Phonon contribution to the ultrasonic attenuation in neutron-irradiated quartz

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It is shown by calculations that the thermal phonon contribution to the attenuation of ultrasonic waves in slightly neutron-irradiated crystalline quartz may be as much as 1 order of magnitude smaller than the measured attenuation for frequencies below 1 GHz.

INTRODUCTION

Recent attention has been directed to the study of neutron-damaged crystalline quartz due to the observation in the material of thermal[1-5] and acoustic[6-8] glasslike properties at low temperatures. Extensive exposure of quartz crystals to the radiation spectrum of a reactor core produces a wide range of lattice defects: electronic and color centers due to the intense ionizing radiation, point, and extended structural defects including amorphous inclusions due to fast neutrons. For neutron doses D below $10^{19} n/\mathrm{cm}^2$ the damage to the lattice appears slight,[3,9] but nevertheless the density of low-energy excitations created in the form of a broad distribution of two-level systems (TLS) is sufficiently large to produce the dominant glassy behavior of the low-temperature properties of neutron-irradiated quartz (NIRQ).

Earlier acoustic measurements in NIRQ[10,11] showed that the ultrasonic attenuation α at a given frequency $\Omega/2\pi$ decreased at high temperatures ($\Omega\tau < 1$) and increased at low temperatures ($\Omega\tau > 1$) relative to the attenuation before irradiation, in qualitative agreement with phonon viscosity losses $\alpha \sim \Omega^2\tau/(1+\Omega^2\tau^2)$ for a thermal phonon relaxation time τ reduced by imperfection scattering, but quantitative agreement has been lacking. More recent ultrasonic experiments in slightly irradiated quartz ($D \simeq 5\times 10^{18} n/\mathrm{cm}^2$) show that the attenuation tends towards an $\Omega^0 T^3$ dependence at and below liquid-helium temperatures.[6,8] These losses have been attributed[6] to the nonresonant interaction[12] of the sound waves with the TLS of the amorphous inclusions for $\Omega\tau_m > 1$, where τ_m is the minimum relaxation time of the distributed TLS. At higher temperatures the attenuation versus T curve shows a general increasing trend and displays a number of anomalous features such as plateaus and maxima presumably due to other radiation created defects. We note that in the frequency range explored here, the measured attenuation at high temperatures shows approximately a linear dependence in Ω characteristic of a distribution of relaxations.[13] Since the high-temperature attenuation in NIRQ is smaller than in virgin quartz it has not been possible to separate the thermal phonon contribution to α from other radiation induced losses. It is the purpose of the present paper to calculate the magnitude of the phonon losses in NIRQ in order to facilitate the analysis of the various high-temperature acoustic losses due to neutron damage.

PHONON VISCOSITY AT LOW AND HIGH TEMPERATURES IN NIRQ

By means of a Boltzmann-equation approach in the relaxation-time approximation it is possible to calculate the ultrasonic absorption coefficient in dielectric solids for arbitrary $\Omega\tau$[14,15] provided that $\Omega < k_B T/\hbar$ and that phonons are well defined, $l_i > \lambda_i$ (l_i and λ_i are, respectively, the mean free path and wavelength of thermal phonons). For pure dielectrics one obtains the following expressions for the attenuation coefficient α_{jk} of a sound wave of polarization j propagating along $\mathbf{k}$ with velocity c_j in anharmonic interactions with thermal phonons of wave vector $\mathbf{q}$, polarization i, and velocity v_i.[16,17] For low temperatures, $(1+v_i/c_j)^{-1} < \Omega\tau < (1-v_i/c_j)^{-1}$,

$$\alpha_{jk} = \frac{\pi^2 k_B^4 T^4 \Omega}{\rho c_j^3 h v_i^3} \gamma_j^2 J_4(2T_{D_i}/\pi T) \tag{1}$$

and for high temperatures, $\Omega^2\tau^2 \ll 1$,

$$\alpha_{jk} = \frac{3KT\Omega^2}{\rho c_j^3 s^2} \Gamma_j^2 . \tag{2}$$

In Eq. (1), γ_j^2 is an average Grüneisen parameter for the dominant anharmonic interaction, J_4 is the transport integral of the specific heat, and T_{D_i} is the Debye temperature of the phonon branch i. In the approximations considered, which hold for frequencies below 30 GHz, the attenuation is independent of the phonon relaxation time and proportional to ΩT^4.

In Eq. (2), K is the thermal conductivity for a heat flux parallel to the sound propagation direction, s is the Debye average wave velocity, and Γ_j^2 is an effective ultrasonic Grüneisen parameter (a thermal-conductivity-weighted average of the mode Grüneisen parameters). For longitudinal waves ($j = \mathrm{L}$) Γ_j^2 includes contributions which arise from local equilibrium effects, such as thermoelastic losses, through the introduction in the theory of a sound-wave-modulated local effective temperature;[18] $T(\mathbf{r},t) = T + \delta T \exp[i(\mathbf{k}\cdot\mathbf{r} - \Omega t)]$, where δT is to be determined from energy conservation. This implies the concept of a local equilibrium phonon distribution function $f(\mathbf{q},\mathbf{r},t)$ for regions of dimension $L \lesssim l_i$. Such contributions to the longitudinal attenuation are usually a small fraction of the total attenuation. Local equilibrium effects do not affect the at-

tenuation of shear waves ($j = \mathrm{T}$) since δT is proportional to specific-heat-weighted averages of the mode Grüneisen parameters which vanish for pure shear strains. These effects are also disregarded for $\Omega\tau >> 1$, where the relaxation is towards global equilibrium at the crystal temperature.

Several authors[19,20] have extended the formalism leading to Eqs. (1) and (2) to the case of impure dielectrics by separately considering contributions to the thermal phonon collision rate due to elastic scattering by defects (relaxation time τ_e) and due to intrinsic anharmonic processes (relaxation time τ_i). It has been shown[17,20] that Eq. (2) remains formally valid for both shear and longitudinal waves with K determined now by the total relaxation rate $\tau^{-1} = \tau_e^{-1} + \tau_i^{-1}$ but while Γ_T^2 can still be explicitly written as thermal-conductivity-weighted averages of the mode Grüneisen parameters (which may now depend on the impurity concentration) the contributions to Γ_L^2 from local equilibrium effects depend additionally on both τ_e and τ_i in a complicated way. For "dirty" crystals ($\tau_e << \tau_i$) it is possible to solve explicitly for Γ_L^2 and α_{Lk} but the results depend on how the mode Grüneisen parameters vary with $\mathbf{q}$ and i. Heuristically, one may retain Eq. (2) for impure dielectrics by taking Γ_j^2 as an adjustable parameter whose value and impurity concentration dependence are to be extracted from the experimental results.[17,21] In addition, when applying Eq. (2) to the attenuation in NIRQ, consideration should be given to the fact that neutrons may introduce such large numbers of defects into the lattice to render it inhomogeneous. However, diffuse x-ray scattering experiments in NIRQ show that neutrons create defective (heavily strained) regions, which are most likely amorphous, of mean radius $r \simeq 10$ Å, and that these clusters are not significantly changed in size with increasing doses.[9] Thus, as long as the local distribution functions (for elastic and inelastic scattering) are defined for $L \lesssim l_i$ the use of the Boltzmann-equation formalism is justified for l_i greater than both λ_i and r. From the thermal conductivity of NIRQ[1] it is found that at about 100 K, $l_i \gtrsim 100$ Å, which satisfies the above requirements. In the present context the radiation induced amorphous regions are considered as elastic scattering centers for thermal phonons.

COMPARISON WITH EXPERIMENTS AND DISCUSSION

From the available experimental evidence, in calculating the ultrasonic attenuation in slightly irradiated quartz, we may neglect changes in the mass density ρ (3.65 g/cm^3), the average phonon velocity s (4.26 km/sec), and the wave velocities, and take the specific heat C of NIRQ for $T \lesssim 5$ K equal to that of virgin quartz.[4] Since neither the average Grüneisen parameter nor the third-order elastic constants of NIRQ are available at present it is not possible to estimate reliably the effect of neutron irradiation on the mean anharmonicity of the quartz lattice. Furthermore, previous estimates of this effect for NIRQ[17] were done assuming that phonon viscosity was the principal cause of the measured high-temperature attenuation which may be correct only for high hypersonic frequencies. We note that if the decrease in the ultrasonic attenuation[10] in NIRQ at 1 GHz and 90 K is solely attributed to changes in its thermal conductivity and its mean anharmonicity [according to Eq. (2)] the corresponding Γ_j^2, for a dose of $10^{19} n/\mathrm{cm}^2$, would increase from 0.6 to 0.84 for ac shear waves and from 2.3 to 4.3 for x longitudinal waves.[17] We find in addition, for amorphous quartz, that anisotropic elastic continuum calculations based on the measured third-order elastic constants[22,23] yield values of $\Gamma_L^2 = 3.23$ and of $\Gamma_T^2 = 0.25$ which are not too different from those corresponding to crystalline quartz. Hence, we may reasonably assume that no large error will be incurred in the present calculations by taking for the mean anharmonic parameters of slightly NIRQ the values corresponding to virgin quartz ($\Gamma_L^2 = 2.3$, $\Gamma_T^2 = 0.6$, $\gamma_L^2 = 2.78$, and $\gamma_T^2 = 0.23$).[16,17] From the mean thermal phonon relaxation time $\bar{\tau} = 3K/Cs^2$ we find that the low-temperature regime sets in below about 20 K.

Our ultrasonic samples were α-quartz natural crystals irradiated in the RV-1 Reactor at the Instituto Venezolano de Investigaciones Científicas (IVIC) to an integrated dose of $5 \times 10^{18} n/\mathrm{cm}^2$ and the attenuation was measured for longitudinal waves along the x axis and shear waves along the ac axis from 0.4 K to liquid-nitrogen temperatures in the frequency range from 225 to 825 MHz. The samples were too small for thermal measurements but we find that the low-temperature value of the ultrasonic attenuation agrees with that corresponding to sample NIRQ 1 of Ref. 24 irradiated to a dose of $6.6 \times 10^{18} n/\mathrm{cm}^2$. Thus, we take for the thermal conductivity in Eq. (2) the values corresponding to sample N_1 of Ref. 2, sample 27 h of Ref. 3, and sample 4 of Ref. 1, all of which show approximately the same K and were presumably similarly damaged. Figure 1 shows the phonon ultrasonic attenuation calculated at 810 and 825 MHz by means of Eqs. (1) and (2) together with our experimental results. Similar results are found for other lower frequencies in the experimental range with the ratio of the measured to the calculated attenuation becoming progressively larger.

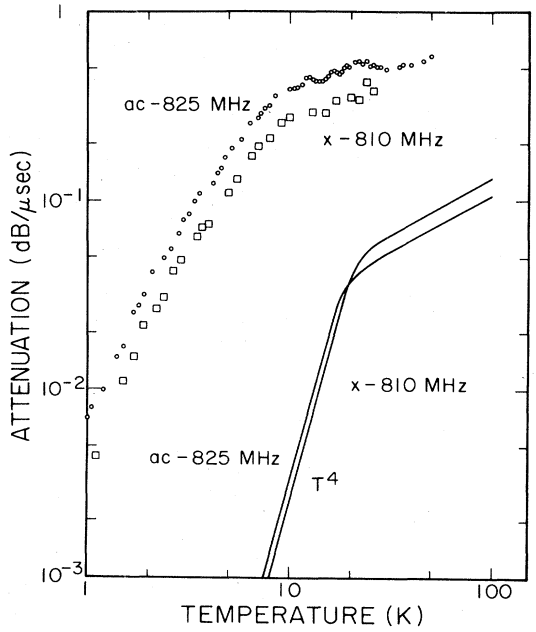

FIG. 1. Ultrasonic attenuation in neutron irradiated quartz for longitudinal waves along the x axis and shear waves along the ac axis. The curves correspond to the calculated phonon contribution from Eqs. (1) and (2), as explained in the text.

As can be observed from comparison with experimental results the calculated phonon viscosity contribution to the high-temperature ultrasonic attenuation in NIRQ may be as much as one order of magnitude smaller than the measured attenuation and can certainly be neglected for frequencies below about 0.5 GHz. The observed attenuation in NIRQ for temperatures above 10 K may be tentatively attributed to the superposition of a discrete spectrum of classical relaxations by thermal activation of radiation induced electronic and structural defects. By extension, it appears that phonon ultrasonic losses may also be disregarded in amorphous quartz at high temperatures except possibly at the higher hypersonic frequencies used in Brillouin scattering determination of the attenuation.[25] Calculated phonon losses in NIRQ are even less important at low temperatures being nearly 2 orders of magnitude smaller than the measured attenuation at 10 K. Thus, the contention that the $\Omega^0 T^3$ attenuation observed in NIRQ at the lower temperatures would reflect the high-frequency part ($\Omega\tau_m > 1$) of the Jäckle relaxation that prevails at high acoustic intensities seems fully justified.

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