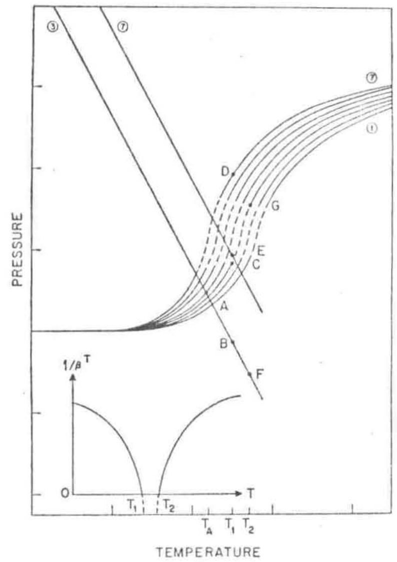

FIG. 3. Behavior of a two-dimensional Ising model at constant volume. The family of *curves* p_I were calculated at seven evenly spaced areas from σ_1 to σ_7. The *lines* $-p_{dl}$ were drawn to represent a disordered lattice with typical compressibility and thermal expansion coefficients. The encircled numbers indicate the spin and lattice isochores at the given areas. The inset represents schematically the temperature dependence of the reciprocal isothermal compressibility $1/\beta^T$.

forbidden zones are shown as dashed lines. The negative disordered-lattice isochores $-p_{dl}(T)$ are also plotted for areas σ_3 and σ_7. Let us assume we want to keep the system at a constant area σ_3. Under zero external pressure, the equilibrium point is at A, corresponding to a temperature T_A. As the temperature is increased, the system can be kept at constant area σ_3 by applying an external pressure. When the temperature reaches T_1 where the appropriate external pressure is of magnitude $p_1=BC$, the system becomes mechanically unstable. Then the area will spontaneously *increase* to (say) σ_7 which is a stable state at temperature T_1 under an external pressure $p_1=BC=DE$. In the range $T_1<T<T_2$ it is impossible, by any manipulation of the external pressure,[8] to keep the area at value σ_3. Above T_2 it is again possible to maintain the area σ_3. If the metastable equilibrium is disrupted before the mechanical instability point is reached, the range of temperature over which it is impossible to maintain constant area is widened somewhat.

An inset on Fig. 3 shows the schematic variation of $1/\beta^T$ as a function of temperature for a constant area σ_3. At T_1 and T_2, $1/\beta^T$ vanishes; the dashed lines represent the behavior expected if the area could be kept constant (i.e., if one could work in an unstable region). Because of the instability, the area of the crystal between T_1 and T_2 depends on the way the experimental run is conducted. The actual area can correspond to an equilibrium point close to or far from an instability point. As a result, experimental values for $1/\beta^T$ between T_1 and T_2 can vary between 0 and an upper value corresponding to the completely disordered state. Consequently, compressibility measurements at constant area σ_3 are meaningful only outside the temperature interval $T_1<T<T_2$.

CONCLUSION

The preceding illustrations of instability and hysteresis near a critical point have been given in terms of a two-dimensional model. The generalization of the discussion to a three-dimensional Ising model is quite easy. For a real three-dimensional crystal, $1/\beta_{dl}^T$ is experimentally known to be finite at temperatures above T_c, and according to our model it is therefore finite at all temperatures. Recent approximate calculations[9,10] indicate that C_I for a cubic Ising model does approach infinity as T approaches T_c. If so, there will be a range of temperatures in the critical region for which the inequality (10) cannot be satisfied. If C_I does not in fact become infinite at T_c the system may display a lambda transition. However, a very large finite value for C_I can still cause a soft crystal (for which $1/\beta_{dl}^T$ is small) to become unstable. If the crystal does become unstable before the critical point is reached, there is also a region of metastability and the strong probability of hysteresis. The general nature of the hysteresis is the same as that shown in Figs. 1–3 since the isotherms and isochores for p_I and p_{dl} have qualitatively the same shape in three dimensions as in two (although the p_I curve is less symmetrical in three dimensions).

In summary, a first-order transition is to be expected in crystals near a lambda point unless some special kind of strong lattice–spin coupling is invoked. The observable effects of this instability should be large only when (a) the lattice is quite compressible (β_{dl}^T large) and (b) the spin interactions are a sensitive function of distance (dJ/dv large). Thus, this phenomenon is difficult to observe in many ferromagnetic solids. In Paper III we hope to show for ammonium chloride, which satisfies both Conditions (a) and (b), that the experimental data conform very well to the predictions of this model.

[8] There is, in principle, a way to keep the volume of a three-dimensional crystal constant. It consists of clamping the crystal in an infinitely rigid holder. This is equivalent to making the disordered lattice incompressible. In this case inequality (10) is fulfilled at any temperature. In practice this can not easily be realized; one usually places the sample in a fluid under pressure which is externally set at some given value.

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