tant? (2) Does our model give a satisfactory explanation of the attenuation of microwave phonons[12]? These problems will be discussed in a forthcoming paper.

We thank Dr. T. Ishiguro for helpful discussions.

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[7] In the case where the acceptor ground state is fourfold degenerate, the attenuation due to the relaxation process is given by $\alpha = (ND_{u'}{}^{2}a^{2}/3\rho v_t{}^3)[\omega^2\tau/(1+\omega^2\tau^2)]$. In this case the relaxation time τ is determined by the phonon-assisted hopping process, the Raman process, and the interaction with excited states, which give too long a relaxation time τ at low temperatures.

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Rayleigh Scattering Studies of NH_4Cl at High Pressures*

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The integrated Rayleigh intensity of NH_4Cl in the region of the order-disorder transition line has been studied as a function of pressure and temperature. Increased scattering is observed near the tricritical point recently detected by Garland. Although the presence of domain scattering makes unambiguous interpretation of the data difficult, these data are consistent with the Landau-Ginzburg theory of critical opalescence.

Ammonium chloride undergoes an order-disorder phase transition involving an orientational ordering of $NH_4{}^+$ tetrahedra within a cubic lattice, and much experimental work has been done on this system in an effort to elucidate the detailed nature of the transition. Recent experiments[1,2] have indicated that certain characteristics of the transition are different at high pressures than they are at atmospheric pressure. For example, Weiner and Garland[2] have measured changes in the length, l, of single crystals of NH_4Cl as a function of pressure along a number of isotherms and found that l exhibits a discontinuous jump at the transition line for temperatures below about 256°K, but is a continuous function of pressure for higher temperatures. (For these temperatures the transition occurs at pressures greater than ~1500 bars.) Thus, their experiment indicates the existence of a tricritical point, where the line of first-order transitions (in the p-T plane) changes into a line of second-order transitions.

The possible existence of tricritical points (or Curie critical points) is included in Landau's phenomenological theory of phase transitions.[3] Ginzburg[4] has used this theory to predict the possible existence of critical opalescence associated with Curie critical points, but this effects has never been definitely observed in any crystal. In order to test the Landau-Ginzburg theory, we have studied the Rayleigh scattering intensity of NH_4Cl as a function of pressure for temperatures between 252 and 264°K.

The sample used in this experiment was a single crystal, approximately $3\times5\times7$ mm^3 in size, which was grown by slow evaporation from an aqueous solution containing about 20% by weight of urea as a habit modifier. The crystal was carefully polished with Linde A and methanol on Pellon paper and mounted in a black Bakelite holder, which served both to mask stray light from the sample and to hold the sample in the high-pressure, three-window optical cell. The cell itself was mounted inside a thermally insulated brass can, which was cooled by a Forma constant temperature bath (0.01°K) and circulator. Three windows through the can and insulation allowed a 90° light-scattering geometry to be utilized. A thermocouple and potentiometer were made to measure the temperature at the outside of the pressure cell to precision of ±0.05°K and an accuracy of ±0.2°K. Pressure was generated with a hand pump and was measured to a precision of ±10 psi and an accuracy of 1% with a Harwood Manganin gauge and a homemade Carey-Foster bridge circuit. Octoil *S* was used for the pressurizing fluid.

Light from a Spectra Physics 125 HeNe laser (6328 Å, 30 mW) was focused into the sample, and the light scattered at 90° was collected, filtered to eliminate Raman lines, and passed through a Fabry-Perot interferometer which was tuned to the Rayleigh line. The wave vectors of the incident and scattered light were directed along cubic axes of the crystal. As no width was ever observed to the Rayleigh line (see below), it is assumed that the signal from the interferometer was proportional to the integrated Rayleigh intensity. The intensity of the beam transmitted through the crystal was recorded along with the Rayleigh intensity. This was necessary because the incident beam is so strongly attenuated near the transition (as much as 70% for the lowest temperature used), that the measured scattered intensity must be corrected in order to reflect the actual cross section.

The total Rayleigh intensity was measured as a function of pressure along seventeen different isotherms, six of which are indicated by the vertical lines crossing the transition line on the phase diagram of Fig. 1. The pressures used typically ranged from 5000 psi above the transition to 5000 psi below. The most serious experimental difficulty encountered was large day-to-day variations in the scattered intensity, due mainly to the beam and crystal shifting relative to each other. Fortunately, the shape of the curve of intensity versus pressure for a given temperature was reproducible from day to day; thus the appropriate correction factor was found to be multiplicative. The data were multiplicatively normalized to have unit intensity at a pressure 5000 psi below the transition line, which is well into the disordered phase, where normal behavior is expected. Separate measurements have indicated that this quantity, $I(p_0(T)-5000)$, may actually decrease slightly with increasing temperature.

After the scans of intensity versus pressure were completed, an attempt was made to mea-

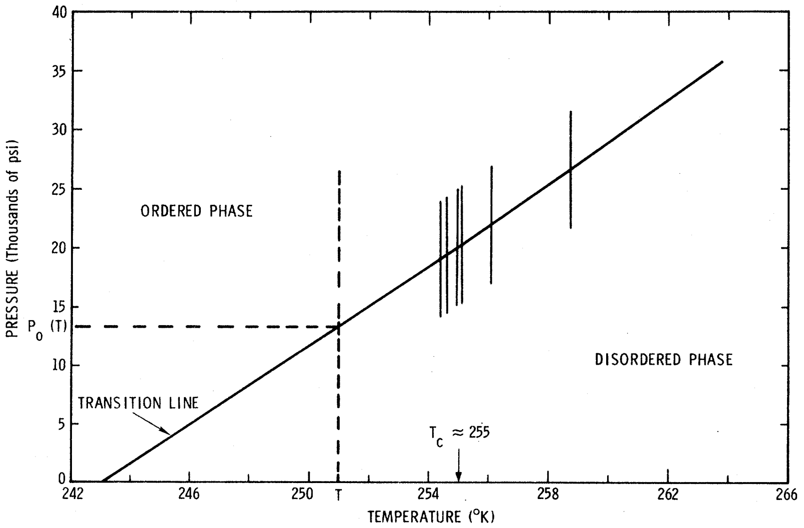

FIG. 1. Partial phase diagram of NH$_4$Cl showing the order-disorder transition line and six isotherms used in the experiment.

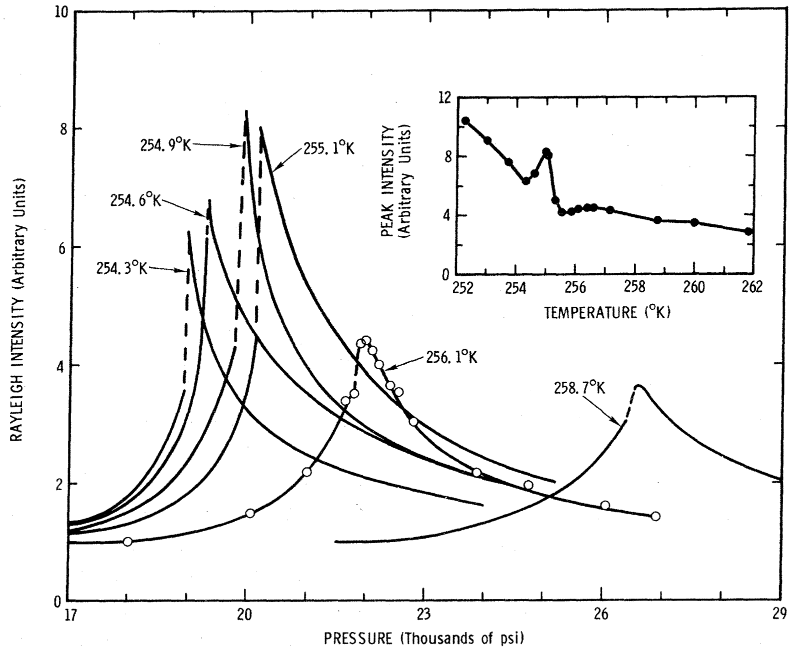

FIG. 2. Rayleigh intensity (corrected for attenuation) versus pressure for the six isotherms indicated in Fig. 1. Insert: peak intensity versus temperature for each of seventeen isotherms.

sure the width of the Rayleigh line. For this experiment a longer spacer was used in the Fabry-Perot (1.3 vs 0.3 cm) which, with a finesse of 20, gave a resolution of 0.02 cm^{-1} (600 MHz). No changes in the width of the Rayleigh line were observable as a function of pressure and temperature, indicating that the scattering either has a very narrow spectrum or is completely elastic.

The intensity versus pressure data were normalized and corrected for incident beam attenuation as discussed above. The resulting intensity-versus-pressure curves for the six isotherms indicated on Fig. 1 are shown in Fig. 2. At the highest temperature (258.7°K) the scattering is relatively weak, but as the transition line is crossed at successively lower temperatures the peak scattering becomes progressively stronger down to a temperature of about 255°K. Below this temperature the scattering becomes weaker. For temperatures below 254°K the scattering again becomes progressively stronger with decreasing temperature. This behavior is shown in the insert of Fig. 1, where the maximum intensities obtained on all seventeen isotherms studied are plotted as a function of temperature.

The peak in the plot of maximum intensity versus temperature occurs at around 255°K, which is within a degree of the temperature where Garland found the transition to change from first order to second order, and we believe that this peak may be associated with the proposed tricritical point. The large increase in the scattering at the lowest temperatures is probably due to domain scattering. At these temperatures the transition becomes noticeably discontinuous, and the domain walls should therefore be highly strained, leading to an increase in elastic scattering.

Unfortunately, the mechanism of the Rayleigh scattering was not determined by this experiment. There are three immediately obvious possible mechanisms, any combination of which might contribute to the scattering: (1) The scattering could be from the fluctuating order parameter η. Because of the high symmetry, the coupling to the dielectric tensor would be quadratic, $\Delta\epsilon \propto \Delta(\eta^2)$, and the Landau-Ginzburg (LG) theory would be applicable. (2) Since longitudinal strains are coupled to η, there could be scattering from critical strain fluctuations.[5] (3) The scattering could be from domains, and critical effects could be observed indirectly through their effect on the domain structure.

With the explicit understanding that these uncertainties (plus the somewhat uncertain normalization and attenuation correction procedures) make any quantitative interpretation of the data highly tentative, we wish to present evidence that out data are consistent with the LG theory of critical opalescence.

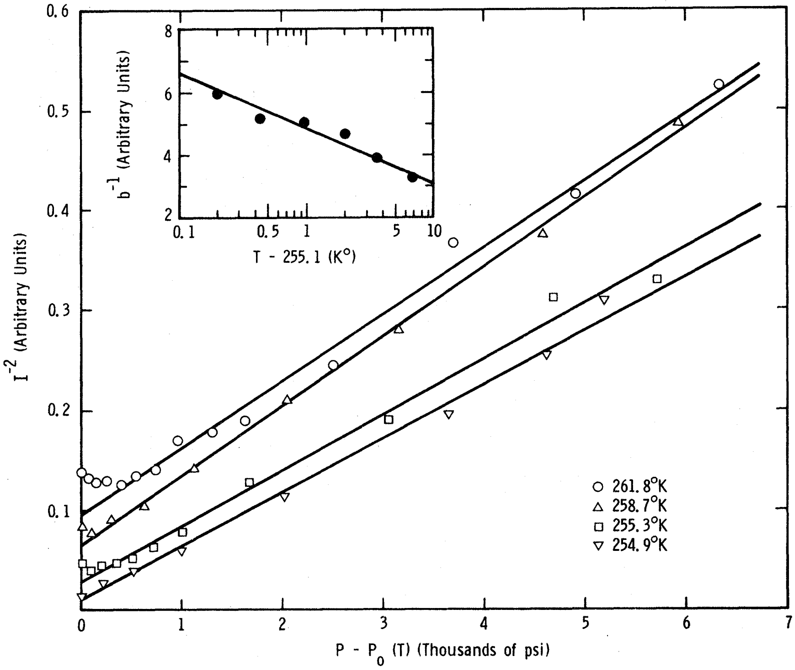

FIG. 3. Linear fits to I^{-2} vs $p-p_0(T)$. Insert: approximately logarithmic divergence of b for $T>255°K$.

LG[4] expand the free energy of the crystal as $A(\eta,p,T)=A_0+a\eta^2+\frac{1}{2}b\eta^4+\frac{1}{6}c\eta^6$, where A_0 is constant and a, b, and c are functions of pressure and temperature. The coefficient a is positive in the disordered phase and negative (except near a first-order transition) in the ordered phase. The p-T dependence of b is unknown, and c is usually taken to be a positive constant. If it is assumed that η couples quadratically to the dielectric tensor, then critical opalescence is expected in the ordered phase with intensity given by[4]

$$I\propto(b^2-2ac)^{-1/2}.$$

If b is positive at the transition, then the transition (which is continuous) occurs at $a=0$, and the maximum intensity is b^{-1}. If b is negative at the transition, then the transition (which is discontinuous) occurs at $a=3b^2/8c$, and the maximum intensity is $2|b|^{-1}$. At the Curie critical point, b is zero and the intensity diverges (critical opalescence).

To describe experiments done along isotherms, we can write a as[6] $a=-a_0[p-p_0(T)]$, where $p_0(T)$ is the transition pressure at temperature T (see Fig. 1), and a_0 is positive. If we make the crude assumption that b is nearly constant along isotherms, then we expect the data plotted as I^{-2} vs $p-p_0(T)$ to fall on straight lines having intercepts of b^2. In Fig. 3 such plots are shown for four different temperatures, along with linear least-squares fits (solid lines). All data are from the ordered phase. Except for the highest temperatures, where the curves turn up near the transition, the data are reasonably well represented by straight lines, as predicted, with b smallest for temperatures near 255°K. The actual divergence of b^{-1} along the transition line in the second-order region is found to be weak. The data are reasonably well represented by either $b^{-1}\propto(T-255.1)^r$ with $r\approx0.2$, or by $b^{-1}\propto\ln(T-255.1)$, for T in °K. The insert to Fig. 3 shows the approximately linear behavior of b^{-1} vs $\ln(T-255.1)$.

Our measurements of the pressure-temperature dependence of the total Rayleigh intensity support the conclusion that the nature of the NH_4Cl phase transition changes at a temperature of about 255°K and a pressure of about 20 000 psi. Although we have not determined how much of the scattering is due to fluctuation of the order parameter, the data are consistent with the LG theory of critical opalescence. Further experimental work is needed to determine the mechanism of the scattering, preferably spectroscopy of high-enough resolution to separate the spectrum of any critical scattering from domain scattering. High-magnification photography to study domain effects, and studies of the angular dependence of the scattering would also be useful.

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Spin-Wave Resonance Fields, Linewidths, and Intensities of a Permalloy Film: Theory and Experiment

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Using only the surface-anisotropy constants and the saturation magnetization as adjustable parameters, we have found good agreement between an observed and a calculated spin-wave resonance spectrum. A uniform magnetization, effects of exchange, conductivity, and Landau-Lifshitz damping are included in the theory.

The purpose of this paper is to show that the observed spin-wave resonance spectrum of a thin metal film can be fitted quite well by an extension of a theory[1] which takes into account four factors: exchange, electrical conductivity, an asymmetric anisotropy at the two surfaces of the film, and an isotropic Landau-Lifshitz damping constant λ. We present, for the first time, *quantitative* theoretical and experimental comparisons of all three important parameters observed in a standing spin-wave resonance spectrum: resonance field positions, line intensities, and linewidths. The results of this study, along with others,[2] suggest that, if care is taken in preparing the film, many of the contributions to the resonance spectrum as a result of inhomogeneities in the sample may be eliminated. Previous papers on this subject were concerned with studying the intensities of the resonance lines either alone or in combination with the positions.[3-6]. No reports to date have been given of a consideration of all of the resonance parameters (including linewidths) simultaneously.

Most of the details of the film deposition and microwave techniques have been described previously.[7] In preparing the film, substrates were polished optically smooth and thoroughly cleaned, and the deposition was carried out only after the melt had come to thermal equilibrium with its vapor. A rather fast evaporation rate of 38 Å/sec was used assuring that the grains of the film were randomly spaced and not columnar across the film thickness.[8] During evaporation, the pressure ranged from 3×10^{-8} Torr to a maximum of 8×10^{-7} Torr. The oven, which maintained a substrate temperature of 250°C during deposition, was turned off 5 min after the deposition was completed.

It was necessary to extend previous magnetic-resonance calculations[1] in order to account for realistic boundary conditions applicable to evaporated Permalloy films. The theory permits an asymmetrical rf excitation of the film for the following reason. Although the film is placed at the center of a rectangular cavity, any error in placement from the cavity center would result in an asymmetric rf excitation. In addition, the calculation permits asymmetrical pinning conditions, between the extreme limits of complete pinning to no pinning at all, at the two surfaces of the film. There may be several reasons why asymmetrical pinning occurs in actual films. One side of the film is rigidly attached to a substrate while the other side is free. In addition, the first few layers of the film structure, deposited at the beginning of the evaporation, may be quite different from the last few layers, deposited at the end of the evaporation. In both