

low T_λ for longitudinal waves, it was assumed that the crystal consisted of a single tetragonal domain. However, there could have been multidomain regions near the surface or a few large domains with their tetragonal axes perpendicular to each other. The velocity measurements cited below strongly support the view that the crystal was effectively a single domain along the acoustic path, but the data are not definitive. Even if the crystal does become a single domain, the orientation of the tetragonal axis is not known and could be different each time the crystal is cooled below T_λ .⁹ Depending on the orientation of the tetragonal axis, different thermal-expansion correction factors are needed to calculate the elastic stiffness $c = \rho u^2$ as a function of temperature. There are two possible cases: (a) The tetragonal axis is parallel to the direction of propagation of the sound wave:

$$\rho u^2 = \frac{\rho^0 (2L^0)^2}{\delta^2} \frac{(a_1^0/a_1)^2}{(a_3^0/a_3)} \quad (2)$$

(b) The tetragonal axis is perpendicular to the direction of propagation of the sound wave:

$$\rho u^2 = \frac{\rho^0 (2L^0)^2}{\delta^2} \frac{a_3^0}{a_3} \quad (3)$$

In Eqs. (2) and (3), δ is the round-trip transit time for the acoustic pulse, ρ is the mass density, $2L$ is the round-trip path length in the crystal, a_1 is the lattice parameter of one of the two equivalent axes, and a_3 is that of the tetragonal axis. The superscript zero denotes the values at a convenient reference temperature, which is 20 °C. The values $\rho^0 = 2.4336 \text{ g cm}^{-3}$ and $a_1^0 = a_3^0 = 4.0580 \text{ \AA}$ were taken from Garland and Yarnell.⁴ The values of a_1 and a_3 as functions of temperature were taken from Bonilla, Garland, and Schumaker,¹⁰ since their values fall between those obtained in two independent measurements by Hovi and co-workers.¹¹

The elastic constants obtained from shear-wave measurements are shown in Fig. 3, and those obtained from longitudinal waves in Fig. 4. Smooth-curve values are given in Table II. In the case of shear waves, two independent sets of measurements on the same crystal were carried out under exactly the same conditions except that the Y -cut quartz transducer (and thus the direction of polarization) was rotated by 90°. The elastic constants calculated from these two sets of data were in excellent agreement if it were assumed that the tetragonal axis was parallel to the propagation direction for the first run and perpendicular to it for the second run. If either Eq. (2) or (3) were used for both sets of data, the resulting ρu^2 values showed a systematic difference of about 0.8%. Thus the shear elastic constant measured in this experiment is almost certainly c_{44} . As can

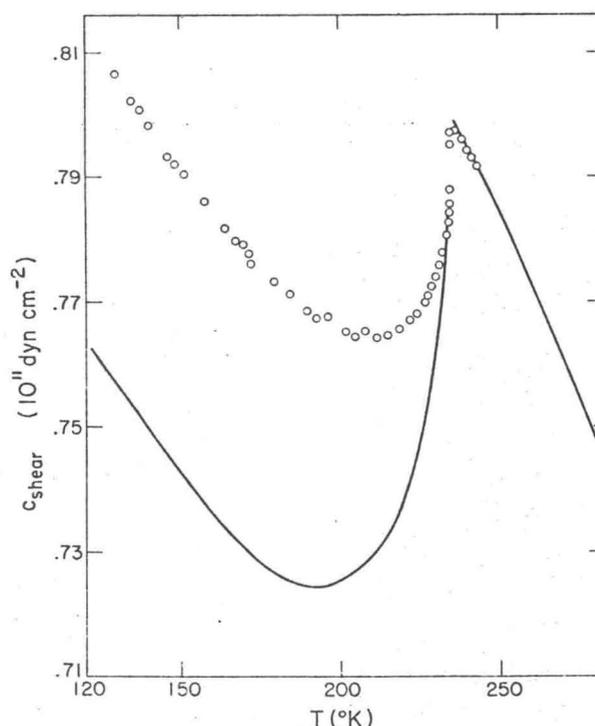


FIG. 3. Temperature dependence of c_{shear} as determined from a "single-domain" crystal in the tetragonal ordered phase. It is probable, but not certain, that these data represent c_{44} values in the ordered phase (see text). The solid line represents the well-characterized variation of c_{44} in the disordered cubic phase above T_λ and the average shear stiffness \bar{c} reported previously from measurements on multidomain ordered crystals (Ref. 4).

be seen from Fig. 3, the variation of c_{shear} is very rapid just below T_λ . Indeed, the value changes from 0.783 at 234 °K to 0.797 at 234.5 °K. It is possible that there is a discontinuous jump in the shear stiffness and hysteresis in the transition temperature, as is seen in NH_4Cl at 1 atm.⁷ However, such effects must be very small if they do occur. We estimate that $\Delta c = 0.032 \times 10^{11} \text{ dyn cm}^{-2}$ and $\Delta T = 40 \text{ m}^\circ\text{K}$ are upper limits on the possible discontinuity and hysteresis.

In the case of longitudinal waves, measurements were made on two different crystals but the orientations must have been fortuitously the same since the ρu^2 values agree on using either Eq. (2) or (3). Thus we do not know which longitudinal elastic constant has been measured in this experiment. It was arbitrarily assumed that the tetragonal axis was parallel to the propagation direction,⁹ and Eq. (2) was used in calculating the c_{long} values in Fig. 4 and Table II. If Eq. (3) had been used instead, the resulting values of c_{long} would all be lower by roughly 0.8%.

The scatter in the data indicates that the random errors in the elastic constants are about 0.1%.