Intrinsic decay lengths of quasimonochromatic phonons in a glass below 1 K

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The temperature dependence and magnitudes of the reciprocal decay length l^{-1} of low-intensity longitudinal and transverse phonon pulses propagating in fused silica glass between 0.02 and 0.5 K for frequencies near 0.5 GHz are reported. The temperature dependence of l^{-1} is in good agreement with resonant scattering from two-level systems, and the magnitudes yield thermal conductivities in good agreement with published data.

At temperatures below 1 K, the thermodynamic and transport properties of amorphous insulating solids are strikingly different from those of pure crystalline dielectrics.[1,2] In order to explain their anomalous behavior, theoretical models of glasses have been proposed which postulate the existence of broad distributions of two-level atomic tunneling or defect states.[3-5] These states are strong resonant scatterers of thermal phonons and thus account for the unusually low thermal conductivities of glassy insulators.[6] In view of the generality of the existing theoretical picture, it is likely that the properties of actual glasses may differ in detail from previous predictions.[7,8] For example, the exclusive existence of two-level systems has not yet been demonstrated from structural models of the glassy state. However, experiments on silica glasses with high-frequency acoustic waves have shown that the states are saturated at "high" phonon intensities, thus supporting the assertion that the defect states can be represented as two-level systems.[9,10] At low phonon intensities a fundamental aspect of a resonant two-level model is the existence of a phonon reciprocal decay length l^{-1} whose frequency and temperature dependence is[3,9]

$$l_i^{-1}(\omega, T) = l_{0,i}^{-1} \tanh(\hbar\omega/2kT) . \quad (1)$$

The $\tanh(\hbar\omega/2kT)$ term is the fractional population difference between two levels separated by energy $\epsilon = \hbar\omega$ in thermal equilibrium and

$$l_{0,i}^{-1}(\omega) = \pi\bar{n}(\hbar\omega)\gamma_i^2\omega/\rho v_i^3 \quad (2)$$

is the $T = 0$ K reciprocal decay length, where $\bar{n}(\hbar\omega)$ is the density of systems contributing to phonon scattering, γ_i is an average linear coupling between a tunneling state and a strain of polarization i, ρ is the mass density, and v_i the phase velocity.

In the present work, we present the first measurements of the intrinsic temperature- and polarization-dependent phonon decay lengths in fused silica glass. Temperatures as low as 0.02 K were reached for phonon frequencies above 0.5 GHz, so that the regime $\hbar\omega > kT$ was achieved. Our results are the first for low-intensity transverse phonon pulses and indicate that phonons of *both* polarizations (transverse and longitudinal) propagate for approximately 250 wavelengths before they undergo resonant scattering when $T \ll \hbar\omega/k$. Finally, our investigation confirms the temperature dependence of l^{-1} expected for resonant scattering, as given by Eq. (1), and lends further support for the existence of two-level systems at low energies.

In order to observe intrinsic phonon decay in a glass it is necessary to work at peak acoustic intensities sufficiently low so that no appreciable saturation of the defect states occurs. Figure 1 shows the effective inverse decay lengths[11] due to resonant scattering for 0.4 μsec, 0.5-GHz phonon pulses in fused silica at 0.023 K. At acoustic intensities $I \gtrsim 10^{-5}$ W/cm^2 the tunneling states are saturated.[12] As the intensity is decreased, the effective l^{-1} increases as the states unsaturate. At sufficiently low I, l^{-1} becomes essentially independent of I, and we shall characterize the decay lengths in this region as "intrinsic."

In Fig. 1 the transverse-mode saturation intensity appears to be greater than that for the lon-

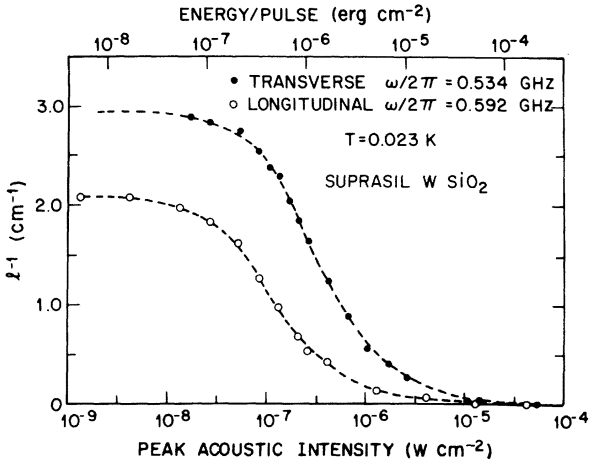

FIG. 1. Reciprocal decay lengths for 0.4-μsec phonon pulses in fused silica at 0.023 K as a function of incident intensity.

gitudinal mode. However, there are uncertainties of ±3 dB in determining the absolute acoustic intensitites for each mode so that we cannot rule out the possibility that the thresholds appear at equal intensities or that the relative positions are reversed. It is significant to note that at the lowest intensity the energy density in a received longitudinal pulse is only three times greater than the energy density of the thermal-phonon background at 0.023 K in a pulse bandwidth (~2 MHz).

The intrinsic reciprocal decay length for the transverse mode is greater than for the longitudinal mode. However, the absorption per wavelength is the *same* for both modes, $l^{-1}\lambda = \pi(\Gamma/\omega) = 2\times10^{-3}$ at 0.023 K. We see that the relative damping is small, $\Gamma/\omega \ll 1$, demonstrating that phonons are indeed well-defined excitations in the frequency and temperature range under investigation here, where $\hbar\omega \approx kT$.

The experiments were carried out on 0.635-cm cubes of Suprasil W fused silica containing OH at concentrations $\lesssim 1\times10^{-6}$. Transducers were ≈1-μm films of ZnO produced by dc reactive sputtering.[13] By careful control of the deposition process ZnO films with longitudinal- or transverse-mode transduction could be grown with effective one-way conversion efficiencies of −8 and −14 dB, respectively. The low temperatures were achieved with a ^{3}He-^{4}He dilution refrigerator and the microwave signals propagated in a low-loss miniature coaxial transmission line. The structure was nonresonant although a small stub stretcher, located in the refrigerator at the sample, functioned as an impedance matcher.[14] This device was in good thermal contact with the refrigerator's mixing chamber so that it served also as a thermal shunt for heat carried down the coaxial center conductor. Nevertheless, heat leaks into the sample prevented the cooling of the glass below 0.023 K even though the mixing chamber was at less than half this temperature. We also became aware of spurious microwave signals which could contribute to local heating in the acoustic-beam region. This effect could contribute a temperature uncertainty of at most ±0.002 K at the lowest temperature and the small correction has been applied to the data.

An experimental technique was used which permitted a direct measurement of the intensity-dependent absorption shown in Fig. 1. A microwave pulse was split into two channels, one consisting of a linear acoustic delay line at room temperature and the other containing the glass sample. The microwave pulse generated acoustic echos in both samples which were combined and amplified. A variable attenuator in the reference line was adjusted to balance the sample-reference echo amplitudes at the single detector. Thus the increased absorption of the glass sample as the power was decreased from saturation amplitudes was directly given by the increased attenuation required in the reference channel.

The temperature dependence of l^{-1} was studied by measuring at discrete temperatures the amplitude of the first round-trip echo relative to a reference echo at a fixed incident intensity 5 dB above the minimum intensity for each mode. The results of these measurements[15] are shown in Fig. 2, where the inverse decay lengths due to resonant scattering are plotted vs $1/T$. The solid line drawn through the data is the function given in Eq. (1). The only adjustable parameter is the magnitude of $l_{0,i}^{-1}$. The data show excellent agreement with a $\tanh(\hbar\omega/2kT)$ dependence and deviations from a T^{-1} dependence are in clear evidence for $\hbar\omega/2kT > 0.5$. We find $l_{0,L}^{-1} = 3.9\pm0.3$ cm^{-1} at 0.59 GHz and $l_{0,T}^{-1} = 5.70\pm0.5$ cm^{-1} at 0.53 GHz. From Eq. (2) we calculate $\tilde{n}\gamma_L^2 = (1.4\pm0.1)\times10^8$ erg/cm^3 and $\tilde{n}\gamma_T^2 = (0.63\pm0.05)\times10^8$ erg/cm^3, so that if both modes interact with the same states $\gamma_L/\gamma_T = 1.5$. We emphasize that $\tilde{n}(\hbar\omega)$ is the density of only a subset of the states contributing to the specific heat since C_p also contains contributions from states not effective in phonon scattering. We previously estimated $\tilde{n}$ to be ~1% of the states contributing to C_p.[9]

Phase-velocity measurements when $\hbar\omega \ll kT$ have been previously analyzed[16,17] for $\tilde{n}\gamma_i^2$. However, for $\hbar\omega \approx kT$ we find that the phase velocity is weakly temperature and frequency dependent.[18] A fit of the velocity data to a numerical Kramers-Kronig inversion of Eq. (1) and (2) (assuming a constant $\tilde{n}$) yields the parameters $\tilde{n}\gamma_L^2 = (2.0\pm0.1)\times10^8$ erg/cm^3 and $\tilde{n}\gamma_T^2 = (0.89\pm0.05)\times10^8$ erg/cm^3. These values are 40% larger than the parameters

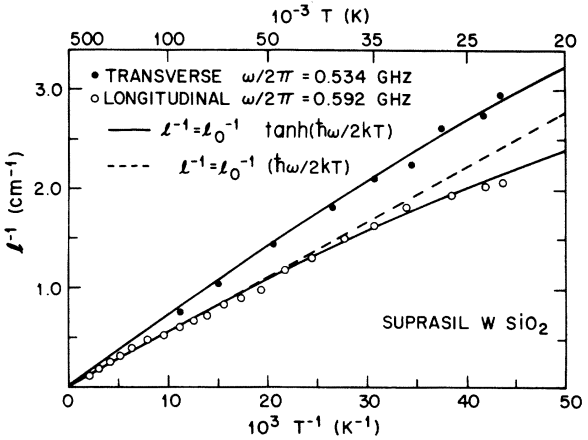

FIG. 2. Temperature dependence of the intrinsic reciprocal decay lengths of phonons in fused silica.

inferred from the l^{-1} measurements. We have no firm explanation for the small discrepancy except to note that all velocities showed a temperature dependence stronger than $\log T$ for $\hbar\omega/kT \ll 1$ which may arise from an energy-dependent density of states.

The phonon contribution to the thermal conductivity may be computed from the kinetic formula

$$\mathcal{K} = \frac{1}{3} \sum_i \int_0^{\omega_D} d\omega\, C_i(\omega) v_i l_i(\omega)$$

in which we sum over a polarization index i, $C_i(\omega)$ is the Debye specific heat

$$C_i(\omega) = \hbar^2\omega^4 e^{\hbar\omega/kT}/2\pi^2 v_i^3 k T^2 (e^{\hbar\omega/kT} - 1)^2 ,$$

and $l_i(\omega)$ is given by Eqs. (1) and (2). It is therefore assumed that dispersionless Debye phonons are responsible for thermal transport. On integrating the above expressions we have

$$\mathcal{K}(T) = A(\rho k^3/\pi^3\hbar^2)(v_L/\tilde{n}\gamma_L^2 + 2v_T/\tilde{n}\gamma_T^2) T^2 \equiv \mathcal{C} T^2 ,$$

where $A = 1.645$. With $v_L = 5.8 \times 10^5$ cm/sec and $v_T = 3.75 \times 10^5$ cm/sec we calculate $\mathcal{C} = (4.4 \pm 0.4) \times 10^{-4}$ W/cm K^3 from the $\tilde{n}\gamma_i^2$ derived from l^{-1} data and $\mathcal{C} = (3.1 \pm 0.3) \times 10^{-4}$ W/cm K^3 from the phase-velocity results. The thermal conductivity of Suprasil W has been measured previously from 0.02 to 0.5 K by Lasjaunias *et al.*[7] who find $\mathcal{K} = 2.6 \times 10^{-4} T^{1.95}$ in this region. No experimental errors are quoted but uncertainties of $\approx 40\%$ in the absolute magnitude of $\mathcal{K}$ below 0.02 K would be compatible with either of our calculated $\mathcal{C}$ values. The $\mathcal{K}$ calculation implicitly assumes an energy independent $\tilde{n}$. However, half of the contribution to $\mathcal{K}$ at a given T comes from phonons of energy $> 2.6kT$, and 10% from energies $> 6kT$, so that a weak ω dependence of $\tilde{n}$ would decrease the calculated $\mathcal{K}$. Of course, measurements of $l_0^{-1}(\omega)$ at higher frequencies could test this idea. It is also reassuring to see that we slightly overestimate $\mathcal{K}$ since there may be other mechanisms which add to the thermal resistance.

We have shown that intrinsic phonon decay in fused silica glass below 1 K exhibits a temperature dependence expected for resonant scattering from two-level systems. Phonons of longitudinal and transverse polarizations are found to have equal characteristic damping parameters in fused silica. The thermal conductivity expected for phonon scattering by two-level systems is in good agreement with published data. The present work emphasizes the need for an independent measurement of the energy dependence of the density of scattering states.

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[11] The effective reciprocal decay length is given by $l^{-1} = (2L)^{-1} \ln(E_0/E_1)$, where E_0 is the incident pulse energy per cm^2 and E_1 is the pulse energy per cm^2 after one round trip through the sample. In the completely saturated or unsaturated regimes where decay is exponential l^{-1} is determined by the ratio E_0/E_1. In the intermediate nonlinear regime l^{-1} will depend on the absolute pulse energy, since the defect state lifetime is much greater than the pulse length [$\Gamma^{-1} \approx 650$ μsec at 0.5 GHz and 0.023 K, B. Golding and J. E. Graebner (unpublished)]. See Ref. 9 for a discussion of propagation in the partially unsaturated case.

[12] A background absorption of 0.3 cm^{-1}, due mainly to diffraction effects, has been subtracted from the curves.

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