

tetragonal distortion of the lattice and the displacements of the bromide ions which occur in the  $\gamma$  phase on ordering. Nor is the Ising model theory<sup>25</sup> applied to the  $\text{NH}_4\text{Cl}$  data valid in the case of the  $\beta$ - $\gamma$  transition in  $\text{NH}_4\text{Br}$ .

The behavior of  $c_{11}$  just above the lambda point in ammonium bromide is very similar to that observed in ammonium chloride, whereas the behavior of  $C'$  is different in the two cases. Unlike the data for the chloride,  $C'$  values for the bromide show a marked anomalous decrease which is apparent as much as  $15^\circ\text{K}$  above the lambda point (see Fig. 4). Attenuation of the ultrasonic waves associated with both  $c_{11}$  and  $C'$

<sup>25</sup> R. Renard and C. W. Garland, *J. Chem. Phys.* **44**, 1125 (1966).

was very high over a considerable range of temperatures below the lambda point. This is presumably due to the presence of domains consisting of tetragonal crystallites with their unique axes lying at random along one of the three original cubic axes. The presence of domains is common in antiferromagnetic crystals and  $\gamma$ -phase ammonium bromide is analogous to an antiferromagnet.

A more extended discussion of the properties of the ordered phase and of the lambda transition region is difficult and inappropriate at this time. New experimental work is now in progress on ammonium bromide in the region  $100^\circ$  to  $250^\circ\text{K}$  and 0 to 6 kbar. This will provide information on both the  $\gamma$  and  $\delta$  phases, as well as new data in the regions of the various transition lines.

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## Order-Disorder Phenomena. I. Instability and Hysteresis in an Ising Model Near Its Critical Point\*

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The mechanical behavior of a system near a cooperative order-disorder transition point is discussed in terms of an Ising model for a set of spins located on mass particles which form a compressible lattice. With the assumption of weak coupling between the lattice and spin systems, it is shown that this Ising model is unstable in the immediate vicinity of its critical point and undergoes a first-order transition. In addition, many properties should show hysteresis in the critical region. These general conclusions are illustrated by several two-dimensional examples.

### INTRODUCTION

IT is a well known but striking fact that substances which undergo cooperative order-disorder transitions usually exhibit anomalous variations in volume which extend over the same temperature range as the lambda spikes in the specific heat. A large amount of theoretical work has been carried out on the thermal properties of such cooperative systems, but little attention has been paid to the mechanical aspects of the problem. Indeed, in most statistical theories the volume of the system is held fixed and it is assumed that experiments could be carried out directly at constant volume. Since it is the pressure rather than the volume which is usually subject to experimental control, the mechanical behavior of a system near a lambda transition point may be of considerable importance.

Our treatment is based on an Ising model for a system of spins located on mass particles which form

a compressible lattice. Due to its simplicity, the Ising model is fairly tractable and a great deal has already been done for the fixed-volume case, including an exact solution of the two-dimensional problem by Onsager.<sup>1</sup> In this paper (I), the model is defined and the character of the transition very close to the critical point is investigated. General conclusions about the instability of the system (and a resulting hysteresis) are illustrated by several explicit, two-dimensional examples. In the following paper (II), the model is generalized in terms of stress-strain variables for the two-dimensional case, and the contributions to the elastic constants due to spin ordering is derived in analytic form. Both of these theoretical developments were inspired by recent ultrasonic measurements on ammonium chloride near its lambda transition; and that system, which is analogous to a simple-cubic ferromagnet, provides several excellent confirmations of our predictions. The experimental results on  $\text{NH}_4\text{Cl}$  and their interpretation are given in Paper III.

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<sup>1</sup> L. Onsager, *Phys. Rev.* **65**, 117 (1944).