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Ultrasonic Attenuation in Quartz at Low Temperatures*

R. C. Purdom and E. W. Prohofsky

Department of Physics, Purdue University, Lafayette, Indiana 47907

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The attenuation of longitudinal ultrasound in X-cut quartz is explained in terms of three-phonon interactions involving slow transverse thermal phonons. The approximately T^6 temperature dependence of the attenuation is understood when the dispersion of the slow transverse branch is properly considered. This mechanism can also account for the measured attenuation of four additional acoustic modes.

Theoretical models for three-phonon interactions in solids have successfully explained the attenuation of longitudinal ultrasound in many insulators at low temperatures with the notable exception of quartz.[1] A thorough analysis of the discrepancies between the theoretical predictions and the experimental results for the attenuation of longitudinal acoustic waves in the x direction of quartz has been given by Lewis and Patterson.[2]

Mechanisms which involve the interaction of the longitudinal acoustic wave with longitudinal thermal phonons such as those proposed by Maris[3] and Shiren[4] are too weak in X-cut quartz to account for the measured attenuation. Kalejs, Maris, and Truell's[5] mechanism where the longitudinal acoustic wave interacts with transverse thermal phonons can explain the large magnitude of the attenuation, but its T^8 temperature dependence differs from the approximately T^6 temperature dependence measured. This mechanism should be the dominant mechanism, however, because the predicted attenuation varies as $(1/v_{th})^8$, and in the x direction of quartz the slow transverse phonon velocity is approximately $\frac{3}{5}$ that of the longitudinal phonon velocity. We believe that this is the dominant mechanism and that if the dispersion of the slow transverse branch is properly considered, the $\sim T^6$ temperature dependence of the attenuation can also be understood.

All such phonon-phonon interaction mechanisms which involve the interaction of longitudinal acoustic phonons with thermal phonons of any polarization are normally forbidden because of the inability of these processes to conserve energy. However, because of the finite lifetime τ of the thermal phonons, energy need be conserved only to the order of $\hbar/\tau$. Therefore, in the calculation of the absorption of longitudinal waves by three-phonon processes, a Lorentzian function replaces the δ function in energy in order to represent the fuzziness of the thermal-phonon energy states.

The attenuation of an acoustic wave of frequency ω and velocity v_a interacting with thermal phonons of frequency ω_1, group velocity v_{g1}, and wave vector K_1 is given by[6]

$$\alpha\left(\frac{\mathrm{dB}}{\mathrm{cm}}\right) = \frac{4.34\hbar\omega}{32\pi^3\rho^3 v_a^3}\iiint \frac{A^2K_1^4}{\omega_1^2}\frac{\partial N_1}{\partial\omega_1} \times \frac{\omega\tau}{1+[1-(v_g/v_a)\cos\theta]^2\omega^2\tau^2}\,d^3K_1\,, \quad (1)$$

where N_1 is the occupation number of the thermal phonons, θ is the angle between the propagation directions of the acoustic and thermal phonons, and A is the elastic coupling parameter.

In the continuum model the allowed thermal-phonon frequencies are not bounded. By including

dispersion, an upper bound in frequency ω_{max} is set for the thermal phonons. If the mode is highly dispersive and $\omega_{max} \lesssim 5\kappa T/\hbar$, the continuum model loses its validity since the continuum model predicts that the attenuation is dominated by thermal phonons of average frequency $\sim 5kT/\hbar$. Therefore, calculating the attenuation with the inclusion of dispersion shifts the dominant thermal phonons down in frequency. Since $\partial N_1/\partial\omega_1$ increases more slowly with temperature for lower thermal-phonon frequencies, the resultant effect of dispersion is an over-all decrease in the temperature dependence of the attenuation.

The exact calculation of the attenuation is difficult because of the anisotropy of the phonon velocity, the coupling parameter, and the dispersion of the phonon velocity. Also, there is uncertainty in both the magnitude and the frequency dependence of τ. In order to make an estimate of the attenuation, we assume that A is isotropic and that τ is given by the phonon lifetimes taken by Maris[3] from thermal conductivity data.

We use the dispersion curves calculated and measured by Elcombe.[7] By performing the calculation numerically for the range of dispersion thus indicated, we find that the attenuation is dominated by large-wave-vector phonons. Therefore, for the thermal phonons of interest, v_g is small and the relative ability of a particular interaction to conserve energy does not depend on the angle between the acoustic and thermal phonons. Because of the increase of $\partial N_1/\partial\omega_1$ with decreasing ω_1, we do not expect the interactions to peak in the collinear direction but in the directions which are the most dispersive.

We assume that over a solid angle Ω the dispersion is similar to that measured by Elcombe in the z direction and that this region dominates the attenuation. We also assume that the form of the coupling of the acoustic waves with these shorter-wavelength thermal phonons can be approximated by Eq. (1) which is valid in the long-wavelength limit. With these approximations, the attenuation reduces to

$$\alpha\left(\frac{\mathrm{dB}}{\mathrm{cm}}\right) = \frac{4.34\,\hbar^2 A^2}{224\,\pi^3 \rho^3 k\, T\, v_a^3} \frac{K_{max}^7}{\omega_{max}^2\,\tau} \times \frac{\exp(\hbar\omega_{max}/kT)}{[\exp(\hbar\omega_{max}/kT)-1]^2}\,\Omega. \quad (2)$$

In Fig. 1 the solid line shows the theoretical prediction of our model for the attenuation of 9-GHz longitudinal waves in the x direction of quartz compared to the data of Lewis and Patterson.[2] We have set $\omega_{max} = 9.4\times10^{12}$ sec^{-1}, $K_{max} = 6.3\times10^{7}$ cm^{-1}, and $A^2\Omega = 3.6\times10^{24}$ dyn^2/cm^4.

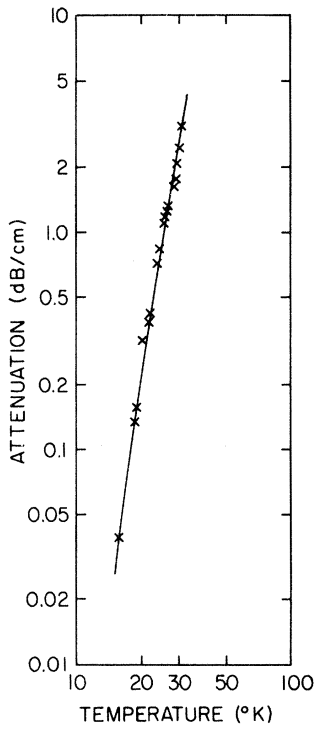

FIG. 1 Longitudinal microwave phonon attenuation in nearly perfect X-cut quartz (Ref. 2): × ; solid curve indicates theory.

Calculating A^2 from the long-wavelength elastic constants[8] for the coupling of x-axis longitudinal acoustic waves to z-axis transverse waves, we obtain $A^2 \cong 1.0\times10^{24}$ dyn^2/cm^4. Therefore, for Ω equaling $\sim\pi$ sr, a value consistent with the calculated dispersion, our model explains both the temperature dependence and the large magnitude of the attenuation.

In addition, other predictions of our model which can be seen in the final attenuation expression lend validity to our approximations. The expression should be valid for all acoustic waves interacting with phonons of the slow transverse branch if the velocity of the acoustic wave is larger than the velocity of the long-wavelength slow transverse waves. Under the same approximations, the predicted temperature dependence of the attenuation of these acoustic waves is independent of the propagation directions, polarization directions, and velocities of the modes. Also, we would expect the magnitude of the attenuation to be approximately the same scaling by the appropriate A^2/v_a^3. This is verified by the experimental data. Lewis and Patterson's[2] measurement of the attenuation of fast transverse waves in X-cut, quasilongitudinal waves in AC-cut, and transverse

waves in BC-cut quartz show nearly identical attenuation with that of x-direction longitudinal waves. Also, Ganapolskii and Chernet's[9] measurement of the attenuation of longitudinal waves in the z direction of quartz shows the same pattern of attenuation.

In summary, we believe that the marked anomalies in the attenuation of acoustic waves peculiar to quartz are a direct result of the unusually large dispersion of the slow transverse branch.

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Excited States in a Model Polaron Hamiltonian*

D. E. Hagen, L. L. Van Zandt, and E. W. Prohofsky
Department of Physics, Purdue University, Lafayette, Indiana 47907
(Received 14 April 1969; revised manuscript received 22 October 1969)

The energy-spectrum problem is treated for a bound stationary polaron. The Pekar Hamiltonian is used to describe the polaron and its polar lattice. This model is based on two main assumptions. First, the carrier is assumed to move much faster than the lattice ions. Second, the polaron is assumed to be spread out over a distance that is much larger than the lattice constant. These assumptions restrict us to a strong-coupling regime. A variational principle especially well suited for finding excited states has been used in a search for solutions of the polaron Hamiltonian. Using trial wave functions having s-, p-, and d-like symmetry, some well-separated approximate eigenstates of this model polaron have been found. The excited states represent internal and self-consistent excited-state solutions of the polaron Hamiltonian. It is found that the energy of each state is proportional to the square of the polaron coupling constant. Also, the qualitative features of the spectrum, i.e., the number of energy levels and their relative positions, are independent of the coupling constant.

I. INTRODUCTION

A conduction electron or a valence hole in a deformable crystal lattice creates a distortion in the over-all periodicity localized about itself. The dynamics of low-density carriers in insulators therefore involves the effects of local lattice deformations. The complex of carrier and associated deformation is a polaron. Reviews of work done on the polaron problem have been given by Appel[1] and by Kuper and Whitfield.[2] Other work relating to polaron excited states has been reported by Evrard, Devreese, and Kartheuser.[3–5]

While it is frequently convenient to consider the polaron as a single entity like a simple particle, it is clear that the many actual particles of which it is composed together with their varying interactions produce a more complex system. We expect these complexities to be made manifest in the dynamical behavior of the polaron; the polaron should show internal degrees of freedom or internal states, and these internal states should be apparent in phenomena involving energies comparable to the carrier-lattice deformation energy.

In this paper, we report the results of a theoretical search for such states. To find them more simply, we have made a number of approximations designed to reduce extraneous considerations to a minimum and focus on the carrier-lattice deformation interaction. We treat the case of the stationary polaron only, i.e., the polaron as a whole does not move through the lattice. We assume a constant optical-phonon frequency. We also assume a sym-