original cubic axes. Such a multidomain crystal scatters sound very strongly, and the echo pattern disappears for longitudinal waves and the c' shear wave. In the present investigation, it was found possible to obtain an essentially single-domain ordered crystal. Thus we were able to measure the velocities of both longitudinal and shear waves and the anomalous attenuation of the longitudinal wave as a function of temperature in the ordered phase.

II. EXPERIMENTAL PROCEDURES

The ultrasonic techniques used in this investigation were very similar to those used previously to study ammonium chloride. Attenuation measurements were carried out with a single-transducer, pulse-echo method using Matec ultrasonic equipment.[6] Velocity measurements were made at 10 MHz with a pulse-superposition method.[7]

A Lauda refrigerating circulator and dry-ice heat exchanger were used for measurements in the disordered phase. Liquid nitrogen and a variable-pressure transfer gas were used for measurements in the ordered phase. In both cases, regulation of the temperature of a copper sample holder was achieved with a Bayley model-250 proportional temperature controller. The resulting temperature stability, as measured with a calibrated Rosemount platinum resistance thermometer, was ± 5 m °K.

The NH_4Br single crystals used in this experiment were from the same batch as those studied in an earlier high-pressure investigation.[1] They were at least 99.9% pure, were free from visible defects, and had well-developed (100) faces which were parallel to within ± 0.0001 cm. Some preliminary measurements in the [100] direction were made using a slightly imperfect crystal with a length $L = 0.7066$ cm. Final measurements in that direction were made on a larger and more perfect crystal, denoted as crystal I. With the exception of attenuation values in the ordered phase, data obtained on these two crystals agreed well with each other. Crystal *Ia* was used as grown for longitudinal attenuation measurements in the disordered phase; the length of this crystal in the [100] direction was 0.8928 ± 0.0001 cm at 20 °C. Subsequently, one face of this crystal was water damaged and a new face was flycut parallel to the undamaged (100) face; the new length was 0.8179 ± 0.0005 cm at 20 °C. This crystal, designated as crystal I*b*, was then used for measurements in the ordered phase. Crystal II was flycut to obtain a pair of (110) faces; the parallelity of these faces was within ± 0.0003 cm and the length along the [110] direction was 0.7562 cm at 20 °C. It should be noted that NH_4Br crystals are soft and hygroscopic and must be handled with considerable care.

The transition temperature T_λ for each crystal was determined by measuring the shear velocity corresponding to the elastic constant c_{44}. This shear wave is not attenuated near T_λ and can even be propagated in a polydomain ordered crystal. Furthermore, this shear velocity undergoes a very rapid variation at the transition. Indeed, the temperature coefficient $(\partial c_{44}/\partial T)_p$ changes abruptly from a small negative value in the disordered phase to a very large positive value, and the transition point is easily established. The λ temperature was found to be (234.3 ± 0.1) °K in crystal I and (234.8 ± 0.1) °K in crystal II. These values agree reasonably with the value 234.5 °K cited by Garland and Yarnell.[4]

Dow resin 276-V9 was used to bond the $\frac{1}{4}$-in.-diam quartz transducers to the NH_4Br crystals. However, this bond broke when the sample was cooled below 160 °K, and the less viscous Dow-Corning 200 silicone fluids were used for measurements in the ordered phase.

It was found possible to obtain an effectively "single-domain" NH_4Br crystal in the tetragonal ordered phase by cooling the crystal slowly through the transition after a temperature gradient has been established along one of the [100] axes. This gradient was obtained by placing a small heating element in good thermal contact with the face opposite from the transducer (which is itself in good thermal contact with the copper sample holder). The optimal gradient, as measured with a gold-chromel difference thermocouple, was 4 to 5 °K cm^{-1}. The gradient heater was turned on and the sample holder was maintained at ~237 °K for several hours to allow the system to achieve a steady-state temperature distribution. The sample holder was then cooled slowly to about 210 °K. The rate of cooling was not crucial, but a constant rate of 4 °K h^{-1} seemed to give the best results. The criterion for deciding whether a single domain had been formed was the ability to propagate longitudinal waves and obtain a good exponential echo pattern with at least five echoes at ~200 °K. Ultrasonic pulses were not propagated in the sample during the cooling process, and the gradient heater was turned off before the echo pattern was inspected. Although there is no direct evidence about the tetragonal domain structure formed below T_λ, the observed ultrasonic behavior is consistent with a single dominant domain and we shall consider the ordered tetragonal phase to be a single-domain crystal. After the gradient is turned off, there is no indication of any change in the domain structure with time or with temperature variations (as long as the temperature is kept below T_λ).