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The Adiabatic Elastic Constants of Rock Salt*

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The velocities of propagation of 10-Mc/sec longitudinal and transverse sound waves in single crystals of rock salt have been measured over the temperature range from 60°K to 300°K by means of the ultrasonic pulse method. From these velocities the adiabatic elastic constants have been calculated, with the results that $c_{11}=4.828\pm0.015$, $c_{44}=1.273\pm0.005$, and $c_{12}=1.276\pm0.025$ at 300°K in units of dyne/cm$^2\times10^{11}$. The values of these elastic constants extrapolated to absolute zero temperature are, in these units, 5.750, 1.327, and 0.986, respectively. The adiabatic compressibility has been calculated from c_{11} and c_{12} and is 4.07×10^{-12} cm^2/dyne at 300°K. The Debye characteristic temperature is computed to be 292.6±0.5°K at $T=0$°K.

INTRODUCTION

THE ultrasonic pulse method has been used to measure the velocity of 10-Mc/sec sound waves in single crystals of rock salt at temperatures from 60°K to 300°K. From the knowledge of the velocity of propagation of longitudinal and transverse waves in the medium, the adiabatic elastic constants c_{11}, c_{44}, and c_{12} may be calculated. The ultrasonic pulse method of determination of mechanical properties of solids,[1-3] made practicable by the development of electrical circuits for pulsing, has certain advantages over the previously used methods.[4,5] Perhaps the most important of these is the fact that the velocity of propagation of sound waves in the medium may be calculated directly from experimentally observed quantities. Since the piezoelectric quartz crystal employed to generate the acoustic pulses is many wavelengths in diameter at high frequencies, the acoustic waves propagated by the crystal are essentially traveling plane wave pulses. Using an oscilloscope such as the Du Mont 256-D, direct measurement of the time required for the pulses to travel twice the length of the specimen is possible, which allows immediate calculation of the wave velocity. From this velocity and the density of the medium the elastic constants may be calculated.

EXPERIMENTAL

The specimens of NaCl employed are single crystals, free from flaws and impurities, obtained from the Harshaw Chemical Company, and are in the form of cylinders approximately three centimeters in diameter. Two rock salt crystals are used in the experiments: one cut so that the end faces of the cylinder are parallel to the (0, 0, 1) crystalline plane, the other with end faces parallel to the (0, 1, 1) plane, where 0 and 1 are Miller indices. The lengths of the crystals are about 6.6 and 12.7 cm, respectively. The advantage in the use of single crystals is that no extraneous effects are introduced due to scattering and reflections of the sound pulses by the grain structure present in polycrystalline specimens. The end faces of the specimens have been carefully ground and polished so that they are both flat and parallel; the lengths of the specimens are thus constant over the area of the faces to ±0.0004 inch. Furthermore, any focusing of the sound beam caused by curvature of the reflecting faces is minimized.

It has been determined experimentally that the quartz transducer must be physically bonded to the specimen in order to obtain the intelligible echoes necessary for precise velocity measurements. If the quartz is merely pressed against the end face of the specimen, the contact is not sufficiently good to allow transmission of the acoustic pulse to the specimen without undesirable pulse distortion. In the measurements on rock salt the choice of the cement used to bind the quartz to the specimen must be made according to the temperature range over which the measurements are to be made. The reason for this is that as the temperature is lowered, differential thermal contraction between the quartz, the binder, and the rock salt may result in one of two effects, either of which renders further measurements impossible. The first of these is the cracking of the binding layer, so that the quartz is no longer in contact with the rock salt. The second is that stresses introduced in the quartz crystal may deform it and hence change its piezoelectric properties, with the result that intelligible echoes are no longer received. Both phenomena have been observed, and in one instance the thermal stress was sufficiently large to cleave the specimen very slightly.

It has been found that ordinary stopcock grease serves as an adequate binder for the quartz transducer in the temperature range from room temperature to 160°K, but below this temperature the binding layer cracks because of thermal stresses. For measurements below 160°K an organic mixture,[6] consisting of ethyl ether, ethyl alcohol, and isopentane, has been employed as a binder. This mixture is applied at a temperature

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[1] H. B. Huntington, Phys. Rev. **72**, 321 (1947).

[2] J. K. Galt, Phys. Rev. **73**, 1460 (1948).

[3] W. C. Overton, Jr., J. Chem. Phys. **18**, 113 (1950).

[4] L. Balamuth, Phys. Rev. **45**, 715 (1934).

[5] F. C. Rose, Phys. Rev. **49**, 50 (1936).

[6] G. N. Lewis and M. Kasha, J. Am. Chem. Soc. **66**, 2100 (1944).

of about 125°K, and with it measurements may be made from 110°K to 60°K. The substitution of pentane for isopentane in this organic binder allows measurements to be made at temperatures as high as 140°K, although the thermal contraction of the medium is greater at lower temperatures than that of the mixture using isopentane. At temperatures below about 50°K the differential contraction between the transducer, the binder, and the rock salt is sufficient to deform physically the transducer. The result of this is a radical alteration in the electrical and acoustical properties of the quartz crystal, and the pulses are so distorted that accurate measurements are impossible. At the present time no binder has been found which will allow measurements to be carried out below this temperature on rock salt.

In the determination of the round-trip travel time of the pulses in the specimen some ten readings are made of the position in time of the leading edge of each echo occurring in the first 100 microseconds after the initial pulse, and an average taken. Then the differences between the times of occurrence of successive echoes give the desired travel time, which in general has an average deviation of ±0.03 microsecond. It is to be noted that although the acoustic pulses may be reflected at grain boundaries in the binder and at the faces of the transducer as well as at the faces of the specimen, measurements at the leading edges of the echoes insure that the distance travelled by the pulses, corresponding to the measured time, is wholly within the specimen.[7] Temperatures are measured with a copper-constantan thermocouple and a helium gas constant volume thermometer. Although there are temperature gradients during measurements at temperatures above 80°K, the rate of cooling of the specimen may be regulated so that the temperature is constant to within one degree during a given measurement. These temperature gradients were not sufficiently large to cause any noticeable pulse distortion. Figure 1 shows the cryostat used in these measurements. The Dewar flask A surrounds the apparatus and serves both as a container for the liquid refrigerant and as a shield against external thermal radiation. In addition another Dewar flask surrounds flask A and is filled with liquid nitrogen for refrigeration purposes. The specimen is held in the brass can B, and layers of paper are used as a sound absorbing medium between the specimen and the can. The rock salt crystal may be raised and lowered and rotated by means of the stainless steel rod C. The leads of the copper-constantan thermocouple thermometer D are soldered to the can, giving good thermal contact with the specimen. The glass capillary E is used for the application of the quartz-to-specimen binder for temperatures below 140°K, and may be rotated from above. The rf pulses to the quartz crystal F are brought down by the coaxial line G, which consists of a stainless steel tube and an inner copper conductor. The electrical contacts to the transducer are the spring-loaded plunger H and the stack of steel disc springs supporting a brass contact I. J is the bulb of the helium gas constant volume thermometer.

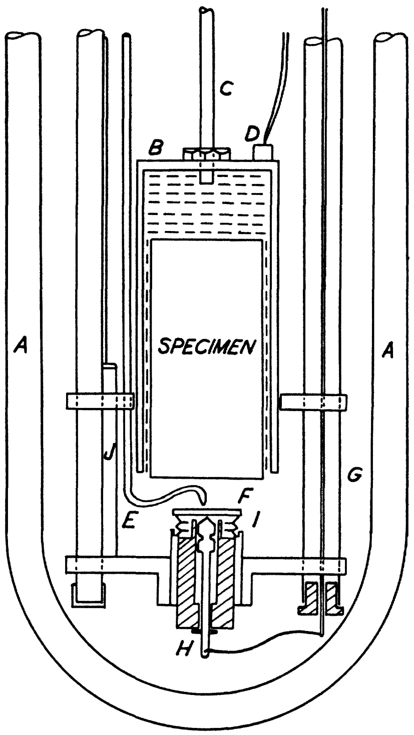

FIG. 1. Cryostat used for low temperature ultrasonic measurements.

In order to calculate the velocity of propagation of elastic waves in a solid, the length of the specimen must be known accurately. This in turn requires knowledge of the temperature dependence of the linear coefficient of thermal expansion, which in the case of NaCl has not been determined experimentally over the temperature interval under consideration. However, it may be calculated from available data in two independent ways. Also the temperature dependence of the density of rock salt must be known in order to calculate the elastic constants. Grüneisen[8] has shown that the ratio α/c_p where α is the linear coefficient of thermal expansion and c_p the specific heat at constant pressure, is independent of temperature for many solids. Using published values of α and c_p at 0°C, α has been calculated from the temperature variation of c_p.[9] From this knowledge of α the volume coefficient of thermal expansion is determined, which allows calculation of the temperature dependence of the density, assuming a

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[7] In this connection Lazarus (see reference 13) reports the necessity of a correction to the observed travel time. However, Huntington (see reference 1) maintains that according to the above discussion no such correction is necessary.

[8] E. Grüneisen, Ann. Physik **26**, 211 (1908).

[9] *International Critical Tables* (McGraw-Hill Book Company, Inc., New York, 1928), Vol. 3, p. 43.

TABLE I. Velocity of 10-Mc/sec sound waves in rock salt. The subscripts l_1, t_1, and l_2 denote longitudinal waves normal to the (0, 0, 1) plane, transverse waves normal to the (0, 0, 1) plane, and longitudinal waves normal to the (0, 1, 1) plane, respectively. The units are cm/sec×10^5.

T	c_{l_1}	c_{t_1}	c_{l_2}
60	5.086	2.447	4.602
70	5.081	2.448	4.596
80	5.070	2.449	4.590
90	5.056	2.450	4.584
100	5.042	2.450	4.578
110	5.027	2.450	4.572
120	5.012	2.448	4.568
130	4.997	2.445	4.562
140	4.980	2.441	4.556
150	4.965	2.438	4.550
160	4.949	2.435	4.544
170	4.933	2.433	4.538
180	4.917	2.432	4.532
190	4.901	2.431	4.526
200	4.885	2.430	4.520
210	4.869	2.429	4.514
220	4.853	2.428	4.508
230	4.837	2.428	4.502
240	4.822	2.427	4.498
250	4.806	2.427	4.492
260	4.790	2.427	4.488
270	4.774	2.427	4.483
280	4.758	2.427	4.479
290	4.741	2.427	4.476
300	4.726	2.427	4.473

cube of unit volume at 0°C. The second method of calculation employs the use of the empirical relation between density and temperature for NaCl,[10] which is

$$\rho(t)=2.1680(1-1.12\times10^{-4}t-5\times10^{-8}t^2),$$

where t represents the centigrade temperature. The density is calculated directly from this, and upon application of the above relation to a unit cube of rock salt at 0°C the linear coefficient of thermal expansion may be calculated. In either case the length is computed by numerical integration of the relation

$$L(T)=L(T_0)\left[1+\int_{T_0}^{T}\alpha(T)dT\right].$$

The values of the lengths of the specimens obtained by these two independent methods differ by not more than one part in 3000.

TABLE II. Adiabatic elastic constants and compressibility of rock salt. The units of the elastic constants are dyne/cm^2×10^{11} and of the compressibility are cm^2/dyne×10^{-12}.

T	c_{11}	c_{44}	c_{12}	k_s
0	5.750	1.327	0.986	3.89
60	5.730	1.327	0.986	3.90
70	5.713	1.327	0.986	3.91
80	5.685	1.327	0.986	3.92
90	5.649	1.326	0.986	3.94
100	5.613	1.325	0.992	3.95
110	5.575	1.324	1.001	3.97
120	5.535	1.321	1.017	3.97
130	5.496	1.316	1.034	3.97
140	5.456	1.310	1.052	3.97
150	5.415	1.306	1.069	3.97
160	5.376	1.302	1.082	3.98
170	5.336	1.298	1.098	3.98
180	5.296	1.295	1.110	4.00
190	5.256	1.293	1.122	4.00
200	5.216	1.290	1.134	4.01
210	5.177	1.288	1.143	4.02
220	5.137	1.286	1.155	4.03
230	5.097	1.284	1.169	4.05
240	5.059	1.282	1.181	4.05
250	5.020	1.281	1.190	4.06
260	4.981	1.279	1.205	4.06
270	4.942	1.277	1.222	4.07
280	4.904	1.276	1.236	4.07
290	4.864	1.275	1.258	4.07
300	4.828	1.273	1.276	4.07
Uncertainty:	±0.015	±0.005	±0.025	±0.03

RESULTS

The velocity of propagation of the sound waves is calculated for each of the two determinations of the length, and the curves of the experimental results are smoothed. Table I gives the velocities measured in this experiment, determined by the algebraic mean of the smoothed values calculated by each of the above methods.

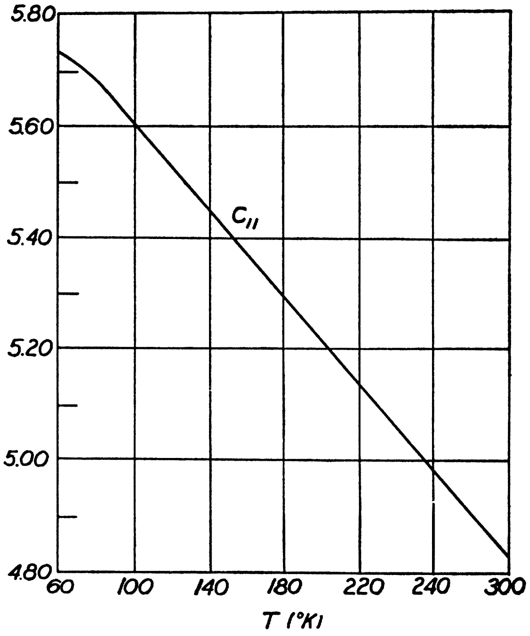

FIG. 2. Elastic constant c_{11} of rock salt in units of dyne/cm^2×10^{11}.

In the case of a cubic crystal such as NaCl rotational and reflectional symmetries require that there be only three independent, nonzero elastic constants, which are c_{11}, c_{44}, and c_{12}. It may be shown[11] that the following relations hold for cubic crystals, where ρ is the density and c the wave velocity:

longitudinal waves normal to the (0, 0, 1) plane,

$$\rho c^2=c_{11};$$

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[10] A. Henglein, Z. physik Chem. **115**, 97 (1925).

[11] A. E. H. Love, *The Mathematical Theory of Elasticity* (Dover Publications, Inc., New York, 1944), pp. 295–299.

transverse waves normal to the (0, 0, 1) plane,

$$\rho c^2 = c_{44};$$

longitudinal waves normal to the (0, 1, 1) plane,

$$\rho c^2 = \tfrac{1}{2}(c_{11} + c_{12} + 2c_{44}).$$

In the calculation of the elastic constants one value of ρc^2 is determined from the density and velocity calculated according to Grüneisen's theory and another from the empirical relation for the density, and an average of the two is taken. These two values of ρc^2 never differ by more than one part in 2500, which is well within the total uncertainty of 0.4 percent in the values of this quantity. Table II gives the values of the adiabatic elastic constants as functions of temperature, and also the adiabatic compressibility, which is

$$k_s = 3/(c_{11} + 2c_{12}).$$

The greater uncertainty in c_{12} is due to the contributions of the uncertainties of c_{11} and c_{44}. The values of these quantities at absolute zero are determined by extrapolation of the results at higher temperatures. Figures 2 and 3 give the plots of the elastic constants as functions of temperature.

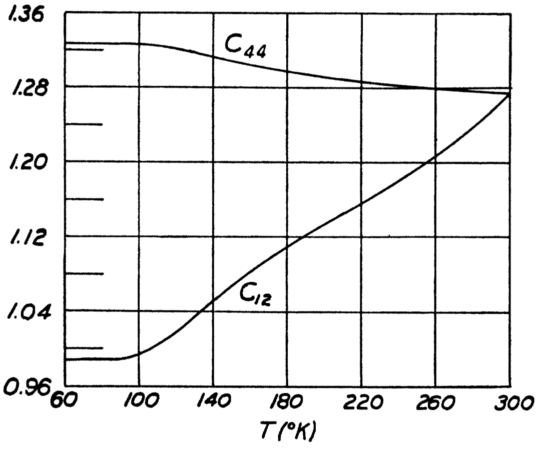

FIG. 3. Elastic constants c_{44} and c_{12} of rock salt in units of dyne/cm^2×10^{11}.

DISCUSSION

The adiabatic elastic constants of rock salt have been determined previously in this temperature range by Rose[5] and Durand,[12] both of whom used the composite oscillator method. Also these quantities have been measured at room temperature by Huntington,[1] Galt,[2] and Lazarus,[13] using the ultrasonic pulse technique. A comparison of the values obtained here with these previous results at room temperature is given in Table III. It is to be noted that in the present work c_{12} is smaller than c_{44}, except at room temperature, showing the invalidity of the cauchy relations, which require the equality of these two elastic constants in a cubic lattice at all temperatures.

TABLE III. Elastic constants of rock salt measured at room temperature in units of dyne/cm^2×10^{11}.

	c_{11}	c_{44}	c_{12}
Overton and Swim	4.86	1.27	1.28
Huntington	4.85	1.26	1.23
Galt	4.87	1.26	1.24
Rose	4.96	1.27	1.30
Durand	4.99	1.27	1.31
Lazarus	4.91	1.28	1.23

It is evident from the experimental results that the elastic constants become independent of temperature at a temperature depending upon the particular elastic constant. It is to be expected that c_{ij} as a function of temperature should have zero slope at $T = 0$°K as can be seen from the strain-energy function and the Nernst theorem of thermodynamics. A more detailed consideration of the strain-energy function shows that $\partial c_{ij}/\partial T$ may vanish at a temperature considerably greater than zero, dependent upon the temperature variation of strain in the crystal. It follows that the temperature at which $\partial c_{ij}/\partial T$ becomes zero is not necessarily the same for all of the elastic constants, as is also shown by the experimental results. Consideration of the strain-energy function also leads to the possibility of the existence of inflection points in the curves of the elastic constants *vs* temperature, which are experimentally found to occur. The hump in the curve of c_{12} *vs* temperature in the region between 120° and 220°K, although generally within the experimental error of the measurement, is due to the inflection of c_{44} in this temperature region. In the relation used to calculate c_{12}, $c_{12} = 2\rho c^2 - c_{11} - 2c_{44}$, ρc^2 and c_{11} are linear functions of temperature over this interval, whereas c_{44} undergoes the abovementioned inflection.

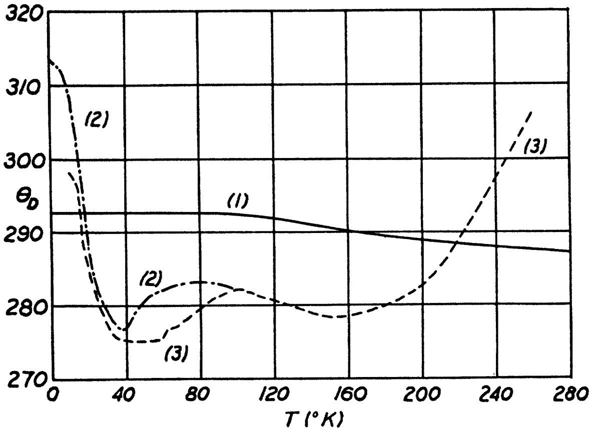

FIG. 4. Debye characteristic temperature of rock salt. Curve (1) is due to Overton and Swim, (2) to Kellermann, and (3) to Clusium, Goldmann, and Perlick.

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[12] M. A. Durand, Phys. Rev. **50**, 449 (1936).

[13] D. Lazarus, Phys. Rev. **76**, 545 (1949).

From the values of the elastic constants obtained here the Debye characteristic temperature θ_D has been computed as a function of temperature, and as is to be expected, is constant over a wide temperature range. The computed values of θ_D are shown in Fig. 4, together with the results obtained by Kellermann[14] from a theoretical analysis of the NaCl lattice vibrations and with those obtained by Clusius, Goldmann, and Perlick[15] from specific heat measurements. Comparison of the three curves shows that the inadequacy of the Debye theory in providing an accurate description of the properties of the crystal may be attributed to the choice of the distribution function for the frequencies, rather than to anomalous behavior of the elastic constants. The value of θ_D at absolute zero temperature is 292.6±0.5°K.

ACKNOWLEDGMENT

The authors are indebted to Professors W. V. Houston and C. F. Squire for helpful discussions concerning this work.

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[14] E. W. Kellermann, Trans. Roy. Soc. (London) **A238**, 513 (1940); Proc. Roy. Soc. (London) **A178**, 17 (1941).

[15] Clusius, Goldmann, and Perlick, Z. Naturforsch. **4A**, 424 (1949).

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Scattering of Protons by Alpha-Particles*

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Differential scattering cross sections for 5.10±0.10-Mev protons by helium have been determined at laboratory angles from 30° to 150°. Probable errors in the cross sections varied from 2.4 percent at 30° to 4.1 percent at 150°. The scattered protons were detected by a proportional counter. The apparatus was calibrated by a study of proton-proton scattering.

I. INTRODUCTION

The scattering of protons by helium has previously been studied at energies up to 3.6 Mev.[1] In the present work the scattering of 5.10-Mev protons by helium has been studied at laboratory angles of 30°, 45°, 60°, 75° 58′, and 150°. The effect of a slight variation in energy on the scattering cross section at 30° was investigated.

The determination of absolute scattering cross sections is usually complicated by difficulties in measuring certain geometrical constants of the apparatus. In the present work, moreover, the capacitance of a standard condenser and associated circuit used in integrating the beam current presents a problem in measurement. These uncertain constants were here lumped into an "apparatus factor." This apparatus factor was then evaluated by an experiment on proton-proton scattering. The best value for the proton-proton scattering cross section based on all the available experimental data was used for this calibration.

II. THE SCATTERING APPARATUS

A schematic diagram of the scattering apparatus is presented as Fig. 1.

The beam from the Washington University cyclotron entered the apparatus through a collimator consisting of two 4-mm diameter slits separated from one another by about 34 cm. Four 6-mm diameter baffles were spaced at approximately equal intervals between the slits. A window of 0.25-mil rubber hydrochloride separated the scattering apparatus from the cyclotron.

The unscattered particles were collected by a faraday cage. The faraday cage, maintained at a pressure of about 4×10^{-5} mm of Hg, was separated from the scattering gas by a window of 0.5-mil polyethylene.[2] The charge was accumulated on a 1μf polystyrene condenser made by John E. Fast and Company of Chicago, Illinois. This condenser showed no appreciable leakage or polarization. The voltage across the condenser was measured by a null method using a Compton electrometer as the null indicator. The error in the charge measurement was estimated to be ±0.2 percent, the largest part of this error being due to slight drifts in the electrometer zero.

The scattered protons were defined by a slit system consisting of two 2-mm×4-mm slits spaced about 8.5 cm apart. The distance from the center of the scattering volume to the second slit, the counter slit, differed slightly at the various angles. All the data were normalized to a distance of 16.15 cm. Uncertainty in this distance contributed an estimated ±0.6 percent error to the final cross sections. The proton-proton

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* Assisted by the joint program of the ONR and AEC.

† Now at Georgia School of Technology, Atlanta, Georgia.

[1] Freier, Lampi, Sleator, and Williams, Phys. Rev. **75**, 1345 (1949); N. P. Heydenburg and N. F. Ramsey, Phys. Rev. **60**, 42 (1941).

[2] The 0.5-mil and 0.25-mil polyethylene used in this work was kindly furnished by the Du Pont Company.