Phonon Propagation by Quasidiffusion

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Experimental observation of quasidiffusive propagation of a nonequilibrium phonon distribution in Al_2O_3:V is reported. Among the features of this propagation mode is the fact that the arrival time of the phonon pulse front varies almost linearly with propagation distance as expected for ballistic transport, even though the time-of-flight spectrum has an extended tail characteristic of diffusive transport.

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We report the first direct experimental observation of quasidiffusional propagation of nonequilibrium phonons in solids.

Phonon transport is said to be "diffusive" if the time of arrival, t_A, for a phonon packet to traverse L is proportional to L^2/D, where D is the diffusion constant. Such propagation is normally observed in the presence of elastic scattering sites.[1] If phonon decay through anharmonic interaction is simultaneously present, a "quasidiffusive" propagation mode[2,3] should be discernible[4] whose qualitative features can be understood as follows.[2] We assume, at $t=0$, an injection at one point in a solid of a monochromatic packet of acoustic phonons of frequency ω_0. If the lifetime against elastic scattering, $\tau^*(\omega)$, is short compared to the lifetime against anharmonic decay, $\tau(\omega)$, then during $\tau(\omega)$ propagation proceeds diffusively. For simplicity, we limit anharmonic processes to the spontaneous decay of the ω_0 phonons into two lower-energy phonons with $\omega_1 \simeq \omega_0/2$.[5] This process repeats itself, producing new "generations" of phonons with $\omega_2 \simeq \omega_0/4, \ldots, \omega_i \simeq \omega_0/2^i \ldots$, until the transformed packet reaches the detector. For our purposes it suffices to express the frequency dependence[6] as $\tau(\omega) = \tau_0(\omega/\omega_0)^{-5}$ and as $\tau^*(\omega) = \tau_0^*(\omega/\omega_0)^{-4}$.

The strong dependence of $\tau^*(\omega)$ and $\tau(\omega)$ on ω has two major consequences; namely, every new generation has a much longer lifetime and it enlarges the spatial extent of the packet (as defined by, say, the root mean displacement) compared with that of its predecessor. A measurement of the temporal evolution of the phonon population at a distance L_A from the site of injection reflects, then, almost exclusively the transport of the last generation. That is, for all intents and purposes, the contributions of all previous generations can be neglected. Thus the distance traveled to the detector $L_A \simeq C(D_A t_A)^{1/2}$ and $t_A \simeq \tau(\omega_A)$, where D_A is the effective diffusion constant of the generation which arrives at the detector, and C is a factor of order unity. But from kinetic theory, $D_A \simeq \langle v^2\rangle \tau^*(\omega_A)$, such that $\langle L_A^2\rangle \simeq \langle v^2\rangle \tau^*(\omega_A) t_A$, where v is the sound velocity. Accordingly, $L_A \simeq v[\tau^*(\omega_A)\tau(\omega_A)]^{1/2}$. Moreover, from above, $\omega_A \simeq \omega_0(t_A/\tau_0)^{-1/5}$. By direct substitution, one finds that

$$t_A = C' t_B (t_B \tau_0^4/\tau_0^{*5})^{1/9}, \quad (1)$$

where $t_B = L_A/v$ (the ballistic arrival time) and C' is another factor, of order unity, which contains all the geometric and other proportionality factors which have so far been neglected. It is assumed that $\tau_0^* < \tau_0 < t_B$. It follows from Eq. (1) that $t_A \propto L^{10/9}$ and $t_A > t_B$.

To summarize, the distinguishing features which characterize quasidiffusional propagation are that t_A is nearly linearly related to L_A and not to L_A^2 as in pure diffusion. On the other hand, $t_A > t_B = L/v$. Moreover, the time-of-flight spectrum has a long tail similar to pure diffusion,[7] but the empirically observed diffusion constant increases as L increases.

The experimental conditions and the sample of Al_2O_3:V have been described elsewhere.[8] In the present case 28.8-cm^{-1} light, from a CO_2-laser–pumped CH_3NH_2 laser, was resonantly absorbed

to excite V^{4+} ions from their $E_{3/2}$ ground state to the ${}_1E_{1/2}$ first excited state.[8] Since at this frequency the density of one-phonon states greatly exceeds that in the photon field, the most likely deexcitation of the electronic system is the emission of phonons of $\omega_0 = 28.1\ \mathrm{cm}^{-1}$. A superconducting tin bolometer, $1 \times 1\ \mathrm{mm}^2$ in cross section, was evaporated on the doped end of the crystal (see inset, Fig. 1) and the center of the focused laser beam was positioned at distances L from the bolometer. In this way the beam passes through the width of the sample which is 7 mm. The full width at half maximum of the laser beam was approximately 1 mm with peak power of about 100 W and a pulse width of 250 ns. The time-of-flight spectrum (arbitrarily normalized to the value at $t = 80\ \mu\mathrm{s}$) as obtained at various fixed values of L is shown in Fig. 1. If a diffusion profile[9] is forced to fit the data, then the empirically obtained diffusion constant is found to increase with L as shown in Fig. 2, which also contains a plot of the arrival time, t_A, as determined from the leading edge of each spectrum. The ratio L/t_A is found to be 2.3×10^5 cm/s which is some 5 times smaller than the longitudinal and some 3 times smaller than the transverse sound velocity.

It is seen that the experimental results fit the characteristics of quasidiffusional phonon propagation. It remains to demonstrate quantitative agreement with Eq. (1).

It is necessary to specify $\tau_0{}^*$ and τ_0. Elastic scattering can occur at V^{3+} ($\simeq 1.4 \times 10^{20}\ \mathrm{cm}^{-3}$) and at V^{4+} ($\simeq 7 \times 10^{18}\ \mathrm{cm}^{-3}$) ions through local changes in the mass and/or force constants (defect scattering) or through interaction with the local electronic system. We estimate the cross section, σ, for defect scattering from the relation[1] $\sigma = \delta a_0{}^2(\omega/\omega_D)^4$, in which the Debye frequency $\omega_D = 720\ \mathrm{cm}^{-1}$, the interatomic distance $a_0 \simeq 5$ Å, and the defect parameter $\delta = 1$ (which is the worst case). This leads to a mean free path $l \gtrsim 1$ cm for ω_0 phonons and $l \gtrsim 16$ cm for $\omega_0/2$ phonons. Since this is longer than the sample, we eliminate this type of scattering from further consideration. Similarly, we estimate the cross section for scattering from the spin-orbit–split ${}^3A_2$ level of V^{3+}. Taking a value[10] of $8.25\ \mathrm{cm}^{-1}$ for the spin-orbit splitting and $10^{-5}\ \mathrm{cm}^{-1}$ for the width of the upper state, we obtain a mean free path $l \gtrsim 5 \times 10^4$ cm, so that this form of scattering may also be neglected.

The ω_0 phonons may scatter resonantly from the two E states of V^{4+} from which they were generated. We estimate the scattering cross section[11] by $\sigma = \lambda_0{}^2/\pi$ with $\lambda_0 = 8.1 \times 10^{-7}$ cm which is the wavelength of the initial ω_0 phonons. Using an average sound velocity $v = 7 \times 10^5$ cm/s, we find a mean free path $l_0 = 6 \times 10^{-7}$ cm. This means that the ω_0 phonons are restricted to the volume in which they are created and can be observed only if the excitation volume overlaps the detector. For all other values of L, it is the properties of the decay phonons ω_1, ω_2, ... which control the propagation, since these alone can leave the excitation volume because they are not resonantly scattered.

The decay phonons may scatter nonresonantly from the E states of V^{4+} for which we take the cross section[11] to be $\sigma = (\lambda_0{}^2/\pi)(\Gamma/\omega_0)^2(\omega/\omega_0)^4$, where Γ is the width of the ${}_1E_{1/2}$ state due to emission of ω_0 phonons. From the width, $\Delta\nu$, of the homogeneous infrared absorption line[12] we take $\Gamma \simeq \Delta\nu \simeq 1\ \mathrm{cm}^{-1}$ and obtain a free path $l_1 = 93$

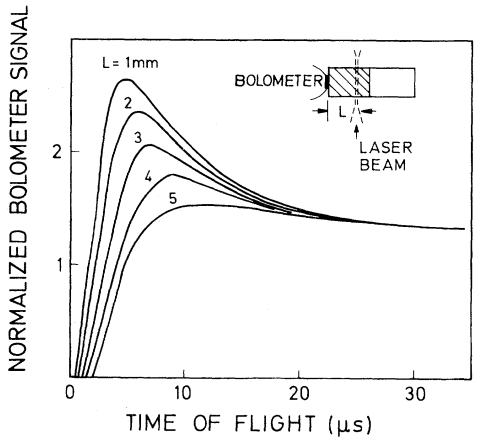

FIG. 1. Time-of-flight spectrum for various propagation distances. The hatched area in the inset represents the vanadium-doped part of the crystal.

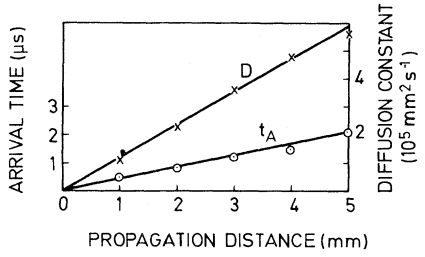

FIG. 2. Pulse front arrival time, t_A, and empirical diffusion constant, D, as functions of the propagation distance.

μm for ω_1 phonons, $l_2 = 1.490$ μm for ω_2 phonons, etc. Since these scattering lengths are well within the dimensions of the experiment, we take nonresonant scattering to be the dominant one and find $\tau_0^* = 0.7$ ns.

For τ_0 we would like to use the published values obtained for the 29-cm^{-1} LA phonons in ruby. Unfortunately, there has been some uncertainty in these values[13] which range from 40 ns[14] to greater than 1 μs.[15] If we choose $\tau_0 \simeq 1$ μs, the ω_0 phonons live on the order of t_A, which is incongruous with their restriction to the excitation volume as a result of the strong resonant scattering and with the observed increase in D_A with L. If we choose the lower value, i.e., $\tau_0 \simeq 40$ ns, we find for $L = 5$ mm that $t_A \simeq 9$ μs which is in fair agreement with the observed 2 μs considering the uncertainty in the factor C'. Caution must, however, be exercised before regarding this result as support for the smaller value of τ_0. The apparent lifetime depends on the excitation level. However, the laser's low (100 W) peak power assures that the excitation in this experiment was well below saturation.[13] On the other hand, the effective τ_0 in the experiment must exceed that for LA phonons, because mode conversion due to repetitive elastic scattering results in roughly a 10 times more efficient[13] conversion into possibly long-lived TA phonons[5] than LA phonons. The TA phonons, however, scatter elastically more efficiently than LA phonons by a factor of roughly 10.[16] Moreover, since the wavelength $\lambda_0 = 0.1 \times 10^{-7}$ cm approximately equals the mean free path against resonant scattering ($l \sim 6 \times 10^{-7}$ cm), a proper treatment of the original 28.8-cm^{-1} excitation may well be a coupled phonon–local-electron state and not a phonon state as has been assumed here. It should, however, be pointed out that there is no evidence in this, or previous, investigations of this crystal system to support or to deny the existence of a coupled mode. It is clear that a rigorous evaluation of t_A is quite complicated, and must be left for some future time.

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