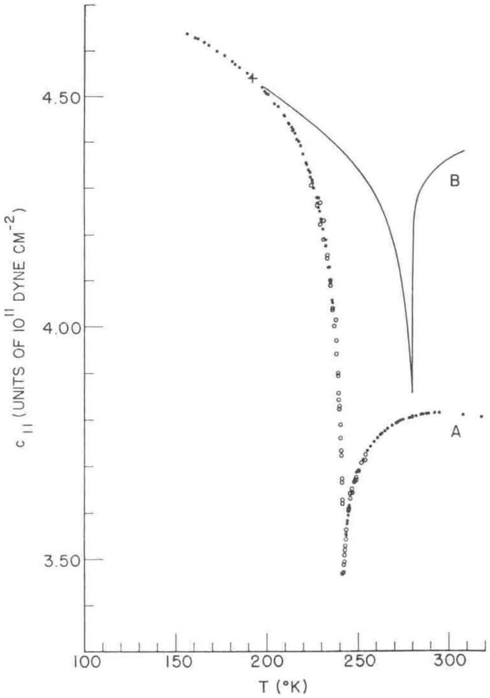

FIG. 4. Variation of c_{11} with temperature. Curve A: data at one atmosphere (solid circles are present measurements, open circles are corrected pulse-echo data[6]). Curve B: calculated curve at constant volume $V_2=34.15$ cm^3 mole^{-1}; V_2 corresponds to V_λ at 280°K and to the volume at 1 atm and 191.25°K (marked by cross).

hysteresis was observed in the critical region. Both c_{44} and C' increased gradually as the temperature was decreased until at 241.4°±0.1°K they suddenly showed a very large increase. At this temperature a new equilibrium was reached only after waiting for about 45 min. During this time c_{44} had suffered a jump of about +3.6% and C' a jump of about +1.2%. On further cooling the variation of these elastic constants was again smooth and gradual. Starting from a low temperature (say 220°K) and warming the sample, an abrupt drop in the values of c_{44} and C' occurred at 242.3°±0.1°K, and a new equilibrium was again reached only after waiting for about 45 min. Below 241.4°K and above 242.3°K experimental values of the elastic constants were the same on cooling and on warming. This hysteresis loop has not been represented on Figs. 5 and 6 because the scale is too small to allow a clear representation of the phenomenon. The strong attenuation of longitudinal waves in the critical region prevented any such observation for c_{11}.

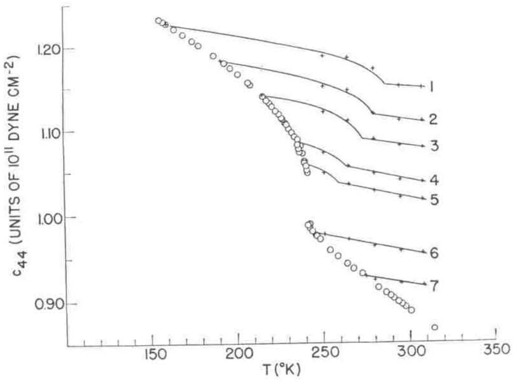

FIG. 5. Variation of c_{44} with temperature. Open circles are experimental data at 1 atm. Curves 1, 2, ···, 7 are calculated for various constant volumes: $V_1=34.002$; $V_2=34.150$; $V_3=34.266$; $V_4=34.428$; $V_5=34.507$; $V_6=34.768$; $V_7=34.928$ cm^3 mole^{-1}.

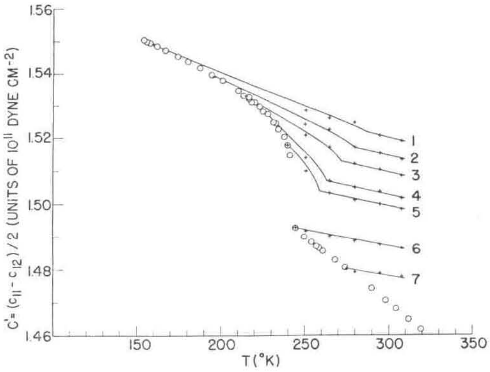

FIG. 6. Variation of C' with temperature. See legend of Fig. 5.

Presented in Table I is a tabulation of smooth-curve values of the experimental results at various temperatures in the range covered in these experiments. Note that values are given at 242.5°K rather than 242°K to avoid the region very close to the critical point which corresponds to a region of hysteresis.

An error analysis indicates that the maximum *random* error in these directly measured elastic quantities is 0.05% at room temperature. The error may be slightly

TABLE I. Smooth-curve values at one atmosphere for the adiabatic elastic constants c_{11}, c_{44}, and $C'=(c_{11}-c_{12})/2$, in units of 10^{11} dyn cm^{-2}. The adopted values of $L(T)/L(296°\text{K})$ are also listed. The c_{11} entries marked by an asterisk are based on corrected data from a previous pulse-echo investigation.[6]

T(°K)	$L(T)/L(296°\text{K})$	c_{11}	c_{44}	C'
155	0.98975	4.6402	1.2316	1.5499
170	0.99043	4.6086	1.2117	1.5462
190	0.99136	4.5473	1.1833	1.5411
210	0.99231	4.4532	1.1499	1.5349
230	0.99353	4.2235	1.1016	1.5260
250	0.99767	3.6892	0.9669	1.4902
270	0.99864	3.7890	0.9328	1.4819
290	0.99967	3.8138	0.9030	1.4738
310	1.00078	3.8117	0.8728	1.4657
236	0.99422	4.055	1.0804	1.5218
237	0.99437	4.008*	1.0758	1.5209
238	0.99452	3.955*	1.0712	1.5198
239	0.99468	3.887*	1.0656	1.5185
240	0.99487	3.813*	1.0592	1.5171
241	0.99508	3.734*	1.0515	1.5152
242.5	0.99724	3.507*	0.9830	1.4937
243	0.99728	3.538*	0.9818	1.4935
244	0.99734	3.582*	0.9796	1.4930
245	0.99739	3.609	0.9775	1.4925

larger at temperatures below the lambda point due to an increased uncertainty in the path–length correction. To check this figure we measured the quantity $(c_{11}+c_{44}-C')$ by exciting a longitudinal wave in the [110] direction; see Eq. (4). The experimental data were compared to the values calculated from the directly-measured elastic constants. The values agreed almost exactly at room temperature. For lower temperatures, the difference was always less than 0.16% in the disordered phase and it never exceeded 0.25% in the ordered phase.

The estimation of systematic errors is more difficult. In the pulse-superposition method an error of $|\Delta n|=1$ in the attribution of the n values to the in-phase conditions would lead to systematic errors greater than 1% in the elastic constants. We tried to prevent such mistakes by using several crystals of different lengths. With Crystals A and B we obtained excellent agreement (within the limits of random error) for the c_{11} values at room temperature. With Crystals B and C the agreement of c_{44} values was not as good: a relative deviation $[c_{44}(\mathrm{B})-c_{44}(\mathrm{C})]/c_{44}(\mathrm{B})=0.4\%$ was observed over the entire range of temperatures. The cause of this disagreement is unknown; it is not due to a misalignment of the crystal axes, a wrong attribution of the n values, or to diffraction effects. We have reported the values obtained with Crystal B since it had natural (100) faces and had not been cut or polished, but these c_{44} values may possible be systematically high by 0.4%.

TABLE II. The adiabatic elastic constants of ammonium chloride single crystals obtained from the present measurements (P) compared with the results obtained by Garland and Jones (G and J), by Haussuhl (H), and by Roa and Balakrishnan (R and B). All values are given in units of 10^{11} dyn cm^{-2}.

Obs.	T(°K)	c_{11}	c_{44}	C'
P	300	3.815	0.8878	1.4698
G and J	300	3.70	0.86	1.41
H	293	3.79	0.83	1.41
R and B	298	3.90	0.68	1.59
P	200	4.507	1.1674	1.5382
G and J	200	4.354	1.122	1.480

The adiabatic elastic constants of single-crystal ammonium chloride have been measured at room temperature by Haussuhl[20] and by Roa and Balakrishnan.[21] They have also been measured over the same range of temperatures as in the present work by Garland and Jones.[6] Table II gives a comparison of the values obtained by these various investigators with the results of the present experiments. It is striking that the present results are systematically 3% to 4% higher than those of Garland and Jones over the entire range of temperatures investigated. They used a pulse-echo method with unrectified pulses and determined the delay times by ranging the position of the rf peaks for a set of successive echoes. An examination of their data indicates that a systematic change in the delay times by a small number of rf periods would explain the discrepancy in most cases.

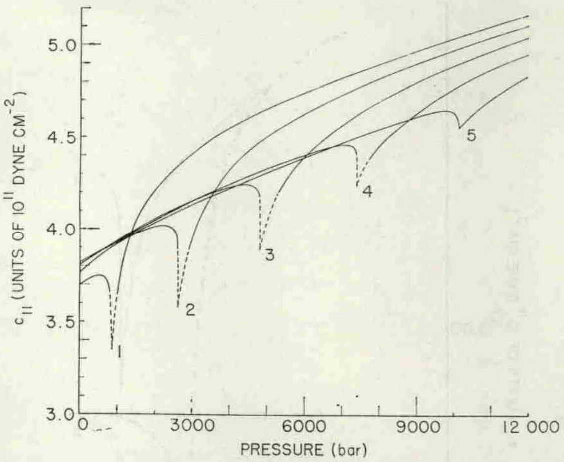

FIG. 7. Dependence of c_{11} on pressure at various temperatures (see Fig. 3 legend). Dashed portions of the curves indicate regions where data are less accurate or are missing due to high attenuation.

Constant-Temperature Data

Shown in Figs. 7, 8, and 9 are the pressure dependences of the effective adiabatic elastic constants c_{11}, c_{44}, and C'. These curves were calculated[15] from the known elastic constants at 1 atm, the adopted path length ratio s (see Fig. 3), and the experimental ratio N_p/N_1, where N_p is the $n=0$ repetition rate under an applied pressure p and N_1 is the value at 1 atm. All the data at high pressure were obtained at 20 Mc/sec. Although data points are not shown on these figures, each isotherm is based on at least 30 experimental points which show very little scatter. A tabulation of the smooth-curve values of experimental results over the range 0 to 12 kbar is presented in Table III.

Because of the strong attenuation of longitudinal waves in the critical region, it was not possible to

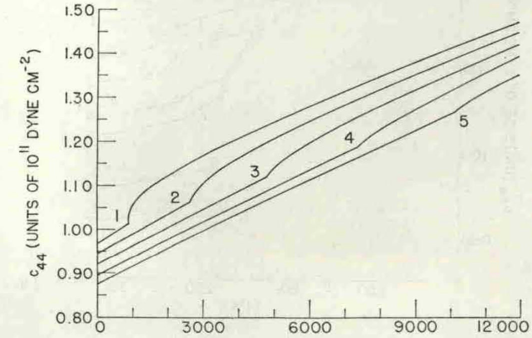

FIG. 8. Dependence of c_{44} on pressure at various temperatures (see Fig. 3 legend).

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[20] S. Haussuhl, Acta Cryst. **13**, 685 (1960).

[21] R. V. G. Sundara Roa and T. S. Balakrishnan, Proc. Indian Acad. Sci. **A28**, 480 (1948).