L_{ii}, \alpha_{ii} c_i q^2]$, i.e., to a fraction $\frac{L_{ii}}{\alpha_{ii} c_i q^2}$, yields a new fraction of the form

$$\frac{\frac{1}{2} L_{i-1} + \frac{\alpha_{i-1} c_i q^2}{\alpha_{ii} c_i q^2} \gamma_{i-1}}{\frac{1}{2} \alpha_{ii} c_i q^2 \gamma_{i-1}},$$

where $\gamma_i$ has been introduced as

$$\gamma_i = \frac{1}{L_{ii} + \alpha_{ii} c_i q^2 \gamma_{i-1}}, \quad (A9)$$

which is the recursive relation for $\gamma_i$. For an $n$-moment system, $\gamma_{n+1} = 0$ because of the closure theorem.\textsuperscript{14} Successive operations of $H_i$, i.e., $\Pi_{i=2}^{n} H_i$, yields a continued fraction of the form

$$\begin{bmatrix} 1 \\ \gamma_2 \end{bmatrix} = L_{22} + \frac{\alpha_{3} c_3 q^2}{L_{33} + \frac{\alpha_{4} c_4 q^2}{L_{44} + \frac{\alpha_{5} c_5 q^2}{L_{55} + \cdots}}},$$

where $\gamma_i = 1/(L_{ii} + \alpha_{ii} c_i q^2 \gamma_{i-1})$. The right-hand side of Eq. (A7) can be expressed in a continued fraction form as

$$\frac{\det Q_n^{(i)}}{\det L_n} = \gamma_i^{(i)} = \frac{1}{L_{11} + \frac{\alpha_{2} c_2 q^2}{L_{22} + \frac{\alpha_{3} c_3 q^2}{L_{33} + \frac{\alpha_{4} c_4 q^2}{L_{44} + \frac{\alpha_{5} c_5 q^2}{L_{55} + \cdots}}}}}.$$ 

A completely analogous discussion can be made for the TA mode, and we obtain from Eq. (A5) that

$$\frac{\det Q_n^{(i)}}{\det T_n} = \gamma_j^{(i)} = \frac{1}{T_{11} + \frac{\alpha_{2} c_2 q^2}{T_{22} + \frac{\alpha_{3} c_3 q^2}{T_{33} + \cdots}}}.$$ 

Thus, from Eqs. (A4) and (A5), we obtain spectra in continued-fraction forms for the pure LA and TA phonon gases, respectively, as

$$S^{(0)}(q, s) \propto \frac{\langle e^{(0)}(q, 0) e^{(0)}(q, s) \rangle}{\langle e^{(0)}(q, 0) e^{(0)}(q, 0) \rangle} = \gamma_j^{(i)} \quad (A10)$$

and

$$S^{(0)}(q, s) \propto \frac{\langle e^{(0)}(q, 0) e^{(0)}(q, s) \rangle}{\langle e^{(0)}(q, 0) e^{(0)}(q, 0) \rangle} = \gamma_j^{(i)} \quad (A11)$$

2. Coupled case

Next, we consider the coupling between the modes. Similarly to Eqs. (A2) and (A4), $e^{(0)}(q, s)$ can be expressed as

$$e^{(0)}(q, s) = (M^{-1})_{11} e^{(0)}(q, 0) = \frac{\det \Theta}{\det M} e^{(0)}(q, 0),$$

where the matrix $\Theta$ has been defined as follows:

$$\Theta = \begin{bmatrix} Q^{(i)} & a \\ a' & T \end{bmatrix},$$

with

$$a = \begin{bmatrix} 0 & A_{22} & 0 & \cdots \\ 0 & 0 & 0 & \cdots \\ \cdots & \cdots & \cdots & \cdots \end{bmatrix}, \quad a' = \begin{bmatrix} 0 & 0 & \cdots \\ A_{22}' & 0 & \cdots \\ 0 & 0 & \cdots \end{bmatrix},$$

where $a$ and $a'$ are obtained by deleting the first row of $A$ and first column of $A'$, respectively. The determinants of the matrices $M$ and $\Theta$ can be calculated as

$$\det \Theta = \det Q^{(i)} \times \det [T - a' (Q^{(0)})^{-1} a]$$

and

$$\det M = \det L \times \det [T - A' L^{-1} A].$$

Hence, we see that

$$\frac{\det \Theta}{\det M} = \frac{\det Q^{(i)} \times \det [T - a' (Q^{(0)})^{-1} a]}{\det [T - A' L^{-1} A]} = \frac{\gamma_i^{(i)}}{\gamma_i^{(i)}} \frac{\det \Phi}{\det P},$$

where we have defined that
\[ \Phi = T - a'Q^{(f)}^{-1}a, \quad P = T - A'L^{-1}A. \]

We see in Eq. (A12) in comparison with the noninteracting spectrum of Eq. (A10) that the interacting spectrum differs by the factor \( \det \Phi / \det P \), which includes information of the interaction between the modes. This modification factor can be computed by Möbius transformation method similarly to the earlier part of this section when \( n = 3 \), and we obtain
\[
\begin{bmatrix}
\det \Phi_n \\
\det P_n
\end{bmatrix} = \begin{bmatrix}
\Phi_{11} \Phi_{22} + \alpha_2 c_2^2 g_2^2 & \Phi_{11} \\

P_{22} P_{11} - P_{11} P_{21} & P_{11}
\end{bmatrix} \begin{bmatrix}
1 \\
\alpha_2 c_2^2 g_2^2 \gamma_2^{(f)}
\end{bmatrix}.
\]

Care should be paid when \( n = 2 \) noting that
\[ \Phi_{11} = T_{11}, \]
\[ \Phi_{22} = T_{22} - A_{22} A_{22}' Y_{22}^{(f)}, \]
\[ P_{11} = T_{11} - A_{11} A_{11}' Y_{11}^{(f)}, \]

The total spectrum for the considered model is given by
\[ w^{(\lambda)} = w^{(\lambda)}/2B \]
\[ = c^{(\lambda)}_3 c^{(\lambda)'}_3 \left\{ B \left[ s + a^{(\lambda)} A c^{(\lambda)'}_3 - 2 A c^{(\lambda)}_3 c^{(\lambda)'}_3 y^{(\lambda)}_1 \right] \right. \\
+ A \alpha_2 g_2^2 \left[ c^{(\lambda)'}_3 \left( c^{(\lambda)}_3 + c^{(\lambda)'}_3 \right) / c^{(\lambda)}_3 \right] + 2 A^2 B c^{(\lambda)}_3 c^{(\lambda)'}_3 c^{(\lambda)}_3 c^{(\lambda)'}_3 y^{(\lambda)}_1 y^{(\lambda)'}_2 \left. \right\}, \]

with continued fractions defined as the following:
\[ y^{(\lambda)}_1 = 1 \\
= \frac{1}{s + a^{(\lambda)} A c^{(\lambda)'}_3 + \alpha_2 g_2^2 y^{(\lambda)}_3}, \]
\[ y^{(\lambda)}_2 = \frac{1}{s + 1/\tau^{(\lambda)}_R + a^{(\lambda)} B c^{(\lambda)}_3 + \alpha_3 g_3^2 y^{(\lambda)}_3}, \]

where \( w^{(\lambda)} = w^{(\lambda)}/2B \)

\[ = c^{(\lambda)}_3 c^{(\lambda)'}_3 \left\{ B \left[ s + a^{(\lambda)} A c^{(\lambda)'}_3 - 2 A c^{(\lambda)}_3 c^{(\lambda)'}_3 y^{(\lambda)}_1 \right] \right. \\
+ A \alpha_2 g_2^2 \left[ c^{(\lambda)'}_3 \left( c^{(\lambda)}_3 + c^{(\lambda)'}_3 \right) / c^{(\lambda)}_3 \right] + 2 A^2 B c^{(\lambda)}_3 c^{(\lambda)'}_3 c^{(\lambda)}_3 c^{(\lambda)'}_3 y^{(\lambda)}_1 y^{(\lambda)'}_2 \left. \right\}, \]

\[ y^{(\lambda)}_1 = 1 \\
= \frac{1}{s + a^{(\lambda)} A c^{(\lambda)'}_3 + \alpha_2 g_2^2 y^{(\lambda)}_3}, \]
\[ y^{(\lambda)}_2 = \frac{1}{s + 1/\tau^{(\lambda)}_R + a^{(\lambda)} B c^{(\lambda)}_3 + \alpha_3 g_3^2 y^{(\lambda)}_3}. \]

As a check, letting \( B \) be zero, Eq. (A13) reduces to a form as

\[ S^{(\lambda)}(q,s) \approx \frac{1}{1/\gamma^{(\lambda)}_1 + (1/\gamma^{(\lambda)'}_1 - (A c^{(\lambda)}_3)(2 A c^{(\lambda)'}_3))}, \]

which is a coupled-oscillator spectrum as one employed by Barker and Hopfield.28

The total spectrum for the considered model is given by the linear combination of the component spectra, viz.,
the following recursive relations:

\[ S_{\text{total}}(q,s) = p_1 S^{(1)}(q,s) + p_2 S^{(2)}(q,s), \]

which was employed in the analysis presented in Ref. 50.

Since each of \( S^{(1)} \) and \( S^{(2)} \) contains the normal modes corresponding to the second sound and Mountain mode the total spectrum is still composed of these two normal-mode spectra (\( S_1 \) and \( S_2 \)) as those systematically shown in Fig. 5.

**APPENDIX B: LINE SHAPE IN THE BALLISTIC LIMIT (\( \text{Kn} \rightarrow \infty \))**

In the ballistic limit, where \( \text{Kn} \rightarrow \infty \), we obtain that \( A = A' = 0 \). Hence the linear system Eq. (A1) can be reduced to two independent systems, namely, Eqs. (A2) and (A3). As shown in Fig. 4 for \( \text{Kn} = 1000 \), the spectrum in the ballistic limit becomes a round-shaped comb with \( N \) sharp peaks, where \( N = n_{\text{max}} \) is the number of moments in the ET equation.

The peak positions of the comblike spectrum are obtained by setting the denominator of the spectral function of Eqs. (A4) and (A5), namely, \( \det L_N \) or \( \det T_N \), to be zero. For a large integer \( i \) such that \( \alpha_{\pm} = 1/4 \), these determinants satisfy the following recursive relations:

\[
\begin{align*}
\det L_i &= s \det L_{i-1} + \frac{1}{4} c_0^2 q^2 \det L_{i-2}, \\
\det T_i &= s \det T_{i-1} + \frac{1}{4} c_0^2 q^2 \det T_{i-2},
\end{align*}
\]

We assume that the frequency shift of the \( i \)th tooth out of \( N \) teeth of the comb is located at

\[ \omega_N^{(i)} = c \lambda q \cos \phi_N^{(i)}(i), \]

where \( (\lambda) \) denotes the mode index, \( (l) \) or \( (t) \), \( i \) is an integer \( (i=1,2,\ldots,N) \), and \( \phi_N^{(i)}(i) \)'s are angles, which are measured counterclockwise from the horizontal axis. Since \( \omega_N^{(i)}(i) \) is the \( i \)th real root of the dispersion relations in the \((\omega,q)\) space, substituting

\[ s = ic \lambda q \cos \phi_N^{(i)}(i) \]

into the recursive relations Eqs. (B1) and (B2) gives that

\[
\begin{align*}
\det L_N &= \frac{ic^2 q^2}{4} \sin(N+1) \cos \phi_N^{(0)}(0), \\
\det T_N &= \frac{ic^2 q^2}{4} \sin(N+1) \cos \phi_N^{(0)}(0).
\end{align*}
\]

Setting these equal to zero (dispersion relation) shows that the angles \( \phi_N^{(i)} \) should satisfy that

\[ \phi_N^{(i)}(i) = i \times \frac{\pi}{N+1}. \]

Equations (B3) and (B5) show that the positions of the comb teeth coincide with the feet of the perpendicular to the \( \omega \) axis from \( N \) points that are equiangularly located \([\Delta \phi = \pi/(N+1)]\) on a half circle of radius \( c \lambda q \) (see Fig. 12).

The envelope function for the round-shaped comb spectrum can be obtained from the asymptotic expressions for \( \gamma^{(i)} \)

\[ \gamma_c = \frac{2}{cq} \sin \phi_N(i), \]

Thus, we see that the spectrum in the ballistic limit has an elliptic envelope.

From Eqs. (B3) and (B7) for the frequency shifts and the envelope, respectively, we see that the spectrum in the ballistic limit for a \( \text{finite} \) number of \( N \) is a “sine-weighted” and “co-sine-intervaled” comblike spectrum that can be expressed as

\[ S_{\text{bal}}(\omega,q,N) \propto \sum_{i=1}^{N} \sin \phi_N(i) \delta(\omega - cq \cos \phi_N(i)), \]

where the teeth of the comb have been expressed as a sum of sine-weighted delta functions with cosine-intervaled frequency shifts.

Equation (B8) is considered to be an approximated expression for the round-shaped, comblike spectra shown in Fig. 4 for large values of \( \text{Kn} \) and for a large \( \text{but finite} \) number of moments, namely, \( N=30 \) or \( 300 \) without the asymptotic expression. We have recently suggested\(^{36}\) that the limiting spectrum when \( \text{Kn} \rightarrow \infty \) be a half ellipse as Eq. (B8) from analyses with finite numbers of moments, viz., \( N \approx 20 \). But, it is not true for an infinite number of moments as we will show below.

Now let us examine the limiting line shape for an \( \text{infinite} \) number of moments, viz., when \( N \rightarrow \infty \). To do so, we just replace the sum in Eq. (B8) with an integral after introducing an appropriate normalizing factor
\[
\lim_{\tau, N \to \infty} S_{\text{int}}(q, \omega) = cq \int_0^\pi \sin \phi \delta(\omega - cq \cos \phi) d\phi \\
= cq \int_0^\pi \sin \phi \frac{\delta(\omega/cq - \cos \phi)}{|cq|} d\phi \\
= \int_0^\pi \sin \phi \frac{\delta(\phi - \arccos(\omega/cq))}{|\sin(\arccos(\omega/cq))|} d\phi
\]
(B9)

where the normalizing factor \(c^2 q^2/2\) has been chosen such that the height at \(\omega = 0\) is 1. Equation (B11) is a rectangular function, which is nonzero only for the interval \([-cq, cq]\).

A rectangular function can be regarded as a transfer function of an ideal low-pass filter as well known in signal processing. Since an ideal low-pass filter is expected in an infinite ladder-type electric circuit, and since a formal correspondence exists between the ET system and a ladder-type circuit, the rectangular spectrum obtained here seems to be quite natural for the linear system of ET with infinite number of moments.

Instead of assuming a delta function for a tooth of the comb spectrum in Fig. 12, it may be assumed that the comb teeth have a finite but very narrow linewidth such that \(1/\tau \ll \omega_0(i)\), where \(\tau\) is the phonon relaxation time defined in Eq. (11). Replacing the delta function in Eq. (B9) with a Lorentzian with a width of \(1/\tau\), we obtain a complex susceptibility as
\[
\chi(q, \omega) = C cq \int_0^\pi \sin \phi \frac{1}{\omega - cq \cos \phi - i\tau} d\phi, \quad (B12)
\]
where \(C\) is a constant. The real and imaginary parts of \(\chi(q, \omega)\) are calculated, respectively, as
\[
\text{Re}[\chi(q, \omega)] = C cq \int_0^\pi \sin \phi \frac{\omega - cq \cos \phi}{(\omega - cq \cos \phi)^2 + 1/\tau^2} d\phi
\]
\[
= C \int_{\omega - cq \tau}^{\omega + cq \tau} \frac{1}{t^2 + 1} dt
\]
\[
= \frac{C}{2} \ln[(\omega + cq)^2 + 1/\tau^2] - \ln[(\omega - cq)^2 + 1/\tau^2]
\]
and
\[
\text{Im}[\chi(q, \omega)] = C cq \int_0^\pi \sin \phi \frac{1/\tau}{(\omega - cq \cos \phi)^2 + 1/\tau^2} d\phi
\]
\[
= C \int_{\omega - cq \tau}^{\omega + cq \tau} \frac{1}{t^2 + 1} dt
\]
\[
= \frac{C}{2} \tan^{-1}(\omega + cq) \tau - \tan^{-1}(\omega - cq) \tau.
\]
(B14)

It is easy to show that, in the ballistic limit \((cq \tau \to \infty)\), the right-hand sides of Eqs. (B13) and (B14), respectively, become the following forms:
\[
\text{Re}[\chi(q, \omega)]_{cq \tau \to 1} = C \ln \left| \frac{\omega + cq}{\omega - cq} \right|
\]
\[
\text{Im}[\chi(q, \omega)]_{cq \tau \to 1} = C \text{ rect} \left( \frac{\omega}{2cq} \right).
\]

Although we have assumed ballistic regime, it is interesting to investigate the case of hydrodynamic regime. Assuming that \(cq \tau \ll 1\) in Eqs. (B13) and (B14), we see that
\[
\text{Re}[\chi(q, \omega)]_{cq \tau \ll 1} = C \frac{\omega}{\omega^2 + 1/\tau^2},
\]
\[
\text{Im}[\chi(q, \omega)]_{cq \tau \ll 1} = C \frac{1/\tau}{\omega^2 + 1/\tau^2},
\]
which are the real and imaginary parts of a central Lorentzian with a width of \(1/\tau\). Such a central Lorentzian is expected for \(S_2(q, \omega)\) defined in Eq. (24). In fact, this is consistent with the result obtained from Eq. (24) if we assume that \(cq \tau \ll 1\). Therefore, the expression of the right-hand side of Eq. (B14) can be employed not only in the ballistic regime but also in the hydrodynamic regime as an expression for \(S_2(q, \omega)\). Although a slight difference is expected between the exact form [Eq. (24)] and the approximate one [Eq. (B14)] in the intermediate regime, we may reasonably adopt the expressions as in the right-hand side of Eq. (B14) as regime-independent spectral formula for \(S_2(q, \omega)\).
It is assumed that if we set $L$ Lorentzian in thermodynamic limit or that the memory function for the second-sound propagator can be approximated as a $\cos(q, \omega) = 2 \times \arccos(\omega/q)$ for $\omega < 0$.

Two-phonon difference process from different phonon dispersion is also possible in general. In such a case, the two-phonon-dispersion curves must be close at a point in the Brillouin zone. In practical system, such points are usually located at the edges of the Brillouin zone, where phonon dispersions are usually flat. From such flat portion of the phonon dispersion, however, TDPS predicts a rather narrow central peak, which is not consistent with the observed spectra.

The pair width should be $1/\tau$ because it is physically identical with the mean rate of collision between phonons.

In Ref. 42, we have just replaced the delta function with the mean rate of collision between phonons.

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