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Lifetimes of high-frequency phonons

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A comparison is made between experimentally and theoretically obtained lifetimes of longitudinal acoustic phonons in Al_2O_3, SrF_2, CaF_2, LaF_3, and α-quartz. Theoretical predictions based on an isotropic, dispersionless Debye solid are shown not be be consistent with most of the experimental results.

Experimentally obtained values for the lifetime τ of acoustic phonons in the frequency range from 0.7 to 3.5 THz are accumulating at an accelerating pace.[1–8] Only a few attempts have, however, been made so far to correlate these results with theoretical predictions. This paper represents a first attempt to carry out an overall correlation in order to illuminate possible weaknesses and strengths in the experimental and theoretical approaches.

Theoretical formulations of phonon lifetimes have been limited to perturbative approaches in which the anharmonic interactions are described in terms of lattice thermal expansion,[9] the Grüneisen parameter,[10] or in terms of linear combinations of the higher-order elastic constants.[11,12] In Refs. 10–12 the density of states for the phonon decay products is determined under the basic simplification of an isotropic, dispersionless Debye solid. More realistic treatments of the dispersion relations are included by Maradudin *et al.*[9,13] and by Nicklasson.[14] The formulations by Klemens[10] and by Orbach and Vredovoe[11] are most readily reducible to calculation and have, accordingly, been most often used in comparison with experimental results. However, since the Grüneisen parameter is not readily available for most of the solids to be discussed here,[15] we shall limit further discussion to results by Orbach and Vredevoe,[11] who find the following expression for the inverse lifetime of a longitudinal (l) phonon which decays into another longitudinal phonon and a transverse (t) phonon, i.e., $l \rightarrow l + t$:

$$\frac{1}{\tau(\omega)} = \frac{\hbar\left[(C_1 + C_4) + \frac{3}{2}(C_2 + C_5)\right]^2}{16\pi\rho^3 v_l^6 v_t^3} \times (1 - \beta^2)(\beta^2 - \beta + \tfrac{3}{8})\omega^5 . \qquad (1)$$

In this expression $\omega = 2\pi\nu$ is the phonon frequency, the C_i are linear combinations of second- and higher-order elastic constants,[16] v_l, v_t are respectively the group velocities of l and t phonons, $\beta = v_t/v_l$, and ρ is the mass density of the solid in which the phonons propagate. Prior to the present paper, published evaluations of Eq. (1) used values of the C_i and v_j for a canonical solid proposed by Orbach and Vredevoe (see Table II below). The ω^5 dependence of τ^{-1} shown in Eq. (1) is a direct consequence of the assumption of an isotropic, dispersionless Debye solid, and was, accordingly, also obtained by Klemens.[10]

Experimental values of τ_l in the presence of $l \rightarrow l + t$ decay have been accumulating from three basic techniques, namely, the so-called ruby phonon spectrometer[1–3,7] (RPS), the vibronic sideband phonon spectrometer (VSPS),[4,5] and coherent phonon generation (CPG) through piezoelectric surface excitation using laser light.[6,17] In the RPS method a nearly monochromatic phonon population is prepared by preferentially trapping in a small region of a sample those phonons which are monoenergetic with a pair of excited electronic levels, as e.g., the $\bar{E}$ and $2\bar{A}$ levels of Cr^{3+} in Al_2O_3 which give rise to the well-known R_1 and R_2 luminescence in ruby. All phonons of other frequencies are not resonantly trapped and accordingly escape from the excitation volume. Under ideal conditions the resonant phonons are completely trapped such that any decrease in time of their number can occur only through anharmonic decay. This method is obviously sensitive to any other mechanism which interferes with complete trapping. The presence of such a competing process in ruby through phonon-assisted energy transport[18] has been deduced by Meltzer in Rives,[19] and the experimental value for τ in ruby listed in Table I below has been corrected for this effect. However, other as yet unidentified competing mechanisms are also thought to be active[7,20] but their contribution to the results in ruby have not as yet been taken into account. In the LaF_3:Pr^{3+} system[8] these competing effects are not thought to be present.[20] The values of τ quoted for the CaF_2:Sm^{2+} case are based on preliminary uncorrected experimental results.[20] Resonant trapping occurs through strong phonon scattering at the excited ions. It is assumed that this results in random mode conversion. Since τ_t may exceed τ_l by orders of magnitude[11] the experimental values of Table I are further corrected for the

TABLE I. Comparison between experimental and theoretical phonon lifetimes.

ν (THz)	Crystal	Method	τ_l (Exp.) (sec)	τ_l (Theor.) (sec)
0.69	LaF_3:Pr^{3+}	RPS	1.5×10^{-8}	6×10^{-7}
0.87	Al_2O_3:C^{3+}	RPS	4×10^{-8}	4.6×10^{-6}
1	SrF_2:Eu^{2+}	VSPS	3×10^{-7}	2.5×10^{-8}
2	SrF_2:Eu^{2+}	VSPS	2.5×10^{-7}	2.1×10^{-9}
3	SrF_2:Eu^{2+}	VSPS	2×10^{-7}	2.7×10^{-10}
3.4	SiO_2	CPG	$\gtrsim 4\times10^{-7}$	2.3×10^{-10}
3.5	CaF_2:Sm^{2+}	RPS	$\gtrsim 4\times10^{-8}$	3.3×10^{-10}

probability that a given mode conversion leads to a longitudinal phonon.[21]

In the VSPS technique the spectral, spatial, and temporal evolution of a phonon distribution in a crystal is monitored through a vibronic interaction between the phonons and certain localized electronic states associated with probe ions placed in dilute solution into the crystal. Preliminary values for the lifetime of 1, 2, 3 THz phonons observed with this spectrometer were reported in Ref. 5. The results quoted in Table I for the SrF_2:Eu^{2+} case have been further corrected for the effect of competing decay modes and for the effect of mode conversion, the latter correction being necessary because of strong defect scattering.

In the CPG method a monochromatic, coherent beam of phonons is produced through a surface interaction of far infrared laser light with a piezoelectric crystal. The phonons injected in this way have the frequency of the laser light and propagate ballistically along a specified direction,[22] i.e., possess a well-defined wave vector and in most cases are predominantly longitudinally or transversely polarized.[22] Any phonon decay products are produced incoherently, do not propagate in the same direction as the injected beam,[23] and require longer periods of time to traverse a fixed length of the crystal. Accordingly, the ratio of the amplitudes of the coherent to incoherent beam in a time-of-flight measurement is a measure of the probability of decay of the injected phonon population. The CPG method has been applied so far to only the single case of 3.4-THz longitudinal phonons propagating in a Y-cut quartz crystal.[6] Analysis of these results yields a lower bound on the lifetime equal to $\gtrsim 0.4$ μsec. Since in this case the phonons travel ballistically no correction due to competing modes or mode conversion is required.

The theoretical values listed for comparison in Table I are obtained on the basis of Eq. (1), the parameters of Table II, and on the following further simplification. Since the RPS and VSPS method involve diffusive phonon flow, v_l and v_t are determined by averaging over velocities in different directions wherever such values are available in the literature. In the same spirit the values quoted in Table II for the C_i's are from linear combinations of averages over the second- and higher-order elastic constants,[24] wherever available liquid helium temperature values were used.

Comparison of the experimental and theoretical τ_l listed in Table I indicates a marked dichotomy. For $\nu < 1$ THz the theoretical lifetimes are longer than those observed experimentally whereas for $\nu > 1$ THz they are shorter than those experimentally observed. Moreover, the largest discrepancies are for the highest phonon frequencies. These results imply that the ω^5 dependence predicted for an isotropic, dispersionless Debye solid is not generally applicable.

The real crystal systems are, of course, anisotropic and dispersive. In the case of zone boundary phonons in quartz, Grill[25] reports that when more realistic dispersion relations[26] are used the density of terminal states is markedly lowered such that the τ_l for these phonons increases by a factor of 10^3 compared to that predicted for the isotropic, dispersionless case. Similar estimates are not as readily performed for the RPS and VSPS methods, since in these cases the phonons propagate diffusively rather than in single well-defined directions. In order to gain some insight into the effects of realistic dispersion relations, we have estimated the effect on the density of terminal states due to propagation along the three highest symmetry direction

TABLE II. Parameters used to evaluate Eq. (1).

Parameter	Units	Al_2O_3	SiO_2	SrF_2	CaF_2	LaF_3	Canonical
v_l	10^5 cm/sec	11.09	5.75	5.24	6.9	5.8	5
v_t	10^5 cm/sec	6.04	4.2	2.64	3.6	2.79	2.9
ρ	g/cm^3	3.986	2.65	4.321	3.18	5.93	3
C_1, C_2	10^{11} dyn/cm^2	23	4.8	6.95	8.6	9	5
C_4, C_5	10^{11} dyn/cm^2	−125	−23	−27	−35.6	−10	5

of Al_2O_3,[27] SrF_2,[28] and CaF_2.[29] Since the dispersion relations in the 1-THz region of Al_2O_3 are essentially isotropic and dispersionless, the source of the observed discrepancy must be elsewhere for this material. SrF_2 and CaF_2 are less isotropic then Al_2O_3 and become progressively more dispersive above 1 THz. An estimate of the correction to τ_l due to those topological effects indicates, at the minimum, a tendency to progressively undo the discrepancies indicated in Table I.

It appears, accordingly, that for SrF_2, CaF_2, and α-quartz that a major component of the discrepancy between experimentally and theoretically obtained values of τ_l lies in the differences in the density of phonon decay states between the real crystal and that of an isotropic, dispersionless Debye solid, and that a, albeit more tedious, computer based evaluations, perhaps of the type proposed by Maradudin *et al.*,[9,13] of this effect is warranted.

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