The possibility of an instability for a compressible lattice near an order–disorder lambda point was first pointed out by Rice,[2] who presented a very general thermodynamic discussion of the problem. A few years later, Domb[3] gave a brief demonstration of the instability of a compressible Ising model, but then the problem appears to have languished for some time. More recently, Bean and Rodbell[4] have demonstrated that magnetic ordering can lead to a first-order transition in the critical region. They based their discussion on the molecular-field theory of ferromagnetism and presented data on MnAs as experimental evidence for their conclusions. Mattis and Schultz[5] have arrived at essentially identical conclusions in their theory of magnetothermomechanics. It is shown here on the basis of a very simple model that an Ising lattice is unstable in the immediate vicinity of its critical point and undergoes a first-order transition with hysteresis. This is proved for a two-dimensional square lattice of ferromagnetic particles in the absence of an external field. Very general conditions are given for observing this effect in a three-dimensional case.

FORMULATION OF THE MODEL

Let us first consider the usual Ising lattice[6] consisting of an array of N fixed sites. Associated with each site are two possible spin orientations—"spin up" and "spin down". For zero external magnetic field, it is customary to write the energy of a given spin configuration as

$$E=-J(q_p-q_a)+\gamma NK/2, \qquad (1)$$

where q_p is the number of nearest-neighbor pairs in the lattice with parallel spins, q_a is the number with antiparallel spins, $\gamma N/2$ is the total number of nearest neighbors ($\gamma=4$ for two-dimensional square lattice, $\gamma=6$ for three-dimensional sc lattice, etc.), and J and K are defined by

$$J=(U_a-U_p)/2; \qquad K=(U_a+U_p)/2. \qquad (2)$$

In the above, U_p is the potential energy due to spin interaction between a nearest-neighbor pair of parallel spins and U_a is the interaction energy for a pair of antiparallel spins. The case $J>0$ corresponds to ferromagnetism. For simplicity, the model is given only for the case of isotropic spin interactions. Thus, the spin partition function is $Q_s=Q_I(0, H)\exp(-\gamma NK/2kT)$, where $Q_I(0, H)$ is the well-known Ising partition function at zero field as a function of $H\equiv J/kT$.

Instead of being concerned with the thermal behavior of a "clamped" system of spins only, we wish to consider the mechanical behavior of a more realistic model in which the localized spins are associated with mass particles (atoms, ions or molecules) which form a compressible lattice. Indeed, an "unclamped" array of spins only is usually unstable and it is the interaction between mass particles which stabilizes the composite system. We *assume* weak coupling between the lattice and spin systems; i.e., we assume that $Q=Q_sQ_l$, where Q_l is the partition function of the particle lattice. This is the crucial feature of our model. Almost all theories of order–disorder phenomena are based on the implicit hypothesis that a configurational partition function can be written without taking into account strong coupling between the spins and the rest of the system.[7] It is possible to check on this weak-coupling assumption for regions far away from the critical point since many properties (e.g., heat capacity, thermal expansion, elastic constants) then depend essentially on the lattice contribution. If the coupling is weak, these properties should have comparable temperature and pressure dependences in the completely ordered and completely disordered state, assuming that these two states belong to the same crystallographic group. It is, however, possible to imagine that the coupling will be strong *only* in the critical region. Since there is no theoretical evidence that this must be true in general, we have made the simpler assumption and present below the consequences of weak coupling in an Ising model.

It proves convenient to rewrite the over-all partition function of the system in a new form

$$Q=Q_sQ_l=Q_I\exp(-\gamma NK/2kT)Q_l=Q_IQ_{dl}, \qquad (3)$$

where Q_{dl}, the partition function for the *disordered lattice*, includes both the usual lattice contribution of a normal crystal and the interactions between randomly-oriented spins. As a result of Eq. (3) all the thermodynamic functions can be written as a sum of two independent contributions; in particular for the Helmholtz free energy, $A=A_I+A_{dl}=-kT\ln Q_I-kT\ln Q_{dl}$. The contribution to the properties of the system which arise from the Q_{dl} term can be deduced empirically from experiments performed on crystals considerably above their lambda points. We emphasize here the contribution to various properties due to the Q_I term which describes the spin ordering. The expressions for the configurational internal energy and specific heat at constant volume are well known[6] and can be written as

$$U_I=-Jd\ln Q_I/dH, \qquad (4)$$

$$C_I=kH^2d^2\ln Q_I/dH^2, \qquad (5)$$

since Q_I is a function only of $H\equiv J/kT$ and J is not a function of temperature. The quantity J will, however, be a function of the spacing between lattice sites.

[2] O. K. Rice, J. Chem. Phys. **22,** 1535 (1954).
[3] C. Domb, J. Chem. Phys. **25,** 783 (1956).
[4] C. P. Bean and R. S. Rodbell, Phys. Rev. **126,** 104 (1962).
[5] D. C. Mattis and T. D. Schultz, Phys. Rev. **129,** 175 (1963).
[6] K. Huang, *Statistical Mechanics* (John Wiley & Sons, Inc. New York, 1963), Chaps. 16 and 17; H. S. Greene and C. A. Hurst, *Order–Disorder Phenomena* (Interscience Publishers, Inc., New York, 1964), Chaps. 2, 3, and 6.

[7] Some work has been done on the coupling between ordering and vibrational motions for a one-dimensional binary alloy; A. A. Maradudin, E. W. Montroll, and G. H. Weiss, Solid State Phys. Suppl. 3., pp. 188–212 (1963).