

## VII. COOPERATIVE PHENOMENA IN SOLIDS AND FLUIDS\*

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### RESEARCH OBJECTIVES AND SUMMARY OF RESEARCH

The general goal of our ultrasonic investigations is to obtain information about bulk systems near cooperative phase transitions or critical points. In particular, ultrasonic velocities give direct information about the equilibrium thermodynamic properties, and ultrasonic attenuation provides data about the dynamical behavior. The majority of the work is concerned with order-disorder lambda transitions in solids, but fluids near their liquid-vapor critical point are also being studied. During the past year, progress was made on a variety of ultrasonic problems. Work on  $\text{KH}_2\text{PO}_4$  and  $\text{BaTiO}_3$  near their ferroelectric Curie temperatures has been completed. The KDP results have been published, but the  $\text{BaTiO}_3$  data are still being analyzed. Considerable progress has also been made on the study of xenon near its critical point. Data have been obtained along several near-critical isochores and along several isotherms. A recent contribution to Physical Review Letters describes the attenuation results and analyzes them in terms of current theories of dynamical critical phenomena.

In addition to ultrasonic investigations, our group has carried out low-temperature x-ray structural studies, and isothermal compressibility measurements. The latter are based on a new capacitance method of measuring the length of a single crystal as a function of pressure. This method was developed by B. B. Weiner and has been applied to the order-disorder transition  $\text{NH}_4\text{Cl}$ . At low transition pressures ( $p_t < 1500$  bar), there is a small first-order discontinuity in the volume at the "lambda" line. For transition pressures above 1500 bar, the variation  $V$  is continuous and becomes progressively more gradual as the pressure increases.

Work continues on dynamical calorimetry, where phase-sensitive detection techniques are applied to the measurement of heat capacities near a critical point. Apparatus has been designed and tested; measurements on chromium near its antiferromagnetic transition are in progress.

C. W. Garland

### Publications

A. Bonilla, C. W. Garland, and N. E. Schumaker, "Low Temperature X-Ray Investigation of  $\text{NH}_4\text{Br}$ ," *Acta Cryst.* A26, 156 (1970).

N. E. Schumaker and C. W. Garland, "Infrared Investigation of Structural and Ordering Changes in Ammonium Chloride and Ordering Changes in Ammonium Chloride and Bromide," *J. Chem. Phys.* 53, 392 (1970).

C. W. Garland and B. B. Weiner, "Anharmonic Potential Function for Libration in Ammonium Chloride and Bromide," *J. Chem. Phys.* 53, 1609 (1970).

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C. W. Garland and D. D. Snyder, "Ultrasonic Attenuation near the Lambda Transition in  $\text{NH}_4\text{Cl}$  at High Pressures," *J. Phys. Chem. Solids* 31, 1759 (1970).

C. W. Garland, "Ultrasonic Investigation of Phase Transitions and Critical Points," in *Physical Acoustics*, W. P. Mason and R. N. Thurston (eds.) (Academic Press, New York, 1970), Vol. 7, Chap. 2, pp. 51-148.

C. W. Garland, D. Eden, and L. Mistura, "Critical Sound Absorption in Xenon," *Phys. Rev. Letters* 25, 1161 (1970).

E. Litov and C. W. Garland, "Ultrasonic Investigation of the Ferroelectric Transition Region in  $\text{KH}_2\text{PO}_4$ ," *Phys. Rev. B* 2, 4597 (1970).

C. W. Garland and B. Weiner, "Changes in the Thermodynamic Character of the  $\text{NH}_4\text{Cl}$  Order-Disorder Transition at High Pressures" (to appear in *Phys. Rev. B*, March 15, 1971).

Theses

B. B. Weiner, "Volume Anomalies in Ammonium Chloride along the Lambda Line," Ph.D. Thesis, Department of Chemistry, M. I. T., September 1970.