Weak localization and anharmonicity of phonons

E. Akkermans and R. Maynard

*Centre de Recherches sur les Très Basses Températures, Centre National de la Recherche Scientifique
et Université Scientifique et Médicale de Grenoble (USMG), Boîte Postale 166X,
38042 Grenoble Cédex, France*

The problem of localization of phonons in disordered materials is studied in the framework of the weak-localization theory. Quantum correction to phonon diffusion is calculated by the resummation technique of maximally crossed diagrams in two and three dimensions. The strong energy dependence of the elastic mean free path—the Rayleigh-Klemens scattering—is responsible for the existence of a threshold frequency ω_3^* where the diffusion constant vanishes. The value of ω_3^* depends only on the local fluctuation of masses and on the Debye frequency in three dimensions. This threshold describes the phenomenon of localization of phonon density fluctuations or second sound. The self-energies of the phonons are strongly affected by this quantum correction via the anharmonic interactions. The two basic anharmonic couplings contribute to the one-phonon renormalization and provide shortening of the mean lifetime as well as excess of spectral density in the vicinity of the threshold. In two dimensions, as for the electrons, the dynamical quantum correction diverges logarithmically when the frequency goes to zero. A procedure of convergence is used by cutting off the low-frequency contributions at the inelastic relaxation rate. Renormalization of phonons are obtained in a self-consistent way. Finally a tentative application of the previous results to the low-temperature properties of glasses is discussed. In particular the existence of a plateau in thermal conductivity accompanied by excess specific heat in all the glasses measured so far could be understood as the manifestation of localization of acoustic-phonon density at the critical threshold ω_3^*.

I. INTRODUCTION

The problem of localization of particles in a disordered medium has received considerable attention in recent years.[1] Important progress has been obtained in describing electron diffusion in the weak-localization regime where both the elastic scattering and the interparticle interaction are treated as perturbations. One important result is the quantum correction to the two-particle propagator which decreases the diffusion constant of the density fluctuations by constructive interferences of the backscattering at the site origin. This quantum correction has been verified by magnetoresistance experiments where the magnetic field destroys the coherence. The transposition of this new concept to phonons is the main motivation of this article and has never been attempted as yet to our knowledge. Passing from electrons to phonons implies many modifications which must be carefully discussed, particularly the relevance of the statistics (bosons or fermions) for building up the diffusion kernel of the two-particle interaction as well as the different dispersions: linear for the phonons and quadratic for electrons. A striking difference arises from the strong frequency dependence of the characteristic length or time in the phonon problem. In particular, the strong energy dependence of the elastic mean free path must be contrasted to that of the electron whose mean free path varies both smoothly and weakly. Moreover, the Fermi-Dirac statistics restrict the range of energy to a narrow vicinity of the Fermi level at low temperatures. Let us call the elastic relaxation time $\tau(\omega)$; it is well known that it obeys the Rayleigh-

Klemens[2] law which is written in d dimensions:

$$\frac{1}{\tau(\omega)} = \sigma_M^2 \omega^2 g_d(\omega) , \qquad (1)$$

where σ_M^2 is the local fluctuation of masses—or harmonic restoring forces—and $g_d(\omega)$ is the spectral density of modes (per atom) in d dimensions:

$$g_d(\omega) = d \frac{\omega^{d-1}}{\omega_D^d} , \qquad (2)$$

ω_D being the Debye cutoff frequency of a unique branch of phonons we will consider here. In three dimensions, the elastic relaxation time varies strongly, as ω^{-4}, which clearly distinguishes phonons from electrons: A dramatic consequence of this ω dependence will be the vanishing diffusivity of phonon density fluctuations—called second sound—at a threshold frequency ω_3^*. Although the critical localization phenomenon is, strictly speaking, beyond the perturbation theory of weak localization, we believe that the occurrence of this critical frequency ω_3^* for the diffusion constant reveals the existence of a genuine threshold of mobility for phonons in three dimensions. The same phenomenon, absence of diffusion, for the static case is a signature of the general localization of modes in two dimensions. This result confirms the recent work of John *et al.*[3] where a mobility edge was found in three dimensions by a renormalization treatment of the nonlinear σ model. Here, a quite different approach based on diagrammatic expansion of the maximally crossed diagrams[4] in the weak-localization regime gives a correct

starting point for a scaling treatment. Another new consequence of the reduction of the diffusion constant at high frequency is the shortening of the mean lifetime of phonons. Phonons are coupled to the phonon density fluctuations by anharmonic interactions; this effect provides the well-known Landau-Rumer[5,6] mean lifetime of phonons in a perfect crystal. The counterpart of the Landau-Rumer attenuation time is obtained here in the weak-localization regime, and the prediction of a considerable reduction of the phonon lifetime at high frequency when ω approaches ω_3^*, or at high temperature when T tends to T^* ($\equiv \hbar \omega_3^* / k_B B$), is obtained directly from the coupling of the phonon with local-density fluctuations.

This drastic shortening of the phonon mean free path seems to us very important in the context of glasses.[7] As a matter of fact, it has been well known for a long time that the thermal conductivity of glasses at low temperature has a plateau, even a dip sometimes, around 10 K. The value of the conductivity at the plateau is very low, of the order of 10^{-1} W m^{-1}K^{-1}. The mean free path derived from the kinetic formula of conduction is very short, typically a few angstroms, sometimes less. Accompanying this plateau, an excess specific heat has been observed in the same temperature range. These observations have inspired many attempts at explanation during the last decade. Let us mention the most recent one where it was assumed that, at short length scales, glasses have a fractal structure.[8] In this model, the spectral density of modes changes from phonons to fractons at a characteristic frequency related to the length scale of the fractal structure. Since the fractons are localized they do not contribute to the thermal conductivity but only to the specific heat. Although describing qualitatively the experimental situation, this model calls for careful structural analysis of the local order in a possible fractal structure. In contrast, without any special assumption as to the structure of glasses, we believe that present weak localization and anharmonicity theory provide an indication for the occurrence of a plateau in the thermal conductivity and the excess specific heat.[7] We note that no special parameters are necessary in our model except the basic features of phonons: sound velocity and Debye temperature. It is, therefore, natural that all glasses exhibit this universal behavior of a plateau in the thermal conduction and excess specific heat.

This article is organized in four sections. In Sec. II, the quantum correction to diffusion is calculated by the well-known series expansion of the maximally crossed diagrams of the two-phonon propagator. The diffusion constant of phonon density (second sound) exhibits a strong frequency dependence and vanishes in three dimensions at a critical value ω_3^*. In two dimensions the same diffusion constant is defined only in the dynamical regime $\Omega \neq 0$, as expected from the general result of localization in two dimensions.[9,10] A simple physical picture of this quantum correction is put forward in terms of the ratio of the residence time of the two-phonon coherent state at the origin over the lifetime of the same two-phonon state. Section III is devoted to the self-energy of phonons coupled via anharmonicity to density fluctuations. The two basic diagrams of the anharmonic expansion, to third and

fourth order, are modified in order to take into account the formation of the two-phonon coherent backscattered state. As a result the phonon lifetime is obtained as a function of frequency and temperature and is strongly reduced when ω tends towards ω_3^* or T towards T^*. In a similar way, the density of states is obtained for both anharmonic interactions and shows an important enhancement around ω_3^*. In Sec. IV, the problem of two-dimensional weak localization is carefully analyzed, since a low-frequency cutoff is necessary for preventing the infrared divergence. A discussion of the basic expressions of the phonon self-energy is developed in terms of the cutoff frequency. The possible applications of the previous results to low-temperature thermal properties of glasses are discussed in Sec. V, while this work is summarized in Sec. VI.

II. QUANTUM CORRECTION TO DIFFUSION AND THE EXPRESSION FOR THE PHONON DIFFUSION KERNEL

The analysis for the phonon diffusion kernel is based on the standard diagrammatic techniques[11] for a particle moving in the field of randomly distributed impurities considered as elastic scatterers. The transport coefficients or the diffusion constant can be derived from the average two-phonon Green's function

$$G_2(\mathbf{k},\mathbf{k}';\omega,\Omega) = \langle G_k^R(\omega + \Omega)G_{k'}^A(\omega) \rangle , \qquad (3)$$

where $\langle \ \rangle$ represents the mean value over all configurations of impurity centers, and where $G_k^R(\omega + \Omega)$ and $G_{k'}^A(\omega)$ describe, respectively, the retarded and advanced one-phonon Green's function with momentum $\mathbf{k}$ and energy $\omega + \Omega$ ($\mathbf{k}'$ and ω, respectively).

The first diagrams for G_2 are depicted in Fig. 1. In order to calculate the dominant contribution to G_2 given in Eq. (3), the most divergent terms in the density expansion must be resummed.[4] This resummation will ultimately lead to localization of density fluctuations.

Let us start from the well-known Bethe-Salpeter equation (rederived in Appendix A) for $G_2(\mathbf{k},\mathbf{k}';\omega,\omega')$:

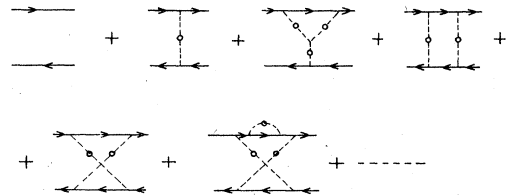FIG. 1. Relevant diagrammatic expansion for $G_2(\mathbf{k},\mathbf{k}',\omega,\Omega)$. The lines with arrows correspond to the one-phonon Green's function and the dashed lines represent the Fourier transforms of the interaction potential between a phonon and an impurity.

$$G_2(\mathbf{k},\mathbf{k}';\,\omega,\omega') = G_k^A(\omega)G_{k'}^R(\omega')\left[\delta_{\mathbf{k},\mathbf{k}'} + \sum_{\mathbf{k}_1} U_{\mathbf{k},\mathbf{k}_1}(\omega,\omega')G_2(\mathbf{k}_1,\mathbf{k}';\,\omega,\omega'\right]\ . \tag{4}$$

This equation relates the two-phonon propagator to the one-phonon propagators $G_k^{A,R}$ and the irreducible part of the vertex interaction $U_{\mathbf{k}\mathbf{k}'}(\omega,\omega')$ as shown in Fig. 2.

U is built up from impurity scatterers which break the momentum conservation law but conserve the energy. At first sight it may be surprising that the interaction $U_{\mathbf{k},\mathbf{k}'}(\omega,\omega')$ couples two phonons of different energy $\hbar\omega$ and $\hbar\omega'$, while the basic scattering is elastic. But this is true and it results from the quantum treatment of diffusion. Let us call $D_0(\omega)$ the diffusion constant of *one phonon* ω. Classically we know that diffusion introduces a relationship between space (or k) and time variables. By breaking the translational invariance and, consequently, changing the $\mathbf{k}$'s at each collision, the diffusive one-phonon motion introduces a frequency dependence $\omega - \omega' \sim D_0(k - k')^2$ which, by virtue of the uncertain energy-time relation, extends the energy shell for the interaction. The dominant contribution to U comes from the maximally crossed diagrams (or fan-shaped diagrams) which can be summed up as a geometrical series.[12] The most divergent part of these diagrams comes from backscattering where $\mathbf{k}' = -\mathbf{k} + \mathbf{q}$, q small $ql(\omega) \ll 1$, and $|\omega' - \omega| = \Omega$ small, $\Omega\tau(\omega) \ll 1$, where $l(\omega) = v\tau(\omega)$ is the elastic mean free path of a phonon ω. In Appendix A, we report the calculation of the vertex U which, at the lowest order in Ω and q in the denominator gives

$$U(q;\Omega,\omega) = \frac{2v^4}{\pi D_0^2(\omega)}\frac{\hbar^2}{g_d(\omega)}\frac{1}{-i\Omega + D_0(\omega)q^2}\ , \tag{5}$$

where v is the sound velocity for the unique branch of acoustic phonons $\omega = vk$ as assumed and the spectral density $g_d(\omega)$ is given by Eq. (2). The expression given by Eq. (5) of the diffusion vertex calls for some remarks.

(1) It is the phonon counterpart of the electron diffusion vertex which has been established in the context of impure metals [see, for example, Eq. (16) of Ref. 12]. In Eq. (5), ω stands for Fermi energy in the electron problem while $D_0(\omega)$ represents the diffusion coefficient of electrons at the Fermi level.

(2) Although the acoustic approximation $\omega = vk$ is used in the expansion in q of the denominator, a term in q^2 is obtained in two and three dimensions as in the electron problem where $E \sim k^2$. This is not true in one dimension where the general condition of validity of perturbation expansion is not fulfilled.[12]

(3) The diffusion coefficient $D_0(\omega)$ is related to the Rayleigh-Klemens scattering time $\tau(\omega)$ or the elastic
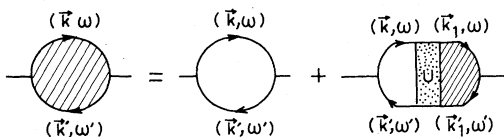
linewidth of the phonon propagator (see Appendix A), $\Gamma(\omega) = \hbar/\tau(\omega)$ by the well-known relation

$$D_0(\omega) = \frac{v^2\tau(\omega)}{d} = \frac{\hbar v^2}{d\Gamma_d(\omega)}\ . \tag{6}$$

Since the Rayleigh scattering frequency is proportional to σ_M^2, the local fluctuations of masses, U varies like $\sigma_M^4 g_d(\omega)$ as expected. Therefore, the first term of the series expansion is a good representation of the X or crossed diagram.

(4) The denominator of Eq. (5) exhibits a diffusion pole: It implies that the dominant contribution will come from the vicinity of the diffusion pole $\Omega \sim D_0 q^2$ in the Ω-q plane. Let us define the diffusion coefficient $D(\omega,\Omega)$ by the velocity currents correlation function.[9,13] This leads, in the limit of acoustic phonons, to the following expression:

$$D(\omega,\Omega) = \frac{v^2}{d}\frac{(\hbar v)^2}{\pi\omega^2 g_d(\omega)}\sum_{\mathbf{k},\mathbf{k}'}\mathbf{k}\cdot\mathbf{k}' G_2(\mathbf{k},\mathbf{k}';\,\omega,\omega')\ . \tag{7}$$

For $\mathbf{k} + \mathbf{k}' = \mathbf{q}$ and $\Omega = |\omega' - \omega|$, as before, we expand the Green's functions in powers of Ω and q^2, and we solve the Bethe-Salpeter equation given by Eq. (2) to first order to obtain

$$G_2(\mathbf{k},\mathbf{k}';\,\omega,\omega')$$

$$= G_k^A(\omega)G_{k'}^R(\omega')\delta_{\mathbf{k},\mathbf{k}'}$$

$$+ G_k^A(\omega)G_{k'}^R(\omega')U_{\mathbf{k},\mathbf{k}'}(\omega,\omega')G_k^A(\omega)G_{k'}^R(\omega')\ , \tag{8}$$

where

$$U_{\mathbf{k},\mathbf{k}'}(\omega,\omega') = U(q,\Omega,\omega)$$

is given by Eq. (5). By replacing Eq. (8) in the expression of $D(\omega,\Omega)$, one obtains two terms. The first one proportional to $\sum_k k^2 G_k^A(\omega)G_k^R(\omega')$ gives $D_0(\omega)$, the single-phonon diffusion coefficient, while the second produces the quantum correction to classical diffusion, and involves the scalar product $\mathbf{k}\cdot\mathbf{k}' = \mathbf{k}\cdot(\mathbf{q} - \mathbf{k})$.

Just as before (cf. Appendix A), the sum $\sum_k \mathbf{k}\cdot\mathbf{q}$ cancels and there remains only a term proportional to $-k^2$ which gives finally, for $D(\omega,\Omega)$,

$$D(\omega,\Omega) = D_0(\omega)\left[1 - \frac{2}{\pi g_d(\omega)}\sum_{\mathbf{q}}'\frac{1}{-i\Omega + D_0(\omega)q^2}\right.$$

$$\left. + \cdots\right]\ . \tag{9}$$

This is the expression for the quantum correction to the dynamical diffusion coefficient at frequency Ω, associated with the density fluctuations. It was first derived by Gor'kov et al.[9] for electrons for $d = 1,2,3$ in a very similar form. The summation $\sum_{\mathbf{q}}'$ indicates a restriction in

FIG. 2. Bethe-Salpeter equation for the two-phonon Green's function.

the range of q for which the regime of density fluctuations is hydrodynamic: $ql \ll 1$ and $\Omega\tau \ll 1$.

The quantum correction term is complex, but the imaginary part is reduced from the real part by a factor $(\Omega\tau)^{1/2}$ for $d=3$ and $(-\ln\Omega\tau)^{-1}$ for $d=2$ and will be neglected. The calculation of the integral over q is straightforward and gives, in the limit $\Omega\tau \ll 1$, the following expressions: for $d=3$,

$$D(\omega)=D_0(\omega)\left[1-\frac{3}{\pi^2\rho}\frac{\tau}{g_3(\omega)l^3}\right], \qquad (10)$$

and for $d=2$,

$$D(\omega,\Omega)=D_0(\omega)\left[1-\frac{1}{\pi^2\rho}\frac{\tau}{g_2(\omega)l^2}\ln\left[\frac{1}{\Omega\tau}\right]\right], \quad (11)$$

where $\rho=N/L^d$ is the density.

It is remarkable that at three dimensions $D(\omega)$ does not depend on frequency Ω, while at two dimensions the ln dependence of $D(\omega,\Omega)$ introduces a singularity when $\Omega\to 0$ which is considered as a signature for the localization of the two-dimensional (2D) systems. Since $l=v\tau$ and $\tau(\omega)$ varies for Rayleigh scattering as $\omega^{-(d+1)}$, we obtain finally the following ω dependence for phonons of energy ω: for $d=3$,

$$D(\omega)=D_0(\omega)\left[1-\left[\frac{\omega}{\omega_3^*}\right]^6\right], \qquad (12)$$

with $\omega_3^*=0.51\sigma_M^{-2/3}\omega_D$, and for $d=2$,

$$D(\omega,\Omega)=D_0(\omega)\left[1-\left[\frac{\omega}{\omega_2^*}\right]^2\ln\frac{1}{\Omega\tau(\omega)}\right], \qquad (13)$$

with[14] $\omega_2^*=(2\sigma_M/\pi)^{-1}\omega_D$.

At $d=3$, the relation (12) describes a vanishing *static* diffusion constant when ω approaches the threshold ω_3^*. For the extreme case of disorder—glasses for instance—$\sigma_M \simeq 1$ and ω_3^* is less, but not far from, ω_D. This result has been established for Rayleigh-type scattering relaxation time which is valid only for dilute concentration of impurities. Although the validity of the present calculation in the weak-localization limit is of a perturbative nature and restricted to $\omega \ll \omega_3^*$, we observe that the quantum correction to the diffusion constant increases strongly with ω up to a threshold frequency ω_3^* where a complete absence of diffusion occurs. It must be realized that this absence of diffusion affects the fluctuations of phonon density. Since these excitations of the phonon density are called second sound, we could describe this effect as *the localization of second sound near ω_3^**. In fact, the one-phonon mean free path is also strongly affected by this coherence effect through the anharmonic coupling to this density fluctuation.

This result distinguishes the present theory from a previous approach of John et al.[3] where a mobility edge is found by scaling considerations on the one-phonon problem. The two-dimensional case needs special discussion since, as shown by Eq. (13), the diffusion constant vanishes for any energy in the static regime $\Omega\to 0$ (see Sec. IV).

A simple physical interpretation of weak localization has been largely used recently for the electron localization problem in terms of coherent backscattering.[15] We think that a similar analysis could be very useful here in the context of phonons and will make more transparent the underlying physical picture.

Let us call η the dimensionless quantum correction to diffusion from Eq. (9):

$$\eta \sim \frac{1}{L^d g_d\rho}\sum_{q}{}'\frac{1}{-i\Omega+D_0q^2}. \qquad (14)$$

The diffusion pole in Eq. (14) given by the expression $(-i\Omega+D_0q^2)^{-1}$, is nothing but the Fourier transform in q and Ω of the probability density

$$P(\mathbf{r},t)\sim(Dt)^{d/2}\exp(-r^2/4Dt)$$

for a diffusing particle starting from $\mathbf{r}=0$ at $t=0$. Then the sum over q up to $|q|=1/l$ describes precisely the residence time $\Theta(\Omega)$ up to $t=1/\Omega$ for the diffusing particle to be found in a volume l^d in the neighborhood of $\mathbf{r}=0$:

$$\Theta(\Omega)=\int dt\, e^{i\Omega t}\int_{(l^d)}d^dr\, P(\mathbf{r},t), \qquad (15)$$

where the spatial integration of $P(\mathbf{r},t)$ represents the return probability near the origin in a sphere of radius l.

By a direct calculation of Eq. (15) using the standard expression of $P(\mathbf{r},t)$ it is easy to derive Eqs. (10) and (11) for $d=2$ and $d=3$: $\Theta\sim\tau$ for $d=3$ and $\Theta\sim\tau\ln(1/\Omega\tau)$ in $d=2$ where a divergence is obtained for $\Omega\to 0$ since the two-dimensional random walk is *recurrent*, but so far this argument is purely classical; how does the quantum nature enter into the problem? First, it is the return probability in the sphere l^d which measures the occurrence of a two-coherent-phonon pair $(+\mathbf{k},-\mathbf{k}+\mathbf{q})$ to build up. Secondly, the residence time Θ in the initial sphere is, therefore, directly related to the lifetime of the coherent backscattered pair. The characteristic time $\rho g_d(\omega)l^d$ represents the lifetime of a two-phonon state confined in a sphere of volume l^d. The quantum correction is, therefore, directly proportional to the ratio of the residence time in the sphere l around the origin to the mean lifetime of the two-phonon state in the sphere l. A natural maximum value of one is then obtained which corresponds to the situation of localization where the residence time of the coherent pair is comparable to the persistence time of the two-phonon state. For this limiting situation D vanishes. The localization of phonon density fluctuations occurs because this ratio increases strongly near ω_3^* due, basically, to the strong frequency dependence of the Rayleigh scattering at three dimensions. At two dimensions all the stationary modes are localized as expected, owing to the recurrent nature of the random walk for $d\leq 2$.

The strong decrease of the diffusion constant D when the frequency increases must also renormalize the diffusion vertex U. We propose to take into account this renormalization, following Gor'kov et al.,[9] by redefining a diffusive vertex $\tilde{U}(q;\omega,\Omega)$:

$$\tilde{U}(q;\omega,\Omega)\simeq\frac{2v^4}{\pi D^2(\omega,\Omega)}\frac{\hbar^2}{g_d(\omega)}\frac{1}{-i\Omega+D(\omega,\Omega)q^2}. \qquad (16)$$

This has not been strictly derived from the diagram resummation techniques but should constitute a better approximation for the calculation of the transport coefficients.

III. THREE-DIMENSIONAL ANHARMONIC INTERACTIONS

In a perfect crystal, the first two anharmonic terms, γ_3 and γ_4, arising from the expansion of the Hamiltonian of the system, give rise to the lowest order of perturbation, to interactions between a given phonon $(\omega, \mathbf{k})$, and the thermal phonons in equilibrium in the system. Such interactions are shown diagrammatically in Figs. 3(a) and 3(b).

The γ_4 diagram represented in Fig. 3(b) corresponds to the excitation of a virtual phonon. In a pure crystal, such an interaction process cannot produce a finite lifetime and only renormalizes the sound velocity.

The γ_3^2 diagram described in Fig. 3(a) represents the coupling of the incoming phonon with two thermal phonons of the medium. The lowest-order diagram of perturbation theory has two vertices of interaction and then is proportional to γ_3^2. Actually, the perturbation expansion of the anharmonic interaction shows that γ_3^2 is of the same order of magnitude as γ_4. Unlike γ_4, the γ_3^2 diagram provides a finite lifetime for the given phonon. It is this diagram which leads to the well-known Landau-Rumer[5] lifetime τ for low-frequency phonons, where τ^{-1} is proportional to $\omega T C_v(T)$, $C_v(T)$ being the specific heat. For impure materials or glasses, the presence of Rayleigh
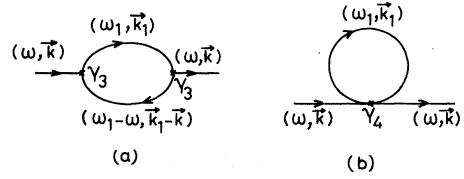
FIG. 3. Relevant diagrams for the anharmonic interactions in crystals.

scattering will produce an additional interaction between the excited phonons and the incoming phonon of the diagram given in Fig. 3(b) or the two thermal phonons of diagram given by Fig. 3(a). Via this anharmonic coupling, γ_3 and γ_4, the phonons, will be coupled to the phonon density fluctuations and will acquire a strongly reduced mean lifetime. Let us calculate separately both contributions to the renormalization of the phonon self-energy.

A. Self-energy for the γ_4 interaction

The modified diagram including the diffusion kernel $\widetilde{U}$ from Eq. (16) is represented in Fig. 4. The basic process is related to the coherent backscattering interference between the incoming phonon $(\omega, \mathbf{k})$ and the thermal excited phonon created by γ_4.

If $V^{(4)}(\mathbf{k}, -\mathbf{k}, \mathbf{k}+\mathbf{q}, -\mathbf{k}-\mathbf{q})$ represents the anharmonic interaction potential, we can write for the self-energy $\Sigma_3^{(4)}(\omega)$ the following expression:

$$\Sigma_3^{(4)}(\omega) = k_B T \frac{V}{g(\omega)} \sum_{\mathbf{k}, \mathbf{k}'} \sum_{\mathbf{q}, p} V^{(4)}(\mathbf{k}, -\mathbf{k}, \mathbf{k}+\mathbf{q}, -\mathbf{k}-\mathbf{q}) \langle G^R_{-\mathbf{k}-\mathbf{q}}(\omega_p) \delta(\omega - \omega(k)) \rangle \delta_{\mathbf{k}', -\mathbf{k}-\mathbf{q}} , \tag{17}$$

where ω_p represents the usual Matsubara frequency and $\langle\ \rangle$ the average value associated with the disorder. In $\Sigma_3^{(4)}(\omega)$, an average over all the $\mathbf{k}$ on the energy shell ω must be performed. With the standard relation

$$\delta(\omega - \omega(k)) = -(\hbar/2i\pi)[G^R_k(\omega) - G^A_k(\omega)]$$

and with the help of Eqs. (4) and (16) we can show that (cf. Appendix B)

$$\Sigma_3^{(4)}(\omega) = \frac{(\hbar v)^4 \omega^2 V}{4\pi \Gamma(\omega) g(\omega)} \gamma_4 \frac{1}{D^2(\omega)} \int d\omega'\, n(\omega'/T) \frac{1}{\Gamma^2(\omega) + \hbar^2(\omega' - \omega)^2} {\sum_{\mathbf{q}}}' \frac{1}{i(\omega - \omega') + D(\omega)q^2} , \tag{18}$$

where $n(\omega'/T)$ is the Bose-Einstein occupation factor.

The inverse lifetime $\Gamma_3^{(4)}(\omega)$ is given by the imaginary part of $\Sigma_3^{(4)}(\omega)$. Considering first the integration over the $\mathbf{q}$ variable from 0 to $l^{-1}(\omega)$, one has
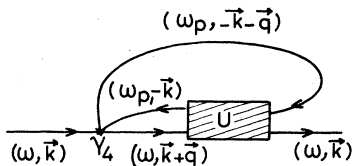
FIG. 4. Diagram contributing to the γ_4 anharmonic interaction. Note the coupling of the loop phonon with the $(\omega, \mathbf{k})$ phonon due to the effective $\widetilde{U}$ interaction.

$$\frac{1}{V} \sum_{\mathbf{q}} \frac{\omega - \omega'}{(\omega - \omega')^2 + (Dq^2)^2} = 2\pi(\arctan 2) \cdot \left[\frac{|\omega - \omega'|^{1/2}}{D^{3/2}(\omega)} \right] . \tag{19}$$

We then obtain for the expression $\Gamma_3^{(4)}(\omega)$,

$$\Gamma_3^{(4)}(\omega) = \frac{\arctan 2}{2} \frac{\hbar^4 v^4 \omega^2}{\Gamma^2(\omega) g(\omega)} V \frac{\gamma_4}{D^{7/2}(\omega)}$$

$$\times \int d\omega'\, n(\omega'/T) \frac{|\omega' - \omega|^{1/2} \Gamma(\omega)}{\Gamma^2(\omega) + \hbar^2(\omega - \omega')^2} . \tag{20}$$

The function

$$\frac{\Gamma(\omega)}{\Gamma^2(\omega) + \hbar^2(\omega - \omega')^2}$$

defines a Lorentzian with width $\Gamma(\omega)$ and height $\Gamma^{-1}(\omega)$, centered at $\omega'=\omega$. We approximate this Lorentzian by a rectangular function with the same width and height, and then

$$\int d\omega' n(\omega'/T)\frac{|\omega-\omega'|^{1/2}\Gamma(\omega)}{\Gamma^2(\omega)+\hbar^2(\omega-\omega')^2}$$

$$\simeq n(\omega/T)\frac{\pi}{2\hbar^{1/2}}\Gamma^{1/2}(\omega) , \quad (21)$$

and finally $\Gamma_3^{(4)}(\omega)$ reads

$$\Gamma_3^{(4)}(\omega)=\frac{\arctan2}{12}\gamma_4\Gamma^2(\omega)\frac{n(\omega/T)}{[1-(\omega/\omega_3^*)^6]^{7/2}} . \quad (22)$$

Some specific features must be emphasized for the mean attenuation $\Gamma_3^{(4)}(\omega)$. The condition for the most divergent diffusion kernel implies that $|\omega-\omega'|\tau\ll1$. This condition is fulfilled here when the excited phonon ω' has nearly the same frequency as the incoming phonon ω. This dominant contribution in the expression of $\Sigma_3^{(4)}(\omega)$ of the thermal phonons belonging to the energy shell $\hbar\omega$ of the incoming phonon explains the Planck occupation number in the final expression given by Eq. (22). At low frequency and low temperature $\Gamma_3^{(4)}(\omega)$ is very weak as compared to $\Gamma(\omega)$, and it is at higher frequency $\omega\sim\omega_3^*$ and high temperature $(T\sim T^*)$ that the effect of localization will act efficiently.

As for the renormalized density of states, it is defined by

$$g^{(4)}(\omega)=-\pi^{-1}\sum_k \mathrm{Im}D_k(\omega) .$$

If $\widetilde{\omega}(k)$ are the new eigenenergies of the system then

$$\widetilde{\omega}(k)=\omega(k)+\Delta_3^{(4)}(\widetilde{\omega}(k)) ,$$

where $\Delta_3^{(4)}(\widetilde{\omega}(k))$ is the real part of $\Sigma_3^{(4)}(\omega)$. An expansion

$$\Delta_3^{(4)}(\omega)=\Delta_3^{(4)}(\widetilde{\omega}(k))+[\omega-\widetilde{\omega}(k)]\frac{\partial\Delta_3^{(4)}}{\partial\omega}$$

of $\Delta_3^{(4)}(\omega)$ around $\widetilde{\omega}(k)$ gives the following expression for the renormalized density of states:

$$g^{(4)}(\omega)=\frac{A(\omega)}{\pi\hbar}g(\omega) , \quad (23)$$

where

$$A(\omega)\equiv\left[1-\frac{1}{\hbar}\frac{\partial\Delta_3^{(4)}}{\partial\omega}\right]^{-1}$$

with for $\Delta_3^{(4)}(\omega)$ the expression derived from Eq. (18):

$$\Delta_3^{(4)}(\omega)=\frac{1}{12}\gamma_4\Gamma^2(\omega)\frac{n(\omega/T)}{[1-(\omega/\omega^*)^6]^3} . \quad (24)$$

This expression shows a great enhancement of the density of states near ω_3^* and $T\sim T^*$. This situation will be confirmed by the calculation of the self-energy $\Sigma_3^{(3)}(\omega)$.

B. Self-energy $\Sigma_3^{(3)}(\omega)$ associated with γ_3^2 interactions

The diagram associated with this interaction is represented in Fig. 5, but the structure of this diagram is more "classic" than those of the γ_4 interaction. The constructive interferences between excited phonons k and $q-k$ is described by the renormalized diffusive kernel $\widetilde{U}$ of Sec. II. By considering the breakdown of translational invariance, it is natural to consider only local fluctuations of phonon density coupled to the incoming phonon. This implies a violation of the momentum conservation law and consequently an unrestricted summation over momentum k and q in the self-energy. Then, we have

$$\Sigma_3^{(3)}(\omega)=k_B T\frac{\hbar}{g(\omega)}\sum_{k,k'}\sum_{q,p}V^{(3)}(k,k',q-k')V^{(3)}(k'-q,k,-k')\delta(\omega-\omega(k))\langle D_k^R(\omega_p)D_{q-k'}^A(\omega_p-\omega)\rangle , \quad (25)$$

where the interaction potential $V^{(3)}$ is defined by

$$V^{(3)}(k_1,k_2,k_3)=\gamma_3(\hbar/2)^{3/2}[\omega(k_1)\omega(k_2)\omega(k_3)]^{1/2} .$$

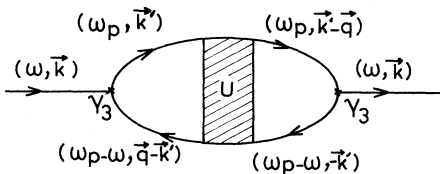In a way quite similar to those studied in Appendix B, we obtain

FIG. 5. Graph related to the γ_3^2 anharmonic interaction. Note the difference between γ_4 and γ_3^2 relative to the nature of the coupling to the density fluctuations.

$$\Sigma_3^{(3)}(\omega)=\frac{\pi}{2}\gamma_3^2\hbar^5 v^4\omega\frac{V}{N}\int d\omega' n(\omega'/T)\frac{(\omega')^2}{\Gamma^3(\omega')D^2(\omega,\omega')}$$

$$\times\sum_q\frac{1}{-i\omega+D(\omega,\omega')q^2} , \quad (26)$$

which gives for the inverse lifetime $\Gamma_3^{(3)}(\omega)$ associated with this anharmonic process

$$\Gamma_3^{(3)}(\omega)=\frac{3\pi}{4}(\arctan2)\gamma_3^2\frac{(\hbar\omega)^{3/2}}{\omega_D^3}$$

$$\times\int d\omega' n(\omega'/T)\frac{\Gamma^{1/2}(\omega')(\omega')^2}{[1-(\omega'/\omega^*)^6]^{7/2}} , \quad (27)$$

and for the real-part $\Delta_3^{(3)}(\omega)$ of the self-energy

$$\Delta_3^{(3)}(\omega)=\frac{3\pi}{2}\gamma_3^2\frac{\hbar\omega}{\omega_D^3}\int d\omega'\frac{\Gamma(\omega')(\omega')^2}{[1-(\omega'/\omega^*)^6]^3}n(\omega'/T) . \quad (28)$$

Both the expressions given by Eqs. (27) and (28) are divergent when ω' approaches ω_3^*, but we know that this limit is out of range of validity of our calculation of weak localization. A convergent version of Eqs. (27) and (28) can be obtained in the approximation of the dominant phonon at temperature T by replacing the summation over ω' by the average value $\omega' \simeq k_B T/\hbar$ in Eqs. (27) and (28). Therefore, approximate expressions are

$$\Gamma_3^{(3)}(\omega) = \frac{3\pi \arctan 2}{4} \sigma_M \gamma_3^2 (\hbar \omega_D)^{1/2}$$

$$\times (\hbar \omega)^{3/2} \frac{1}{[1-(T/T^*)^6]^{7/2}} \left[\frac{T}{\Theta_D}\right]^5 \qquad (29)$$

and

$$\Delta_3^{(3)}(\omega) = \frac{3\pi}{2}(\hbar \omega_D)\sigma_M^2 \gamma_3^2(\hbar \omega) \left[\frac{T}{\Theta_D}\right]^7 \frac{1}{[1-(T/T^*)^6]^3} , \qquad (30)$$

where the correction to the density of states is always given by Eq. (23), i.e., the relative variation $\delta g/g$ is given by

$$\frac{\delta g}{g} \simeq \frac{3\pi}{2}(\hbar \omega_D)\sigma_M^2 \gamma_3^2 \left[\frac{T}{\Theta_D}\right]^7 \frac{1}{[1-(T/T^*)^6]^3} . \qquad (31)$$

Just as before, this expression provides a great enhancement of the density of states for $T \sim T^*$ for all the spectrum, even for phonons with $\omega < \omega^*$.

IV. TWO-DIMENSIONAL CASE

We can, as before, find the correction to the lifetime and the density of states in a two-dimensional disordered system due to anharmonic coupling. Now, however, we must take into account the *dynamic singularity* given by Eq. (13) for the diffusion coefficient. Because of the logarithmic divergence of the quantum correction in the limit $\Omega \tau \ll 1$, we define a cutoff frequency $\tilde{\Omega}$ such that $D(\omega,\tilde{\Omega})=0$. Such a cutoff is imposed by the necessarily positive value of the diffusion coefficient. Then $\tilde{\Omega}$ is defined by

$$\left[\frac{\omega}{\omega_2^*}\right]^2 \ln\left[\frac{1}{\tilde{\Omega}\tau}\right] = 1 ,$$

which implies

$$\tilde{\Omega} = \frac{1}{\tau(\omega)} e^{-(\omega_2^*/\omega)^2} . \qquad (32)$$

This value $\tilde{\Omega}$ can serve to define a wave-vector cutoff $\tilde{q}$ or, equivalently, a characteristic length $\xi(\omega)$ such that $\xi(\omega) = [D_0(\omega)/\tilde{\Omega}]^{1/2}$. We then obtain

$$\xi(\omega) = l(\omega) e^{(\omega_2^*/\omega)^2} . \qquad (33)$$

This length coincides with the localization length $\xi_0(\omega)$ defined by John et al.[3] for the case of a noncorrelated potential. It must be noted that $\xi(\omega)$ is much greater than $l(\omega)$ and defines a second characteristic length and a weak-localization regime for $\xi(\omega) \gg l(\omega)$ in the two-dimensional case; by contrast, in one-dimensional systems

we have only one characteristic length $\xi_0(\omega)=l(\omega)$ and only the strong localization regime occurs.[16] However, for two-dimensional films, as for the one-dimensional case, the mobility edge, defined by a diverging localization length, is obtained for $\omega=0$ only, with an essential singularity, which ensures localization for density fluctuations over the whole spectrum.

A. γ_3 anharmonic interaction

The structures of γ_4 and γ_3^2 diagrams given by Figs. 3(a) and 3(b) are the same in the two-dimensional case. Starting from the general expression for the self-energy $\Sigma_2^{(3)}(\omega)$ given by Eq. (25), one obtains for the attenuation

$$\Gamma_2^{(3)}(\omega) = \frac{1}{8}\gamma_3^2 \hbar^5 v^6 \frac{\omega^2}{\omega_D^2} \int d\omega' \frac{(\omega')^2}{\Gamma^3(\omega')D^2(\omega,\omega')}$$

$$\times n(\omega'/T) \sum_q' \frac{1}{\omega^2 + D^2 q^4} , \qquad (34)$$

where the notation $\sum_q'$ always indicates a sum over the hydrodynamic modes $ql(\omega) \ll 1$, but with the supplementary condition imposed by Eq. (33), $q > \xi^{-1}(\omega)$:

$$\sum_q' \frac{1}{\omega^2 + D^2 q^4} = \frac{\pi^2}{2D\omega} .$$

Then,

$$\Gamma_2^{(3)}(\omega) = \frac{\pi^2}{16}\gamma_3^2 \frac{\hbar^2 \omega}{\omega_D^2} \int d\omega'(\omega')^2 n(\omega'/T)$$

$$\times \left[1 - \left[\frac{\omega'}{\omega_2^*}\right]^2 \ln\left[\frac{1}{\omega\tau(\omega')}\right]\right]^{-3} . \qquad (35)$$

We can, just as before, approximate this relation by keeping only the contribution to the integral of the dominant phonons which $\hbar\omega' \sim k_B T$ to obtain finally

$$\Gamma_2^{(3)}(\omega) = \frac{\pi^2}{16}\gamma_3^2(\hbar\omega_D)\hbar\omega \left[\frac{T}{\Theta_D}\right]^3$$

$$\times \left[1 - \left[\frac{T}{T^*}\right]^2 \ln\left[\frac{1}{\omega\tau(T)}\right]\right]^{-3} , \qquad (36)$$

where

$$T^* \equiv \frac{\hbar\omega_2^*}{k_B} .$$

We can in the same way derive an expression for the correction to the density of states given generally by Eq. (23). But in the 2D case, the sum $\sum_q' Dq^2/(\omega^2 + D^2 q^4)$ reduces to $(\pi/2D)\ln(1/\omega\tau)$ because of the weak-localization condition $\xi(\omega) \gg l(\omega)$. Then with the same approximation of the dominant phonon, keeping only the most important correction, one finally has

$$\Delta_2^{(3)}(\omega) = \frac{\pi^2}{16}\gamma_3^2(\hbar\omega_D)(\hbar\omega)\left[\frac{T}{\Theta_D}\right]^3 \ln\left[\frac{1}{\omega\tau(T)}\right] , \qquad (37)$$

and the correction to the density of states is given by

$$\frac{\delta g}{g} \simeq \frac{\pi^2}{16}\gamma_3^2(\hbar\omega_D)\left[\frac{T}{\Theta_D}\right]^3 \ln\left[\frac{1}{\omega\tau(T)}\right] , \qquad (38)$$

which still provides a greater enhancement of the density of states in the regime $\omega\tau(T) \ll 1$ previously defined.

B. γ_4 anharmonic interaction

As for the three-phonon interaction, the diagram given by Fig. 4 remains relevant in the case of 2D films. But here, the specific dependence of the diffusion constant on the frequency Ω given by Eq. (13) gives rise to a strong divergence for the expressions of $\Gamma_2^{(4)}(\omega)$ and $\Delta_2^{(4)}(\omega)$.

The origin of this divergence is quite clear and comes from the peculiar structure of the γ_4 diagram. Here, the

dynamic singularity is indeed associated to the energy difference $\Omega = |\omega - \omega'|$ of phonons in the coherent backscattering situation. Then, for all energy values $\hbar\omega$ of the incident phonon, there exists a value ω' in the spectrum of the system for which $\Omega = |\omega - \omega'| \simeq \tilde{\Omega}$ leading to the divergence of $\Gamma_2^{(4)}(\omega)$ and $\Delta_2^{(4)}(\omega)$. But, it must be emphasized that this singularity is to be attributed equally to the structure of the γ_4 diagram and to the two-dimensional character of the system. Indeed, the previous expressions of $\Gamma_2^{(3)}(\omega)$ and $\Delta_2^{(3)}(\omega)$ do not produce such a singular behavior because, in this case, the energy difference Ω is fixed at the value of the incident phonon energy ω.

Now, in order for the self-energy to converge, we introduce a natural cutoff for frequencies: the inelastic width $\hbar/\tau_i(\omega,T)$ of the modes, which restricts the dynamical quantum correction to a range of frequencies between $1/\tau_i$ and $1/\tau$. This leads to the following modified expression for $D(\omega,\Omega; T)$:

$$D(\omega,\Omega; T) = D_0(\omega)\left[1 - \frac{2}{\pi g(\omega)}\sum_q' \frac{1}{-i\Omega + D_0 q^2 + \dfrac{1}{\tau_i(\omega,T)}} + \cdots\right] , \qquad (39)$$

instead of Eq. (9), where no special assumption is made on the physical nature of $\tau_i(\omega,T)$. In the limit of weak localization, $\tau_i \gg \tau$, one obtains

$$D(\omega,T) = D_0(\omega)\left[1 - \left[\frac{\omega}{\omega_2^*}\right]^2 \ln\left[\frac{\tau_i(\omega,T)}{\tau_e(\omega)}\right]\right] . \qquad (40)$$

The relation (40) will lead to a convergent expression for $\Gamma_2^{(4)}(\omega)$ and $\Delta_2^{(4)}(\omega)$ generally given by Eq. (18). We now have

$$\Gamma_2^{(4)}(\omega) = \frac{\pi(\hbar v)^4 V\omega^2}{8\Gamma(\omega)g(\omega)}\gamma_4$$

$$\times \int d\omega'\, n(\omega'/T)\frac{D^{-3}(\omega',T)}{\Gamma^2(\omega) + \hbar^2(\omega - \omega')^2} , \qquad (41)$$

where the Lorentzian can be summed to give

$$\Gamma_2^{(4)}(\omega) = \frac{\gamma_4}{32}\hbar\omega\Gamma(\omega)n(\omega/T)$$

$$\times \left[1 - \left[\frac{\omega}{\omega_2^*}\right]^2 \ln\left[\frac{\tau_i(\omega,T)}{\tau_e(\omega)}\right]\right]^{-3} . \qquad (42)$$

In the same way, we can derive for $\Delta_2^{(4)}(\omega)$ the following expression:

$$\Delta_2^{(4)}(\omega,T) \simeq \frac{\gamma_4}{8\pi}(\hbar\omega)\Gamma(\omega)n(\omega/T)\ln\left[\frac{\tau_i(\omega,T)}{\tau_e(\omega)}\right]$$

$$\times \left[1 - \left[\frac{\omega}{\omega_2^*}\right]^2 \ln\left[\frac{\tau_i(\omega,T)}{\tau_e(\omega)}\right]\right]^{-3} . \qquad (43)$$

C. Self-consistent approximation

As for the electrons case,[17,18] we can develop a self-consistent approximation and identify the inelastic scattering time and the mean lifetime of quasiparticles given by $\hbar/\Gamma_2^{(4)}(\omega,T)$. It gives at low frequency ω less than T and ω_2^*

$$\Gamma_2^{(4)}(\omega,T) = \frac{\gamma_4}{32}k_B T\Gamma(\omega) \qquad (44)$$

and

$$\Delta_2^{(4)}(\omega,T) = \frac{\gamma_4}{8\pi}k_B T\Gamma(\omega)\ln\left[\frac{T_1}{T}\right] , \qquad (45)$$

where

$$k_B T_1 \equiv \frac{32}{\gamma_4} .$$

Then the relative variation of the density of states is proportional to

$$\frac{\delta g}{g} \propto \omega^2 T\ln\left[\frac{T_1}{T}\right] , \qquad (46)$$

an expression which recalls the corrections of the electronic density of states in the weak-localization regime for interacting fermions.[17,18]

V. APPLICATIONS TO LOW-TEMPERATURE THERMAL PROPERTIES OF THREE-DIMENSIONAL GLASSES

We have seen that the elastic relaxation time or Rayleigh-Klemens time plays a crucial role in the theory of weak localization. Therefore, it is natural to turn towards the situation where the harmonic disorder is the strongest: the glasses. This situation has already been discussed in a classical article by Zeller and Pohl[19] where

the Rayleigh-Klemens mechanism was used to explain the plateau of the thermal conductivity. In this paper, they model a glass as a crystal where each atom can be considered as a mass defect. In such a case $\sigma_M^2 = 1$. By using the classical approximation of the dominant phonon, they were able to explain both the magnitude and the temperature dependence of the "kinetic" mean free path derived from the gas kinetic expression of the thermal conductivity: $K = \frac{1}{3} Cvl$. If the value $\sigma_M^2 = 1$ seems justified for the case of glasses, the association of the dominant phonon approximation with the gas kinetic thermal conductivity is misleading for a strong frequency dependence of relaxation time. Here we will retain from the basic work of Zeller and Pohl the limit value $\sigma_M^2 = 1$ for glasses. It also must be indicated at this point in the discussion that the two-level systems[20] cannot play the role of elastic scatterers we had considered previously. We will neglect them for the present analysis.

The present theory of phonon weak localization and anharmonicity provides two sets of expression for the attenuation and the excess spectral density contained in relations (22), (24), (29), and (30). These relations are frequency and temperature dependent and involve the threshold frequency ω_3^* or threshold temperature T^* at which a divergence occurs. We will consider only regimes of low frequency $\omega < \omega_3^*$ and low temperature $T < T^*$.

The expressions involving γ_4 are valid for any frequency or temperature, especially for thermal phonons where ω is of the order of T and can be applied to the analysis of the thermal conductivity. The expression (22) of $\Gamma_3^{(4)}$ can be compared to the Rayleigh-Klemens scattering Γ by forming the ratio $\Gamma_3^{(4)}/\Gamma$. The known expressions for the anharmonic coupling constants are

$$\gamma_4 \simeq \gamma_3^2 \simeq \frac{\gamma_G^2}{Mv^2} \sim \frac{1}{\alpha \Theta_D} ,$$ (47)

where γ_G is the Grüneisen parameter ($\simeq 1$) and M is the mass of the atom. The parameter α is typically of the order of 10 for a canonical glasslike vitreous silica. One obtains

$$\frac{\Gamma_3^{(4)}}{\Gamma} \simeq \frac{\sigma_M^2}{\alpha} \frac{\omega^4}{\Theta_D^4} \frac{1}{[1-(\omega/\omega_3^*)^6]^{7/2}} n\left[\frac{\omega}{T}\right]$$ (48)

(ω and T in same units), which shows that $\Gamma_3^{(4)} \ll \Gamma$ in the regime $\omega, T \ll \omega_3^*$. It is only in the range of $\omega \lesssim \omega_3^*$ and $T \lesssim T^*$ that $\Gamma_3^{(4)}$ can be more efficient than Γ. For the excess density of modes we find

$$\frac{\delta g}{g} \simeq \frac{\sigma_M^2}{\alpha} \left[\frac{\omega}{\omega_D}\right]^7 n(\omega/T) \frac{1}{[1-(\omega/\omega_3^*)^6]^{7/2}} .$$ (49)

The very strong frequency dependence comes from the Rayleigh scattering, and here, as for the lifetime, the excess density is negligible except in the vicinity of ω_3^* and T^*.

A similar conclusion can be achieved for the γ_3^2 interaction which applies only to low-frequency phonons as compared to the temperature. The expressions (29) and (30) of $\Gamma_3^{(3)}$ and $\Lambda_3^{(3)}$ are candidates for predicting the sound at-

tenuation and spectral density for hypersound phonons. Naturally we are led to compare $\Gamma_3^{(3)}$ to the Landau-Rumer[5,6] expression:

$$\Gamma_{LR} = \gamma_3^2 \omega T (T/\Theta_D)^3 .$$ (50)

By forming the ratio $\Gamma_3^{(3)}/\Gamma_{LR}$, the γ_3^2 coupling constant is eliminated and gives

$$\frac{\Gamma_3^{(3)}}{\Gamma_{LR}} \simeq \sigma_M^2 \left[\frac{\omega}{\omega_D}\right]^{1/2} \frac{T}{\Theta} \frac{1}{[1-(T/T^*)^6]^{7/2}} .$$

The value of this ratio is less than 1 except when T approaches T^* where the divergence occurs.

A similar result is obtained for the excess spectral density for the γ_3^2 interaction:

$$\frac{\delta g}{g} = \frac{3\pi}{2\alpha} \sigma_M^2 \left[\frac{T}{\Theta_D}\right]^7 \frac{1}{[1-(T/T^*)^6]^{7/2}} ,$$

the contribution of which becomes sizable only when T tends towards T^*. Actually, the general conclusion of this analysis is that a strong enhancement of the excess density of states as well as the attenuation is predicted when ω and T approach $\omega_3^* = k_B T^*/\hbar$. This prediction must be compared to the experiments on glasses which exhibit a plateau in thermal conduction accompanied by a bump in the specific heat around 10 K in all glasses which have been tested.[7] The standard analysis of these features exhibits, therefore, an abrupt decrease of the phonon mean free path and, at the same time, a strong excess of the spectral density. It is tempting to apply the previous conclusions to this situation by identifying the critical frequency ω_3^* or temperature T^* with the temperature of occurrence of the plateau in the thermal conduction namely $T^* \simeq 10$ K. From the expression (12), we can derive a value of ω_3^* less than ω_D: $\omega_3^* = 0.51\omega_D$ for $\sigma_M^2 = 1$. This value is very high, typically above 100 K, and cannot be applied directly to the case of glasses. We think that it is a consequence of the approximation of weak localization which cannot produce a good value of the threshold of localization. We believe that higher-order contributions to the diagram expansion as well as more realistic spectral density could result in a lower value of ω_3^*. If ω_3^* is considered as a parameter which fits, the present theory of weak localization is able to describe the qualitative features of glasses, namely, the occurrence of a plateau in the thermal conduction and, simultaneously, an excess specific heat at low temperature. At $T = 0$ K the present model does not predict any localization for the single-mode problem. As a matter of fact, the mean lifetime shortening, as well as the spectral density contribution, vanish either exponentially or as a power law for both the γ_4 and γ_3^2 interactions. But the two-phonon correlator contributing to the diffusion constant is localized when ω approaches ω_3^*. This prediction does not contradict the recent numerical study of Nagel et al.[21] which was performed at $T = 0$ K on a three-dimensional model of glass where atoms interact via 6-12 potential. They only observed a localization of vibration modes at high frequency near the cutoff. Our analysis leads to a situation of localization for "second sound" at

critical frequency which is reflected in the first sound via the anharmonic coupling. A finite temperature is necessary in order to observe localization in the one-phonon problem.

As a final remark on the application of the localization in glasses, let us specify the domain of validity of the present model. The energy of phonons is limited by ω_3^* at high frequency in order to satisfy the condition of perturbation expansion incident plane waves instead of localized modes. A supplementary condition is that the inelastic collision frequency must be less efficient than the elastic or the Rayleigh-Klemens relaxation. This is true for the anharmonic interaction as reported by expression (48). In glasses, however, at low frequency and low temperature, we know that two-level systems are present and inelastically scatter the phonons. More precisely it is accepted[19,20] that below 1 K, the two-level systems are more efficient than mass defects for scattering phonons. Actually, the domain of validity of the weak localization is reduced in the range of temperature or frequency between 1 and 10 K where the mean free path drops abruptly to interatomic distances.

VI. SUMMARY

Let us summarize the main steps of the transposition of the weak-localization theory from electrons to phonons.

(1) Quantum corrections to diffusion are obtained by the resummation technique of the maximally crossed diagrams. A pole of diffusion in the effective vertex interaction subsists, despite the change in statistics, from fermions to bosons and in dispersion from quadratic to linear.

(2) The strong energy dependence of the elastic Rayleigh-Klemens scattering produces a threshold energy ω_3^* in the renormalized diffusion constant in three dimensions. At this critical value ω_3^*, the two-phonon density fluctuations—or second sound—become localized.

(3) In two dimensions the divergence at low frequency, $\Omega \rightarrow 0$, of the dynamical quantum correction indicates the general localization effect in low dimensionality ($d \leq 2$).

(4) The anharmonic interactions couple the one-phonon propagator to the phonon density fluctuations. Phonon self-energies are calculated for two basic anharmonic interactions and diverge when ω or T approach the critical energy ω_3^*. Strong shortening of the phonon mean free path and additional spectral density reflect the approach to this localization threshold.

(5) In the two-dimensional case, a low-frequency cutoff (the inelastic rate of collisions) is indispensable to ensure the convergence of the quantum correction when one kind of anharmonic interaction is switched on. The resulting phonon self-energy is quite similar to the electronic one.

(6) A tentative application of the previous concepts to glasses is developed: Qualitative features of low-temperature anomalies of glasses—plateau in thermal conduction and bump in excess specific heat—are understandable in the framework of weak localization providing that the value of the phonon threshold energy is reduced to around 10 K.

APPENDIX A: DERIVATION OF THE EFFECTIVE INTERACTION U OF PHONONS

In this appendix we would like to sketch the expansion of Green's functions in order to obtain the expressions (4) and (5) of the kinetic equation for $G_2(\mathbf{k},\mathbf{k}',\omega,\Omega)$ and the effective U interaction between interfering phonons leading to the coherent backscattering effect. First, if $\hat{H} = \hat{H}_0 + \hat{H}_{\text{des}}$ represents the Hamiltonian for the phonon gas moving in the field of randomly fixed impurities, we can write the usual development for the Green's operator $\hat{D}(\omega)$:

$$\hat{D}(\omega) \equiv \hat{D}_0(\omega) + \hat{D}_0(\omega)\hat{t}(\omega)\hat{D}(\omega) , \qquad (A1)$$

where

$$\hat{D}_0(\omega) = (\hbar\omega\hat{I} - \hat{H}_0)^{-1}$$

and where

$$\hat{t}(\omega) = \hat{H}_{\text{des}}(\hbar\omega\hat{I} - \hat{H}_0 - \hat{H}_{\text{des}})^{-1}(\hbar\omega\hat{I} - \hat{H}_0)$$

represents the collision operator. Equation (A1) can be written more explicitly in the form

$$\hat{D}(\omega) = \hat{D}_0(\omega) + \sum_n \hat{D}_0(\omega)\hat{t}(\omega,\mathbf{X}_n)\hat{D}_0(\omega)$$

$$+ \sum_{\substack{m,n \\ m \neq n}} \hat{D}_0(\omega)\hat{t}(\omega,\mathbf{X}_n)\hat{D}_0(\omega)\hat{t}(\omega,\mathbf{X}_m)\hat{D}_0(\omega) + \cdots ,$$

$$(A2)$$

where $\{\mathbf{X}_n\}$ represents the impurities' positions.

From Eq. (A2), we can write

$$G_2(\mathbf{k},\mathbf{k}'; \omega,\Omega) = \lim_{\epsilon \rightarrow 0} \lim_{\delta \rightarrow 0} \langle G_{\mathbf{k}}(\omega + \Omega + i\delta)G_{\mathbf{k}'}(\omega - i\epsilon) \rangle$$

in the form

$$G_2(\mathbf{k},\mathbf{k}'; \omega,\Omega) = \delta_{\mathbf{k},\mathbf{k}'}G_{\mathbf{k}}^A(\omega)G_{\mathbf{k}}^R(\omega + \Omega)$$

$$+ G_{\mathbf{k}}^A(\omega)G_{\mathbf{k}}^R(\omega + \Omega)\Gamma_{\mathbf{k},\mathbf{k}'}(\Omega)$$

$$\times G_{\mathbf{k}'}^A(\omega)G_{\mathbf{k}'}^R(\omega + \Omega) , \qquad (A3)$$
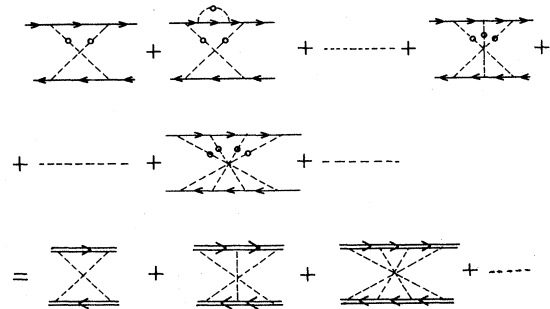

FIG. 6. Resummation of the most divergent diagrams in the hydrodynamic limit $ql(\omega) \ll 1$. The simple lines $\rightarrow$ represent the free propagator and the double lines $\Rightarrow$ the renormalized phonon propagator.

where the vertex function $\Gamma_{k,k'}(\Omega)$ describes the interaction of phonons with impurity centers as shown in Fig. 1. Now, we can express $\Gamma_{k,k'}(\Omega)$ as the sum of two terms, one of them, $U_{k,k'}(\Omega)$, giving the sum of irreducible diagrams. Then we have

$$\Gamma_{k,k'}(\Omega) = U_{k,k'}(\Omega) + \sum_{k_1} U_{k,k_1}(\Omega)\Gamma_{k_1,k'}(\Omega)$$

$$\times G_{k_1}^A(\omega)G_{k_1}^R(\omega+\Omega) , \qquad (A4)$$

and (A3) and (A4) lead to

$$G_2(\mathbf{k},\mathbf{k}';\omega,\Omega) = \delta_{\mathbf{k},\mathbf{k}'}G_k^A(\omega)G_k^R(\omega+\Omega) + G_k^A(\omega)G_k^R(\omega+\Omega)G_{k'}^A(\omega)G_{k'}^R(\omega+\Omega)$$

$$\times \left[U_{k,k'}(\Omega) + \sum_{k_1} U_{k,k_1}(\Omega)G_{k_1}^A(\omega)G_{k_1}^R(\omega+\Omega)\Gamma_{k_1,k'}(\Omega) \right] ,$$

which implies

$$G_2(\mathbf{k},\mathbf{k}';\omega,\Omega) = \delta_{\mathbf{k},\mathbf{k}'}G_k^A(\omega)G_k^R(\omega+\Omega) + G_k^A(\omega)G_k^R(\omega+\Omega)U_{k,k'}(\Omega)G_{k'}^A(\omega)G_{k'}^R(\omega+\Omega)$$

$$+ G_k^A(\omega)G_k^R(\omega+\Omega)\sum_{k_1} U_{k,k_1}(\Omega)G_{k'}^A(\omega)G_{k'}^R(\omega+\Omega)\Gamma_{k_1,k'}(\Omega)G_{k_1}^A(\omega)G_{k_1}^R(\omega+\Omega) ,$$

and from Eq. (A3) we finally get

$$G_2(\mathbf{k},\mathbf{k}';\omega,\Omega) = G_k^A(\omega)G_k^R(\omega+\Omega)\left[\delta_{\mathbf{k},\mathbf{k}'} + \sum_{k_1} U_{k,k_1}(\Omega)G_2(\mathbf{k}_1,\mathbf{k}';\omega,\Omega)\right] ,$$

which coincides with Eq. (4).

The effective interaction between phonons is then defined by the sum of irreducible diagrams. The most divergent set of these diagrams, as shown by Langer and Neal,[4] is given by the maximally crossed diagrams and can be resummed (cf. Fig. 6) to give $U_{k,k'}(\Omega)$ as the sum of a geometric series

$$U_{k,k'}(\Omega) = \frac{b_d^2 I_d(\omega,\Omega;q)}{1 - b_d I_d(\omega,\Omega;q)} , \qquad (A5)$$

where

$$I_d(\omega,\Omega;q) \equiv \sum_k G_k^A(\omega)G_{k-q}^R(\omega+\Omega) ,$$

and b_d is given by the Fermi golden rule,

$$b_d = \frac{\hbar}{\pi}\frac{\Gamma_d(\omega)}{g_d(\omega)} . \qquad (A6)$$

In this last expression, $g_d(\omega)$ is the d-dimensional density of states and $\Gamma_d(\omega)$ is the inverse Rayleigh-Klemens lifetime proportional to ω^{d+1}.

We would like now to have a perturbative expression in powers of Ω and q^2 (where we recall that $\mathbf{k}+\mathbf{k}'=\mathbf{q}$) in the limit $ql(\omega)\ll 1$ and $\Omega\tau(\omega)\ll 1$ of the product $G_k^A(\omega)G_{k-q}^R(\omega+\Omega)$. First, we write

$$G_{k-q}^R(\omega+\Omega)G_k^A(\omega) = \frac{G_{k-q}^R(\omega+\Omega) - G_k^A(\omega)}{[G_k^A(\omega)]^{-1} - [G_{k-q}^R(\omega+\Omega)]^{-1}} ,$$

where the Green's functions for phonons in the field on impurities are defined by

$$G_k^A(\omega) = \frac{1}{\hbar[\omega - \omega(k)] + i\Gamma_d(\omega)}$$

and

$$G_{k-q}^R(\omega+\Omega) = \frac{1}{\hbar[\omega+\Omega - \omega(k-q)] - i\Gamma_d(\omega+\Omega)} .$$

We prefer these definitions of the phonon propagators to those generally given[22] because of the fundamental role played by time reversal in the constructive interference effect between phonons. On the other hand, the acoustic approximation for the dispersion relation allows us to write

$$[G_k^A(\omega)]^{-1} - [G_{k-q}^R(\omega+\Omega)]^{-1}$$

$$= \hbar\left[\frac{2i}{\hbar}\Gamma_d(\omega) + \Omega\left[1 + \frac{2i\Gamma_d(\omega)}{\hbar\omega}\right] + \frac{vq^2}{2k} + v\frac{\mathbf{k}\cdot\mathbf{q}}{k}\right] ,$$

and the denominator of Eq. (A5) with the help of Eq. (A6) reads

$$1 - b_d I_d(\omega,\Omega;q) = \frac{b_d}{2i\Gamma_d(\omega)}\left[-\frac{\hbar\Omega}{2i\Gamma_d(\omega)}\frac{2i\pi}{\hbar}g_d(\omega) + \frac{v^2\hbar^2 q^2}{4\Gamma_d^2(\omega)}\frac{2i\pi}{\hbar}g_d(\omega) + \frac{\hbar v q^2}{2i\Gamma_d(\omega)}\sum_k \frac{1}{k}[G_k^A(\omega) - G_k^R(\omega)]\right] , \qquad (A7)$$

where the term proportional to q cancels because it appears only in the sum

$$\sum_k k^{-1}\mathbf{k}\cdot\mathbf{q} \propto q\int_0^\pi \cos\theta\,d\theta ,$$

where $\theta = \arccos(\mathbf{k}\cdot\mathbf{q}/kq)$. In Eq. (A7) the last term proportional to $\Gamma^{-1}(\omega)$ is negligible because the Rayleigh-Klemens expression for $\Gamma(\omega)$ is proportional to n, where n is the impurity density assumed to be very small and then $\Gamma^{-2}(\omega)\gg\Gamma^{-1}(\omega)$. Finally, we get

$$1-b_d I_d(\omega,\Omega,q)=\frac{\hbar}{2\Gamma_d(\omega)}\left[-i\Omega+\frac{\hbar v^2}{\Gamma_d(\omega)}q^2\right]$$

and

$$U_{k,k'}(\Omega)=\frac{2v^4}{\pi D_0^2(\omega)}\frac{\hbar^2}{g_d(\omega)}\frac{1}{-i\Omega+D_0(\omega)q^2} , \qquad (A8)$$

where $D_0(\omega)$, defined by $D_0(\omega)=\hbar v^2/\Gamma_d(\omega)$, is the single-phonon diffusion constant. Equation (A8) is the desired Eq. (5) given in the text.

APPENDIX B: SELF-ENERGY $\Sigma^{(4)}(\omega)$ ASSOCIATED WITH THE FOUR-PHONON ANHARMONIC PROCESSES AND THE $\widetilde{U}$ INTERACTION

Starting from the general relation given by

$$\Sigma_3^{(4)}(\omega)=k_B T\frac{V}{g(\omega)}\sum_{k,k'}\sum_{q,p}V^{(4)}(k,-k,k+q,-k-q)\langle D^R_{-k-q}(\omega_p)\delta(\omega-\omega(k))\rangle\delta_{k',-k-q} ,$$

we can begin to calculate the sum S defined by

$$S=\frac{\hbar^2}{4}\gamma_4\sum_k\omega^2(k)D_k^R(\omega_p)D_k^A(\omega_p)D_k^R(\omega)D_k^A(\omega) , \qquad (B1)$$

where the renormalized propagators in the presence of impurities can be expressed by

$$D_k^{R,A}(\omega)=\frac{1}{\hbar[\omega-\omega(k)]\pm i\Gamma_d(\omega)} ,$$

in which $\Gamma_d(\omega)=\hbar v^2/dD_0(\omega)$ is the Rayleigh width of the modes proportional to the impurity density, assumed to be small. Then Eq. (B1) can be replaced by

$$S=\frac{\hbar^2}{4}\gamma_4\frac{1}{\Gamma_d^2(\omega)}\frac{\pi}{\hbar}\int d\omega''(\omega'')^2 g(\omega'')\delta(\omega-\omega'')$$

$$\times\frac{\Gamma(\omega)}{\Gamma^2(\omega)+\hbar^2(\omega'-\omega'')^2} ,$$

and finally we get for $d=3$

$$S=\frac{\hbar}{4}\gamma_4\frac{\pi}{\Gamma(\omega)}\frac{\omega^2 g(\omega)}{\Gamma^2(\omega)+\hbar^2(\omega'-\omega)^2} . \qquad (B2)$$

Equation (B2) combined with the expression given by Eq. (6) for the effective interaction $\widetilde{U}$ leads for $\Sigma_3^{(4)}(\omega)$ to the expression

$$\Sigma_3^{(4)}(\omega)=V\frac{(\hbar v)^4\omega^2}{4\pi\Gamma(\omega)g(\omega)}\gamma_4\int d\omega'$$

$$\times\sum_q n(\omega'/T)\frac{D^{-2}(\omega,\omega')}{-i(\omega'-\omega)+D(\omega,\omega')q^2}$$

$$\times\frac{1}{\Gamma^2(\omega)+\hbar^2(\omega'-\omega)^2}$$

which is the Eq. (18) given in the text for a three-dimensional system.

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