XI. 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 in a variety of areas. Ultrasonic investigations of xenon near its critical point and KH_2PO_4 near its ferroelectric Curie temperature have been actively pursued. Dr. Eliyahu Litov has completed a study of the transition region in KDP as a function of applied electric field. A field of at least 3 kV cm^{-1} was found necessary to obtain a single-domain crystal and avoid scattering losses. The most interesting result of this work involves comparison of the polarization relaxation times in KDP with those in KD^*P (that is, KD_2PO_4). In the paraelectric phase $\tau(KDP) \approx 0.1\ \tau(KD^*P)$, whereas the two relaxation times are essentially equal in the ferroelectric phase. This indicates that hydrogen tunneling is an important mechanism above T_c for KDP. Considerable progress has also been made on the study of xenon. Data have been obtained by Peter E. Mueller and Don Eden along the two near-critical isochores and along several isotherms. Recently, the sensitivity of the ultrasonic equipment was greatly enhanced by modifying a coherent detection scheme for use in variable-path measurements. This permits velocity accuracy of ± 0.7 dB cm^{-1}, even when dealing with pulses that are attenuated as much as 1000 dB cm^{-1}. The most important result of the xenon work is the observation of appreciable velocity dispersion and nonclassical attenuation behavior between 0.5 MHz and 5 MHz. Recently, Dr. Strukov and Dr. Litov have begun an ultrasonic study of polarization fluctuations near the Curie point in $BaTiO_3$, which is a typical displacive ferroelectric.

In addition to ultrasonic investigations, the group has carried out infrared measurements on ammonium halide crystals near order-disorder transitions, low-temperature x-ray structural studies, and isothermal compressibility measurements. The last 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 Bruce B. Weiner and has been applied to the order-disorder transition in NH_4Cl. At low transition pressures ($p_t < 1700$ bar), there is a small first-order discontinuity in the volume at the "lambda" line. For transition pressures between 1700 bar and 2500 bar, the transition appears to be of the classical lambda type (that is, V varies continuously but infinitely rapidly at

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the transition). Above 2500 bar, the volume variation indicates that the transition is second-order in the Ehrenfest sense.

The newest work in our group involves dynamical calorimetry, where phase-sensitive detection techniques have been applied to the measurement of heat capacities near a critical point. The sample is subject to a heat input at a fixed frequency ω, and its temperature variation is detected with a thermocouple whose output is measured with a "lock-in" detector. This output signal, which is inversely proportional to the heat capacity of the sample, can be measured with considerable precision as a function of the mean sample temperature. The dynamic behavior of a system with oscillatory heat input is determined by two time constants. It is desirable that $\omega\tau_1 > 1$ and $\omega\tau_2 \ll 1$, where τ_1 is the relaxation time for heat flow between the sample and the surroundings, and τ_2 is the characteristic time for the sample to reach internal thermal equilibrium. Apparatus has been designed and test measurements are in progress on the application of this method to chromium near its antiferromagnetic transition.

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Publications

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Theses

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