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THE ELASTIC CONSTANTS OF NaCl AT 77·3°K AND 4·2°K

R. Q. FUGATE* and D. E. SCHUELE

Case Institute of Technology, Cleveland, Ohio

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Abstract—The adiabatic elastic constants of NaCl have been measured at 77·3°K and 4·2°K using the ultrasonic pulse echo technique. The results are as follows (in units of 10^{11} dyne cm^{-2}):

	C'_{11}	C'	C_{44}
77·3°K	4·807	2·272	1·335
4·2°K	4·850	2·321	1·337

where $C'_{11} = \frac{1}{2}(C_{11}+C_{12}+2C_{44})$ and $C' = \frac{1}{2}(C_{11}-C_{12})$. These values have been used to calculate the adiabatic bulk modulus and to re-evaluate the low temperature value of the Gruneisen parameter as determined from thermal expansion data and microscopic mode gammas.

INTRODUCTION

ANHARMONICITY in crystals is usually considered in the light of the Quasi Harmonic Model.[1] In using this model, one expands the potential energy in terms of the displacements of the atoms or ions from their equilibrium positions, keeping only terms to second order. By considering the free energy in this approximation and by using thermodynamic relations, one arrives at the Mie–Gruneisen equation of state

$$\gamma_G = \frac{\beta B_s}{C_p/V}, \tag{1}$$

where γ_G is known as the Gruneisen parameter [defined by equation (1)], β is the volume coefficient of thermal expansion, B_s is the adiabatic bulk modulus, and C_p/V is the heat capacity per unit volume. The Quasi Harmonic Model also leads to a value of γ_G in terms of the microscopic parameters γ_i defined by

$$\gamma_i \equiv -\frac{d \ln \omega_i}{d \ln V}, \tag{2}$$

where the ω_i depend only on the volume and correspond to the normal mode frequencies. Defined in this way the γ_i are a measure of the anharmonicity of a crystal.

The calculation of γ_G in the region of low temperature requires a precise determination of not only β and C_p but also B_s. WHITE[2] has recently determined β for NaCl from which he calculates γ_G (denoted as γ_0 in his work) in the T^3 region of temperature and in this calculation has used the value of B_s as reported by OVERTON and SWIM.[3] A plot of Overton and Swim's B_s vs. temp. leads one to believe that their values are low. Furthermore, the corresponding low temperature gamma reported by BARTELS and SCHUELE,[4] $\bar{\gamma}_L$ (calculated from the γ_i) is some 17% greater than White's γ_0. Hence this work was undertaken to measure the elastic constants of NaCl at very low temperatures and thereby redetermine B_s. Furthermore, it was of interest to test the supposition of Bartels and Schuele that a change of the value of B_s at low temperatures would not appreciably change their calculated value of the low temperature $\bar{\gamma}_L$. Finally, it was considered advantageous to determine low temperature elastic data on samples from the same parent crystal that provided samples for the pressure data collected by Bartels and Schuele.

EXPERIMENTAL PROCEDURE

Sample preparation

Two NaCl single crystals were prepared from one-in. cleavage cubes obtained from the Harshaw Chemical Company. One of the crystals was oriented with its ends normal to the [110] direction by mounting the cleavage

* Now at Iowa State University, Ames, Iowa.

cube in an appropriate steel fixture and lapping the protruding edges with water on a felt covered motor driven turntable. The other crystal was oriented with its ends normal to the [100] direction and required no special orientation fixture since the cleavage cube was of the desired orientation. After rough shaping, both crystals were mounted in steel lapping rings, and the end surfaces were ground flat and parallel on fine grit emery paper backed by plate glass. The final shapes of the two crystals were approximate right circular cylinders with diameters of about 1·7 cm for the [110] sample and 1·3 cm for the [100] sample and with lengths (at 22°C) of 1·9068 cm and 2·2873 cm, respectively.

Measurements

Figure 1 shows the assembly used to hold the NaCl samples for low temperature measurements. This assembly, surrounded by a stainless steel jacket not

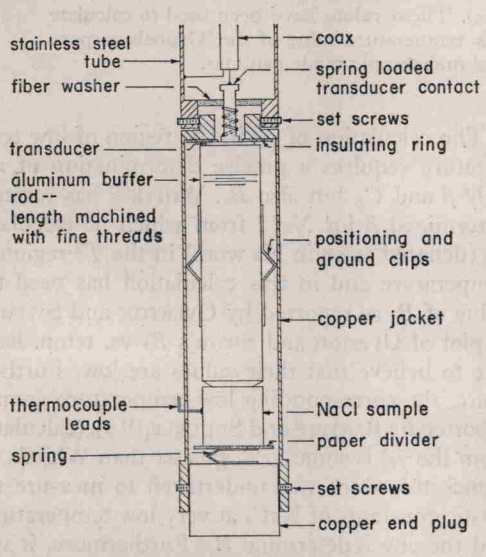

FIG. 1. Arrangement used to hold NaCl samples for low temperature measurements.

shown, was held in a double jacketed dewar, the inner jacket being equipped with a special cap so that the cryogenic liquid could be introduced slowly, thus permitting a slow rate of cooling (about 2°K per min from 295°K to 77·3°K and about 1°K per min from 77·3°K to 4·2°K). The temperature of the sample was determined by means of a copper–constantan thermocouple referenced at 0°C in an air saturated ice bath. A Leeds and Northrup potentiometer Type K-2 and a Leeds and Northrup D-C Null Detector model 9834 were used to measure the thermocouple voltage.

The fundamental measurements involved in this work were the transit times of 10 Mc/s acoustic pulses. (Transit time is defined as the time required for the pulse to traverse the length of the sample twice.) The pulse echo method was used, and a good description of measurements of this kind may be found in earlier papers from this laboratory.[5] The system used in the present work was based on a Tektronix type 547 variable delay oscilloscope equipped with a Type 1A1 plug-in unit. The oscilloscope was modified by replacing the sweep delay helipot by a more linear (±0·015% linearity) helipot in combination with external resistance boxes. In this way, any fraction of the total sweep time could be put on the helipot, which allowed a more sensitive measurement of the time between successive echoes. The portion of the helipot used for each run was calibrated after each run with one microsecond time marks from a Tektronix TM180A time mark generator. The 10 Mc/s pulses were generated by an Arenberg model PG-650c pulse generator, and reflected signals were first amplified with an Arenberg model PA-260 preamplifier before being displayed on the oscilloscope.

An aluminum buffer rod (shown in Fig. 1) was used in all measurements. Glycerin was found to be an ideal seal material between the transducer and aluminum rod. A mixture of 50% isopentane and 50% rubber cement solvent was found to be a satisfactory seal material for the NaCl crystals. This 50–50 mixture begins to solidify (in the form of a glassy state) at about 90°K and undergoes all major phase changes by 77·3°K. The 50–50 mixture is very volatile at room temperature and thus an airtight seal of rubber cement was painted around the end of the NaCl crystal where it was in contact with the aluminum buffer rod. Rubber cement was tried once as a seal material on the [100] crystal. However, in cooling down from 77·3°K to 4·2°K, the strain caused by differential contraction cleaved the [100] crystal rather seriously. On no occasion did the 50–50 mixture seals cause cleaving of the NaCl crystals.

In measuring the transit times, only the first three echoes were used. The first echo is referred to as the first buffer echo since it comes from the buffer rod–NaCl crystal interface. The next two echoes (the first and second crystal echoes) come from transmissions through the crystal. These three echoes should be spaced at intervals of the transit time of the sample under investigation. The transit time was measured by recording the reading of the helipot for several cycles of one echo and then recording the helipot readings of the corresponding cycles in the next echo. In order to insure that the corresponding cycles were measured, special double exposure photographs were taken. This was done by aligning a particular cycle with a vertical line on the oscilloscope screen, making an exposure, and then aligning what was thought to be the corresponding cycle of the next echo with the same vertical line on the oscilloscope and then making another exposure without moving the film. The amplitudes of the corresponding cycles were set equal by means of the variable gain on the oscilloscope. If the corresponding cycle was chosen correctly the photograph showed two echo patterns almost exactly superimposed upon one another. It was clearly evident, however, if the corresponding cycle was chosen improperly because of the large amplitude discrepancies between the corresponding cycles of the two echoes. This photographic method has also proven