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LIQUID-VAPOR-LIKE CRITICAL POINTS IN ANISOTROPIC FERRIMAGNETS*

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A magnetic critical phase transition which involves no change in symmetry is described theoretically. In the example, $Yb_6Fe_{10}O_{24}$ with applied field along [100], a critical point is predicted at $T^c = 7.51°K$ and $H^c = 43\,640$ Oe. There is a large associated anomaly in C_H: $C_H(H=H^c) \approx 0.3(T-T^c)^{1/\delta-1} R$ mole^{-1}, where δ should realize the classical value 3.

Traditional phase transitions in magnetic systems are characterized by broken symmetry. In a space of external variables which do not themselves change the symmetry, the phases must be completely separated by phase transition boundaries. We describe here a magnetic transition between phases of like symmetry. The boundary need not be complete, and in general it should end in a critical point reminiscent of that in liquid-vapor systems. The critical point occurs for some substances in experimentally accessible magnetic fields, but no measurements of the predicted anomalies have yet been reported.

We illustrate the specific-heat anomaly at a critical point in ytterbium iron garnet (YbIG, $Yb_6Fe_{10}O_{24}$). Remarkably, true classical critical behavior is expected for this case. In other systems there should be some effect due to fluctuations. In any case the critical exponents expressing the nature of the singularities in thermodynamic functions should be different from any which have heretofore been observed.

As is well known[1] a ferrimagnet near its compensation temperature likes to assume a canted configuration in an applied field. The angle which a given sublattice makes with the field depends on the field magnitude and on the size of the various (temperature-sensitive) sublattice moments. Treating one particular sublattice angle as a free variable, we may say that the free energy (from exchange and Zeeman interactions) has a minimum at the equilibrium canting angle, and that this exchange minimum moves in angle as field and temperature vary. We add anisotropy by superimposing a background with wells at the sublattice easy directions.[2] In high fields the exchange minimum can be steeper than the wells and as it sweeps along (as, for example, when the temperature passes a compensation point) it can carry the sublattice continuously between them. In low fields the minimum is not so steep and the system will only be able to follow by jumping in first-order transitions to the an-

isotropy well nearest the exchange minimum. There is some intermediate field at which there occurs a critical point marking the end of the first-order phase boundary.

At the critical point the particular sublattice is just barely stable with regard to its mean position; however, it will not necessarily lose its internal rigidity. An analogy can be made to a ferromagnet which loses its anisotropy but not its intra-sublattice exchange: It becomes more susceptible to fluctuations but even so these will be important only at temperatures which are some fraction of the (somewhat reduced[3]) Curie temperature. From another point of view the internal rigidity of the sublattice effectively increases the range of interaction of each of its constituents with those on other sublattices. Thus, unlike real liquid-vapor critical points, here there need be no great buildup of fluctuations and classical critical behavior might be realized.

We now outline a very approximate but quantitative theory of liquid-vapor-like transitions in a two-sublattice ferrimagnet. A more rigorous approach (such as we will use in the YbIG example) requires detailed knowledge of the physical system under consideration.

We denote the sublattices by F and R, respectively. For a given direction of F, the exchange plus Zeeman free energy (neglecting here some entropy and susceptibility contributions) is

$$F^{ex} = -M_R(H^2+\lambda^2 M_F{}^2 - 2\lambda H M_F x)^{1/2} - HM_F x, \quad (1)$$

where M_F and M_R are the F and R magnetic moments (magnitude), H is the applied field, x the direction cosine that M_F makes with H, and λ is the positive (antiferromagnetic) intersublattice exchange. M_F and M_R are assumed to be functions of both H and T. We now minimize F^{ex} against x, take the second derivative at the minimum (called x^{ex}), and arrive at the following approximation to Eq. (1):

$$F^{ex} \approx F_0 + \tfrac{1}{2}[(M_F{}^2H^2)/\lambda M_R{}^2][x - x^{ex}(H,T)]^2, \quad (2)$$

where

$$x^{ex}(H,T) = (H^2+\lambda^2 M_F{}^2 - \lambda^2 M_R{}^2)/2\lambda H M_F, \quad |x^{ex}|<1,$$

$$|x^{ex}| = 1 \quad (3)$$

otherwise.[4] F^{ex} is our exchange minimum; its position moves as $x^{ex}(H,T)$ changes and its "steepness" increases as H^2.

Anticipating the case of YbIG, we represent anisotropy by a term $A(x^2-\frac{1}{3})^2$ with A positive. Adding this to Eq. (2) and minimizing yields the following equation of state for x:

$$x^3 + x[M_F{}^2H^2/4A\lambda M_R{}^2 - \tfrac{1}{3}] - (M_F{}^2H^2/4\lambda A M_R{}^2)x^{ex}(H,T) = 0 \quad (4)$$

and the solutions have the following properties:

(i) For $M_F{}^2H^2/4A\lambda M_R{}^2 - \frac{1}{3} \equiv p > 0$, one real root. There are no discontinuities in x as $x^{ex}(H,T)$ varies. The exchange minimum is steeper than the anisotropy wells.

(ii) For $p<0$, three real roots for small x^{ex}, two stable. Lowest free energy is one with the same sign as x^{ex}. There is a first-order phase transition when x^{ex} changes sign. The anisotropy well is steeper than the exchange minimum.

(iii) For $p=0$, critical point at $x^{ex}=0$. The size of the critical field (H^c) is of order $(H^a H^{ex})^{1/2}$ where H^a and H^{ex} are sublattice anisotropy and exchange fields, respectively.

All critical exponents have their classical values.[5] Particularly, the specific heat at constant field (C_H) along $H=H^c$ behaves as $(T-T^c)^{1/\delta-1}$ where T^c is the critical temperature and $\delta=3$.

YbIG is a system where a mean-field treatment is extremely well justified. The Fe sublattice (F) has a strong internal exchange coupling equivalent to about 550°K. The Fe ions couple anisotropically to Yb ions (R sublattice) on six inequivalent sites (exchange energy about 25°K) while the Yb's couple very weakly (less than 1°K) to each other. At the critical temperature of about 8°K the unstable iron sublattice is too rigid to support fluctuations while the Yb system is essentially noncooperative (it is weakly coupled and in large exchange and magnetic fields). Thus a given Fe spin, constrained to maintain the mean Fe direction, cares almost as much about the state of distant Yb's as it does about its neighbors. The F-R interaction is effectively long range, so is truly represented by the mean field.

The theoretical field-temperature phase diagram of YbIG for field along [100] is shown in Fig. 1. The outer phase boundary, between aligned and disaligned states, cannot end; there is always a second-order phase transition. The

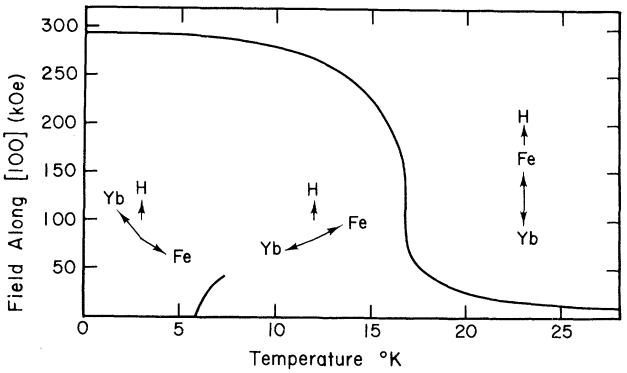

FIG. 1. Phase diagram for YbIG with field along [100]. The critical point occurs at the end of the first-order phase boundary which starts from the T axis at 6°. The outer phase boundary is second order.

first-order phase transition near the compensation point (6°K), on the other hand, is between states of the same symmetry, and there are paths between these states which never traverse a state of higher symmetry. This phase boundary could end. In our calculation for YbIG it does end at a field of 43 620 Oe and a temperature of 7.51°K. This then is an example of a liquid-vapor-like critical point in a real ferrimagnet.

The specific heat at six constant fields in the neighborhood of the critical point has been calculated and is shown in Fig. 2. The calculation employed the data of Wickersheim[6] for anisotropic g and G factors and those of Harris and Meyer[7] for the lattice contribution. The mean-field formulation of Wolf et al.[8] was used for calculating the state of the Yb's and various numerical interation schemes were employed to find the minimum energy direction of the Fe lattice on the unit sphere.[9] Since the magnetization changes by only about 5 emu near T^c, the demagnetization correction is unimportant.

As it turns out the approximate treatment [Eq. (4), etc.] contains the major features of the transition. (There is a quantitative discrepancy but the functional forms should be correct.) Let $A = -\frac{3}{4}K_1 - \frac{1}{3}K_2 = 2\times10^6$ erg cm^{-3}, $M_F = 200$ emu cm^{-3}, $\lambda = 700$, $M_R = 200 + 3.6(T^c - T)$. These imply that (quite accurately) $H^c \approx (4/3\lambda A)^{1/2} = 42 \times 10^3$ Oe. Also we have $x = 0.27(T - T^c)^{1/3}$ for $H = H^c$. Thus

$$C_H(H^c) = 1.1\times10^5(T-T^c)^{-2/3} \text{ erg cm}^{-3}\,^\circ\text{K}^{-1}$$

$$= 0.36(T-T^c)^{-2/3}R \text{ mole}^{-3}. \quad (5)$$

Above H^c, C_H does not diverge but rises to a peak whose maximum is $[5\times10^3/(H-H^c)]R$ mole^{-1} and whose width in temperature is about $4\times10^{-6}(H-H^c)^{3/2}$. Below H^c the maximum value of C_H before the first-order phase change is $[2.5\times10^3/(H^c-H)]R$ mole^{-1}. These effects are thus seen to be quite large, and it seems that efforts to observe them would be well justified.[10]

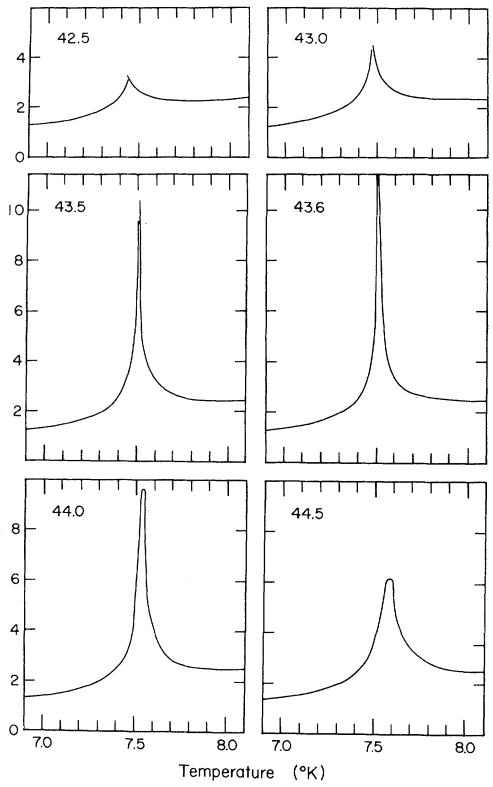

FIG. 2. Specific heat (R/mole) at constant applied field near the critical point. The fields (kOe) are indicated at the upper left of each plot. At the critical field C_H behaves as $(T-T_c)^{-2/3}$. Results are from numerical calculations using data of Refs. 6 and 7.

In YbIG we expect mean-field (classical) critical behavior because the temperature is so far below the disordering (Curie) temperature of the cooperative sublattice. There are surely other cases where the sublattice is far from saturation near the critical temperature. Then there would be fluctuations and the vanishing of apparent anisotropy would change their character; the critical behavior would be nonclassical.

In summary we have described a magnetic critical phase transition with two new aspects. First there is no change in symmetry, and so the coexistence curve ends at some point in the H-T plane, not necessarily on the $H = 0$ axis. Second, and more significant, the role of fluctuations is mediated by the residual order and so

the critical behavior is different from that in typical order-disorder transitions.

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NEUTRON-MATTER RADII FROM AN ANALYSIS OF PIONIC ATOMS*

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We use the low-energy pion-nucleus interaction as a means for investigating the distribution of nucleons in nuclei. An optical-model analysis of existing experimental data on pionic atoms is used to study possible differences in the rms radii of the neutron and proton densities. The difference in these radii, assumed to be independent of mass number, was found to be -0.01 ± 0.16 F. The absence of any significant difference in these radii should be considered an average behavior for a wide range of nuclei.

The present situation concerning possible differences in the rms radii of the neutron and proton distributions in nuclei is not clear. For example, Greenlees, Pyle, and Tang[1] used an optical-model analysis of proton-nucleus elastic scattering for several nuclei from Ni^{58} to Pb^{208}, and found that the rms radius of the neutrons is about 0.6 F larger than the rms radius of the protons. This result is supported by shell-model analyses of single-particle energies,[2] although a more recent study[3] indicates that the difference in these radii approaches zero in the lighter nuclei. In contrast to these studies, investigations of Coulomb displacement energies[4] lead to the general conclusion that the rms radius of the neutron distribution is just slightly larger than that of the proton distribution.

In this Letter we use pionic atoms because of the strong pion-nucleus interaction which furnishes an additional means for studying the distribution of neutrons and protons in nuclei. The possibility of using pionic atoms as a tool for the investigation of nucleon distributions has previously been recognized,[5] although in these studies no attempt was made to examine possible differences in the nucleon densities. However, the utility of this probe has been exploited in a study of pion atomic transitions in the nickel isotopes,[6] and it was found that the experimental data are consistent with equal rms radii for the neutron and proton densities.

Experimentally, the pion atomic-transition energies and the energy-level widths (due to nuclear absorption of the pions) are measured, and these quantities provide information concerning the pion-nucleus interaction. The energy levels and widths can be calculated by finding the (complex) eigenvalues E of a modified Klein-Gordon equation

$$\{\nabla^2 + 2m_r[(E - V_{em}) + (E - V_{em})^2/(2m_\pi)]\}\Psi = 2m_r V_{opt}\Psi, \quad (1)$$

where m_π and m_r denote the pion and pion-nucleus