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Quasidiffusion of phonons in Si

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We show that frequency-down conversion and elastic scattering treated as quasidiffusion can explain quantitatively the recent experimental results by Shields *et al.* and Monte Carlo simulations by Tamura. The ballistic component is demonstrated to be negligible in forming the exponential decay of the phonon signal observed experimentally. The decay is governed by the special bottleneck at some frequency $\bar{\omega}$ which depends upon the boundary conditions and belongs to the quasidiffusive region of the phonon spectrum. The bottleneck frequency separates the phonons which decay ($\omega > \bar{\omega}$) and those which diffuse to the detector ($\omega < \bar{\omega}$).

We still lack a quantitative comparison between experiment, simulations, and analytic work on phonon propagation in semiconductors. This work is devoted to one of the most interesting modes of propagation—quasidiffusion—which it is necessary to understand for applications, in particular for particle detectors.[1] Quasidiffusion is somewhat analogous to cosmic showers: phonons scatter elastically and multiply by frequency-down conversion. The cascade slows down with time and therefore quickly enters the similarity stage[2] which has been studied by Kazakovtsev and Levinson.[3] The frequency-down conversion, or anharmonic decay, occurs with the rate $\tau_a^{-1}(\omega)$, and the elastic scattering with the rate $\tau_s^{-1}(\omega)$ (the latter is larger for Si). The similarity solution implies that a characteristic frequency can be found from $\tau_a(\omega)=t$ which gives $\omega(t)$, and the space expansion is on the order of the lifetime diffusion length $l(t)\sim s[\tau_a(\omega(t))\tau_s(\omega(t))]^{1/2}$. For low-frequency phonons the conventional approximation is $\tau_a^{-1}(\omega)\sim\omega^5$, $\tau_s^{-1}(\omega)\sim\omega^4$, and we get $l(t)\sim t^{9/10}$. Such sublinear expansion was observed by Bron, Levinson, and O'Connor in heavily doped Al_2O_3.[4] The phonon signal had a diffusive shape with a maximum at some intermediate time.

However, attempts to find quasidiffusion in classic semiconductors, for example in Si, have met with significant difficulties.[5] Liquid He, if it is present close to the excitation point, quickly absorbs most of the phonons and the time trace consists of ballistic phonons which escaped at the very beginning of the cascade.[6] If liquid He is eliminated from the vicinity of the excitation point by means of a special seal[6] then the input power should be quite low in order not to excite electron-hole droplets;[6,7] at these powers the phonon signal is weak and data accumulation is required. The phonon signal has its maximum at about the TA-phonon ballistic time, t_b, and is followed by an exponential tail with the relaxation constant $3.6t_b$ for the sample size used in Refs. 6 and 8. The average travel duration is then approximately $4.6t_b$, and it has been unclear to what extent quasidiffusion is applicable. Monte Carlo (MC) simulations have been done by Tamura, which showed the exponential behavior and allowed a fit of the relaxation constant $3.6t_b$ under specific choice of decay parameters[8–10] when the decay of both transverse modes is not allowed.

This work is devoted to analytic explanation of the signal shape. We first discuss the diffusion approximation for a finite sample. After that it is shown that a one-branch model of the phonon spectrum is effective if the elastic scattering is quick enough. The kinetic equation for this model leads to an integral equation for the first eigenfunction. The relaxation constant is the smallest eigenvalue and can be found analytically.

At low enough frequencies phonons can travel ballistically through the sample. Such phonons are not described within the diffusion approximation and require special care. The ballistic phonons escape from the sample quickly, in time t_b. Since the signal relaxation is prolonged, the bottleneck of the process is *not* due to the escape of ballistic phonons. In order to find the bottleneck we need to consider higher energies, where escape becomes comparable to the anharmonic decay. In this case, the frequency appears to be high enough to justify the diffusion approximation.

When particles diffuse in a given region with absorbing boundaries, their concentration at late times decrease exponentially. For the case of a sphere of radius L the relaxation constant is

$$\tau_L^{-1}=\pi^2 D/L^2 \tag{1}$$

and for a rectangular parallelepiped with half-sides L_x, L_y, L_z

$$\tau_L^{-1}=\frac{\pi^2 D}{4}\left(\frac{1}{L_x^2}+\frac{1}{L_y^2}+\frac{1}{L_z^2}\right). \tag{2}$$

Here D is the diffusivity.

Let us consider the elastic scattering and mode conversion. In Si elastic-scattering conversion between LA and TA phonons distributes the phonons among branches in accordance with their density of states (0.531:0.376:0.093 for slow, fast transverse and longitudinal phonons). Given the value of $\tau_s^{-1}(\omega)=2.43\times10^{-42}(\omega/2\pi)^4$ s^3 (Ref. 11) and the value of the anharmonic decay rate $\tau_{a,L}^{-1}(\omega)=1.2-1.8\times10^{-55}(\omega/2\pi)^5$ s^4 (Refs. 9 and 12–14),

$\tau_{a,FT}^{-1}(\omega)=0.66\times10^{-55}(\omega/2\pi)^5$ s^4 (Refs. 12–14) with the mode-averaged rate $0.41\times10^{-55}(\omega/2\pi)^5$ s^4, one can check that for phonons with frequency $\omega/2\pi<2.5$ THz, the elastic mode conversion dominates. This implies that equilibration between modes proceeds faster than frequency-down conversion, and that the one-effective-branch model is a reasonable approximation. However, the precision of this approximation may be insufficient at some level. We note that the direction of motion and mode of a phonon before and after the isotope scattering are correlated,[11] and the process becomes non-Markovian. Three distinct phonon modes have to be considered and the anisotropy of the anharmonic decay[12,14] is to be taken into account. Thus the simulation of the full Boltzmann equation can no longer be avoided. Note that *all* the mentioned complications are controlled by the same paramener $s\tau_s/L$ and should be taken into account simultaneously.

Summarizing what has been written, we arrive at the kinetic equation (cf. Ref. 3)

$$\left[\frac{\partial}{\partial t}-D(\omega)\nabla^2+\frac{1}{\tau_a(\omega)}\right]N(\omega,\mathbf{r},t)=\int_\omega^\infty d\omega'\rho(\omega')N(\omega,\mathbf{r},t)P(\omega'\to\omega)\ , \quad (3)$$

where $D(\omega)=\langle(\mathbf{sn})^2\rangle\tau_s(\omega)$ is the averaged diffusivity, $P(\omega\to\omega')$ describes averaged anharmonic decay distribution, which is normalized as

$$\int_0^\omega d\omega'\rho(\omega')\omega'P(\omega\to\omega')=\omega/\tau_a(\omega)\ . \quad (4)$$

$\rho(\omega)$ is the combined density of states, and $\tau_a^{-1}(\omega)$ is the mode-averaged decay rate. The diffusivity is diagonal (we assume the symmetry of a cubic crystal) and is calculated with the help of mode- and angle-averaged square projection of the phonon group velocity, $\mathbf{n}$ parallel to one of the main axes is the vector. For Si the numerical value of $\langle(\mathbf{sn})^2\rangle$ can be obtained by using Every formulas[15] and is equal to $\langle(\mathbf{sn})^2\rangle=1.16\times10^{11}$ cm^2/s^2.

Equation (3) is considered in a finite sample with the boundary condition $N(\omega,L,t)=0$ meaning that phonons that have escaped from the sample into liquid He cannot return. The longest relaxation is experienced by the first Fourier mode [$r^{-1}\sin(\pi r/L)$ in the case of a sphere], and its amplitude $N_1(\omega,t)$ obeys the integral equation

$$\left[\frac{\partial}{\partial t}+\frac{1}{\tau_a(\omega)}-\frac{1}{\tau_L(\omega)}\right]N_1(\omega,t)=\int_\omega^\infty d\omega'\rho(\omega')N_1(\omega,t)P(\omega'\to\omega)\ . \quad (5)$$

It is now easy to get the value of the phonon signal at late times. We shall give the formulas for the case of sphere (1); the obvious modifications are to be made in other geometries. The signal

$$S(t)=\int_0^\infty d\omega\,\rho(\omega)\omega N_1(\omega,t)\tau_L^{-1}(\omega) \quad (6)$$

has the same exponential time dependence as the slowest decay of $N_1(\omega,t)$, i.e., e^{-t/t_0},

$$t_0^{-1}=\min_\omega\left[\frac{1}{\tau_a(\omega)}+\frac{1}{\tau_L(\omega)}\right]\ . \quad (7)$$

The two terms in (7) are proportional to ω^5 and ω^{-4}, respectively, and their sum has a minimum at

$$\bar\omega=\beta_1\omega_0\left[\frac{\pi^2\langle(\mathbf{sn})^2\rangle\tau_s(\omega_0)\tau_a(\omega_0)}{L^2}\right]^{1/9}\ ; \quad (8)$$

this expression is independent upon the arbitrary frequency ω_0. The numerical factor β_1 is equal to $(\frac{4}{5})^{1/9}$. For the slowest rate t_0^{-1} we get

$$\frac{t_0}{t_b}=\frac{1}{\beta\pi^{10/9}}\left[\frac{\tau_a^4(\omega_0)}{\tau_s^5(\omega_0)}\right]^{1/9}\left[\frac{L^{1/9}s_{FT}}{\langle(\mathbf{sn})^2\rangle^{5/9}}\right]\ , \quad (9)$$

with $\beta=(\beta_1^5+\beta_1^{-4})^{-1}=1.9877$, and s_{FT} is the velocity of the prompt phonons. A formula related to (9) namely, $t_0/t_b\sim(\tau_a^4L/\tau_s^5s)^{1/9}$ can be found in Ref. 3. As expected, the bottleneck is situated quite high on the frequency scale, higher than the mean frequency of the detected phonons.

Let us discuss the validity of Eq. (9). We have not specified the form of the scattering distribution $P(\omega\to\omega')$, which may be of importance. Indeed, if the majority of phonons down-convert missing the frequency ω, the relaxation constant t_0 would be smaller. Such a phenomenon may occur if the source in Eq. (3) is monochromatic and the function $P(\omega\to\omega')$ is strongly peaked at, say, $\omega'=\omega/2$. It is convenient to choose the parameter β entering (9) to be dependent upon the selected form of $P(\omega\to\omega')$ and therefore upon the source frequency ω^*. To investigate this dependence we solved Eq. (5) numerically and investigated the "apparent" value of β as measured over experimentally accessible times of order of $1-2t_0$. We found that while the δ-functional $P(\omega\to\omega')$ indeed results in oscillations of β with the logarithm of source frequency (period is ln2, and $2<\beta<4$), the other more realistic profiles of $P(\omega\to\omega')\sim\omega'^n(\omega-\omega')^n$, $2\le n\le8$, provide less sensitive values of β. We think that the special computed profile by Tamura[9] should also belong to this broad class. To conclude, we do not expect an error of more than 5% comparing the simple formula (9) and the measured "apparent" relaxation constant. This precision is comparable with the experimental error reported in Refs. 6 and 8.

Tamura has compared our Eq. (9) with the special one-branch MC simulations[9,10] where the Debye sound velocity was used and found that for a 0.55-cm spherical sample and $0.12\times10^{-55}(\omega/2\pi)^5$ s^4 effective decay rate Eq. (9) gives $t_0=3.01\ t_b$, while the MC result is $t_0=3.1\ t_b$. If there is indeed a discrepancy, then in addition to the above-mentioned influence of the $P(\omega\to\omega')$ form factor and correlations in elastic scattering one may attribute it to the fact that Eq. (3) possesses a whole set of time-dependent relaxation modes among which (7) is the slowest. However, the mode spectrum is continuous and we deal with an integral of the type $\int d\omega\exp[-(\omega^5+\omega^4)t]$, which gives a power-law prefactor $\sim t^{-1/2}$.

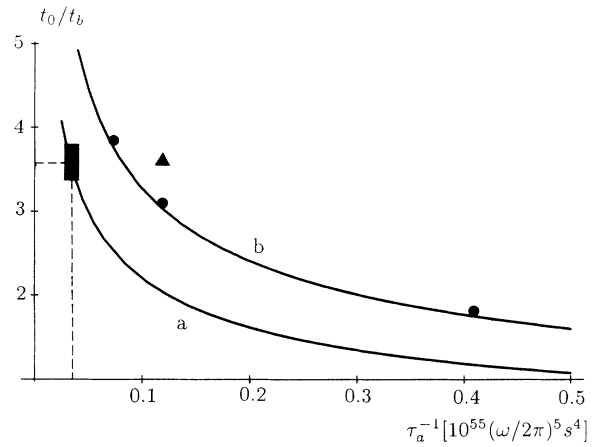

FIG. 1. Dependence of the relaxation constant t_0 in units of t_b upon the strength of the mode-averaged anharmonic decay rate. Curve (a) is computed using Eqs. (2) and (7), the extra factor 1.41, and the parameters given in the text for the experimental sample (Ref. 6). The experimental value of t_0 and the corresponding estimates for the decay rate are depicted by the rectangle to show the experimental error. Curve (b) is computed using Eq. (1) for a sphere of radius 0.55 cm to make the comparison with MC simulations. Circles are the MC results by Tamura (Refs. 9 and 10) for the branch-averaged model, and the triangle corresponds to three branches with angle-averaged anharmonic decay (Refs. 8–10).

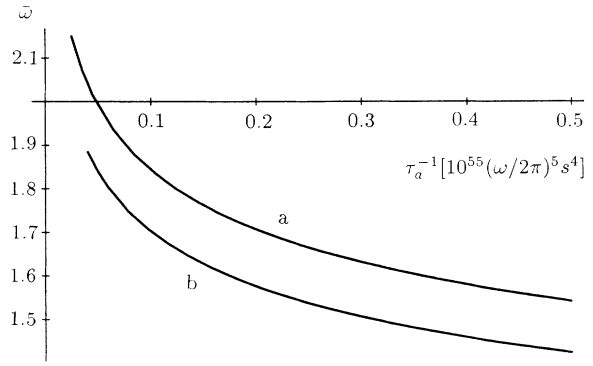

FIG. 2. The bottleneck frequences for the two curves shown in Fig. 1. For the notations and the parameters see Fig. 1.

Having established an agreement between analytical and numerical treatments we now return to the experiment. The sample used in Refs. 6 and 8 is a parallelipiped with dimensions $0.55\times0.6\times1.0$ (in cm), one side of which (0.6×1.0 side) was sealed from liquid He. If the entire side had been sealed we might have used Eq. (2) with parameters $L_x=0.55$ cm, $L_y=0.3$ cm, $L_z=0.5$ cm. However, the seal had a circular shape with diameter of 0.3 cm and was attached to the center of the sample side.[16] We have solved the model diffusion problem with such a boundary condition and found that the decay rate (2) acquires an additional factor of 1.41. We can see in Fig. 1 that the reported decay rates are not in accord with experiment, and we would rather suggest the value of $(0.036\pm0.01)\times10^{-55}(\omega/2\pi)^5$ s^4 as an effective decay rate. Figure 2 shows the relevant bottleneck frequences. One may see that the frequencies are high enough to question the low-frequency approximation. We note that in addition the phonon spectrum at 2 THz may deviate by more than 25% from the low-frequency linear dispersion.[17] Apparently, to improve significantly the precision of the result (9) one has to take into account many factors. For example, Tamura has found that including the three phonon branches while keeping the isotropic approximation for the anharmonic decay may give a change of 20%,[9,10] see Fig. 1. From the experimental point of view the bottleneck-frequency $\bar{\omega}$ dependence (8) upon the size of the sample $\bar{\omega}\sim L^{-2/9}$ provides some opportunity to access the effective decay rates at different frequences, and measure corrections to $\tau_a^{-1}\sim\omega^5$. To give an extreme example, we note that if, say, this fifth power is to be replaced by the fourth, ω^4, there would be no size dependence of t_0/t_b; such a regime has been suggested for GaAs,[18] where mode conversion is slow.

To conclude, we have found a quantitative agreement between the one-branch quasidiffusion model by Kazakovtsev and Levinson as applied to a finite-geometry boundary-value problem and MC simulations of the same process. The geometry of the experiment is important and can be used to check the model. To fit the relaxation constant $3.6t_b$ we have to use the anharmonic decay rate $(0.036\pm0.01)\times10^{-55}(\omega/2\pi)^5$ s^4, which is three times less than the value used by Tamura,[8–10] and ten times less than the value by Berke, Mayer, and Wenner.[12,13] Along with the mentioned factors which can influence this estimate, one has to take into account finite reflectivity of the Si-He interface.

The general picture of three frequency regions of decaying, diffusing, and ballistically moving phonons, discussed by Maris[13] applies to the theory presented. It appears that the slowest phonon path in phase space does not go through the very last region of ballistic motion. This allows us to show that the tail of the phonon signal is due to quasidiffusive phonons.

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