PHASE TRANSITIONS OF AN ISOTROPIC FERROMAGNET IN AN EXTERNAL MAGNETIC FIELD

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The thermodynamics of transitions between uniformly and nonuniformly magnetized states of an isotropic ferromagnet in an external field is described in the molecular-field approximation. These results are found to be in substantial agreement with many experiments.

Griffiths[1] has recently proved a theorem about model ferromagnets consisting of localized spins on a lattice with dipole-dipole interactions included. He has shown that the equilibrium bulk free energy in the absence of a magnetic field is independent of shape in the thermodynamic limit of a very large system. Arrott[2] has pointed out that an immediate consequence of this theorem is that, in the absence of a magnetic field, the lowest energy state of an isotropic magnetic system is not a state of uniform magnetization (the formation of domains occurs only in the presence of anisotropy). Arrott further suggested the existence of a possible connection between this conclusion and the recent observations of van der Hoeven, Teaney, and Moruzzi[3] on the heat capacity of EuS in external magnetic fields.

In this Letter we examine the consequence of Arrott's suggestions. Assuming that the low-energy states of an isotropic ferromagnet are indeed states of nonuniform magnetization, we find that such a system can have phase transitions in finite external magnetic fields for certain geometries. These transitions are physically distinct from the usual zero-field Curie point. The zero-field Curie point is that temperature at which the system changes from a state of vanishing long-range order to a state of finite long-range order. In the finite-field transitions described here, the system changes from a state of one kind of long-range order into a state of a different kind of long-range order (from a nonuniformly magnetized state into a uniformly magnetized state). The thermodynamics of such transitions is examined here from the molecular-field point of view for a simple geometrical arrangement. The results of this calculation appear to verified by a number of experimental observations in various geometries.

Consider a thin toroid of cubic, isotropic, ferromagnetic material.[2] Because of the dipole-dipole interactions (or equivalently the demagnetizing field) the nonuniformly magnetized state (N) is that in which the magnetization is directed around the toroid. The uniformly magnetized state (U) is that in which the magnetization is in the direction of a field applied perpendicular to the toroid. The problem of finding the conditions under which N transforms into U is similar to that of calculating the perpendicular susceptibility of an antiferromagnet.[4]

For an applied field $\vec{H}=H_0\hat{z}$ ($\hat{z}$ is perpendicular to the plane of the toroid), we decompose the magnetization of a given volume element of the toroid: $\vec{M}=M\hat{m}=M_z\hat{z}+M_\theta\hat{\theta}$, where $\hat{\theta}$ is the local circumferential direction. Then, in the effective-field approximation, the total field acting on a given localized spin is $\vec{H}=H_0\hat{z}-DM_z\hat{z}+\gamma M\hat{m}$, the resultant of the applied field, the demagnetizing field, and the exchange field. $D\approx 2\pi$ for this geometry. Application of the requirement that the torques on all moments must vanish at equilibrium yields the result $M_\theta(H_0-DM_z)=0$, the solutions of which are the equilibrium states N and U. For state U, $M_\theta=0$ and $M_z=M$, and a measurement of the magnetization along H_0 would yield the usual total temperature-dependent moment. For state N, however, $M_\theta\neq 0$ and $M_z=H_0/D$, and a measurement of the magnetization along H_0 would yield a constant, analogous to the perpendicular susceptibility of an antiferromagnet. We note that while the condition $M_z=M$ can always be satisfied, the condition $M_z=H_0/D$ can only be satisfied when the total magnetization M exceeds H_0/D.

The ranges of stability of N and U are found by computing the difference in free energy between the two states as a function of temperature and applied field. For simplicity, we use the molecular-field approximation with spin $\frac{1}{2}$. In state U, the effective field is $\vec{H}=(H_0-DM+\gamma M)\hat{z}$. In state N, the external and demagnetizing fields in the z direction cancel and $\vec{H}=\gamma M\hat{m}$. The respective free energies of the two states are[5] (in units of γM_0^2)

$$F_U=F_0(\tau)-\tau\ln\{2\cosh[(h-d\sigma_U+\sigma_U)/\tau]\}-\tfrac{1}{2}d\sigma_U^2+\tfrac{1}{2}\sigma_U^2,$$

$$F_N=F_0(\tau)-\tau\ln[2\cosh(\sigma_N/\tau)]-\tfrac{1}{2}h^2/d+\tfrac{1}{2}\sigma_N^2, \quad (1)$$

where $h = H_0/\gamma M_0$, $d = D/\gamma$, M_0 is the magnetization at 0°K, $\tau = T/T_c(0)$, and $T_c(0)$ is the Curie point. The long-range order parameters are $\sigma_N = M_N/M_0$ and $\sigma_U = M_U/M_0$. Minimization of F_U and F_N with respect to σ_U and σ_N, respectively, at constant reduced temperature and applied field H_0 yields the equilibrium values of the order parameters,[5]

$$\sigma_U = \tanh[(h - d\sigma_U + \sigma_U)/\tau],$$

$$\sigma_N = \tanh(\sigma_N/\tau). \tag{2}$$

Transitions from N to U can occur when $F_N = F_U$. This condition is satisfied when $\sigma_N = \sigma_U = h/d$ and, from Eq. (2), the transition temperature is

$$\tau_t = (h/d)[\operatorname{inv}\tanh(h/d)]^{-1}. \tag{3}$$

For very small fields, Eq. (3) reduces to

$$\tau_t = 1 - \tfrac{1}{3}(h/d)^2.$$

For $\tau < \tau_t$, $F_N < F_U$ and N is the stable state; for $\tau > \tau_t$, σ_N is less than h/d and hence U is the only possible stable state. Note that $\sigma_N = \sigma_U$ at τ_t. Thus, the magnetization is continuous through the transition as is its measurable component M_z. Above τ_t, $M_z = M_0\sigma_U$ and below τ_t, $M_z = H_0/D$.

Numerical solutions of Eq. (2) for magnetization curves in various applied fields are shown in Fig. 1. The most prominent feature of these results is the joining of the constant magnetization (proportional to H_0) below τ_t with the decreasing magnetization above τ_t at a "kink" in the magnetization curve in low fields. This kink and/or the associated field dependence of the magnetization appear to be rather general phenomena and have been observed by numerous investigators. The earliest references to the linear field dependence include the work of Néel[6] and Fallot.[7] Striking examples of kinks in the low-field magnetization curves have been observed by Arajs and Colvin[8] in spheres of Fe, Co, and Ni. Arrott and Noakes[9] have also described such behavior in their investigation of Fe and its alloys. More recently, Miyatani[10] has observed very sharp kinks in single-crystal $CdCr_2Se_4$ and yttrium iron garnet and in polycrystalline $CdCr_2S_4$ and $CdCr_2Se_4$. Note that for $h/d \geq 1$ or $H_0 \geq DM_0$ the uniformly magnetized state remains the equilibrium state down to 0°K. Differentiation of the free energies with respect to τ at constant H_0 yields the entropies[5] (in units of R, the gas constant):

$$S_U = S_0 + \ln\{2\cosh[h - d\sigma_U + \sigma_U]/\tau\} - (h\sigma_U - d\sigma_U^2 + \sigma_U^2)/\tau,$$

$$S_N = S_0 + \ln[2\cosh(\sigma_N/\tau)] - \sigma_N^2/\tau. \tag{4}$$

Since $\sigma_U = \sigma_N = h/d$ at τ_t, the entropy is continuous through the transition. Differentiating Eqs. (4) with respect to τ at constant H_0 yields the heat capacities[5] (in units of R):

$$C_U = C_0 + \frac{(1-\sigma_U^2)(h - d\sigma_U + \sigma_U)^2}{\tau^2 - \tau(1-\sigma_U^2)(1-d)},$$

$$C_N = C_0 + \frac{(1-\sigma_N^2)\sigma_N^2}{\tau^2 - \tau(1-\sigma_N^2)}. \tag{5}$$

The heat-capacity curves for various different applied fields as computed from Eqs. (5) are shown in Fig. 2. As in the usual molecular-field theory of the zero-field case, a discontinuity in the heat capacity is found at τ_t. As the small applied field is increased, τ_t decreases as discussed above, the discontinuity decreases in size, and the transition broadens above τ_t. At larger values of h/d, we begin to observe the appear-

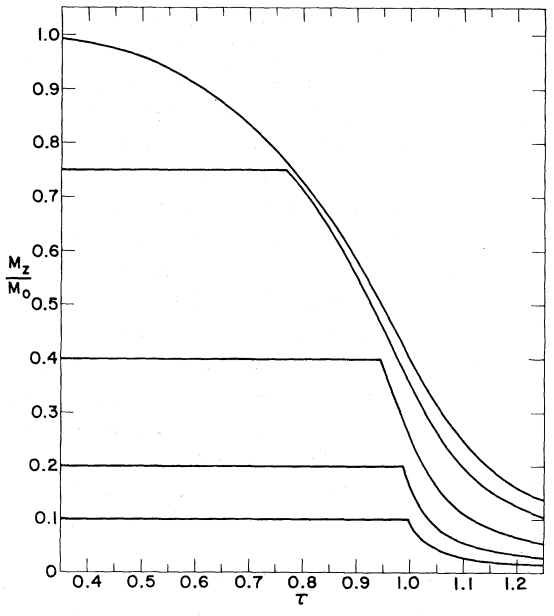

FIG. 1. Magnetization curves for various fields. The respective values of h/d are given by the values of M_z/M_0 at the kinks. In all cases, $d = 0.04$. The top curve corresponds to $h/d = 1$.

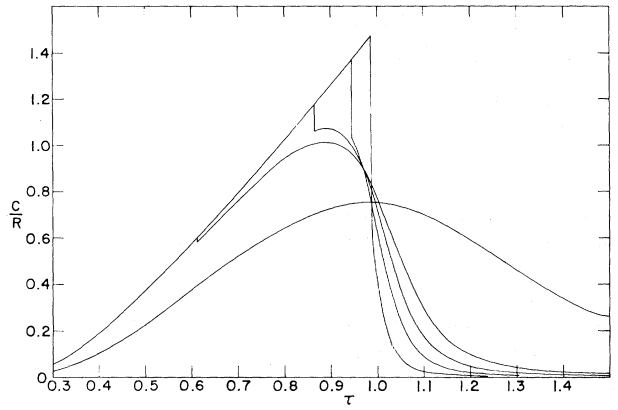

FIG. 2. Heat capacity versus reduced temperature for various fields. In order of decreasing maximum C, the curves correspond to $h/d=0.2$, 0.4, 0.6, 0.9, and 5.0. For $h/d=0.9$, the discontinuity (at $\tau=0.61$) has nearly vanished. In all cases, $d=0.04$.

ance of a broad anomaly above τ_t associated with the gradual development of appreciable long-range order in the uniform state (see Fig. 1). For values of h/d near unity, this broad anomaly is the dominant part of the heat capacity, but the discontinuity remains at (lower) τ_t. The temperature at which the broad anomaly (corresponding to the development of ordering in the U state) has its maximum increases with increasing field.

The features displayed in Fig. 2 are, of course, at best qualitatively correct. We know that the discontinuity is merely an artifact of the molecular-field approximation and that the correct critical behavior must be sought in theories which take account of the fluctuations in the effective field. Nonetheless, we expect experimental results on real materials to exhibit qualitatively the behavior described here. A sharp heat-capacity anomaly associated with the transition from the N to the U state is expected to occur at a τ_t which decreases roughly quadratically for small increasing fields and this anomaly is expected to broaden as the field increases, as observed in Ref. 3. We believe that the T_M defined in Ref. 3 is the τ_t of this work. It is further predicted that these sharp anomalies will be superimposed on the much broader anomalies associated with the development of appreciable long-range order in the uniform state. We have not been able to determine whether such anomalies have been observed in the EuS work,[3] but some earlier experiments of Miedema and his collaborators[11] on $CuK_2Cl_4 \cdot 2H_2O$ and $Cu(NH_3)_4SO_4 \cdot H_2O$ exhibit the qualitative features of the curves in Fig. 2. In particular, the results for $CuK_2Cl_4 \cdot 2H_2O$, a low-temperature isotropic ferromagnet, are an anomaly which broadens with increasing field and a temperature of the maximum which first decreases and then increases with increasing field. No combinations of two types of anomalies were observed in this experiment, but a temperature-independent low-field susceptibility was reported. We make passing reference to their result on $Cu(NH_3)_4SO_4 \cdot H_2O$ because of the striking similarity of the heat-capacity curve for this material to the curves for large (but less than unity) h/d in Fig. 2. Clearly, the question of whether the two anomalies can be observed at the same field awaits further experiments.

We believe that Arrott's idea of the existence of a critical line in the H-T plane for an isotropic ferromagnet is essentially correct. However, we find that this line (which continues to $T=0$) represents transitions which are qualitatively different from the zero-field Curie point. We expect these transitions to be most readily observable when dipole-dipole interactions are not negligible compared with exchange interactions. Further experiments on low-temperature isotropic ferromagnets would appear to provide the best test of these ideas.

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