PHYSICAL REVIEW B VOLUME 38, NUMBER 16 1 DECEMBER 1988

Ferromagnetism of ultrathin films

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(Received 10 August 1988)

We discuss the nature of ferromagnetism in ultrathin films of magnetic ions, here regarded as two-dimensional Heisenberg ferromagnets subject to uniaxial anisotropy with the easy axis normal to the film. We show that a phase transition to ferromagnetism occurs always for arbitrarily small anisotropy. Renormalization-group scaling relations for the transition temperature and the temperature variation of the correlation length are obtained. Implications of these results are discussed.

Remarkable advances in materials technology now allow preparation of ultrathin films (monolayer, bilayer, etc.) of extraordinary quality, from the magnetic transition metals Fe, Co, and Ni. These are deposited on single-crystal substrates,[1] or incorporated within multilayer structures.[2]

The nature of the magnetism in such ultrathin films is of fundamental interest, since it may differ from that realized in bulk single crystals by virtue of their reduced dimensionality. In the absence of dipolar couplings, and anisotropies of spin-orbit origin, these materials can be regarded as physical realizations of the two-dimensional Heisenberg ferromagnet.[3] Thus, there can no longer be range order at any finite temperature in this picture.[4] The recent Mössbauer study of monolayer Fe films described by Koon *et al.*[2] shows that at and considerably below room temperature, large-amplitude, slow-spin fluctuations are present. These are evident as broadening of the spectra; in fact, resolvable structure appears only below room temperature. These results are consistent with the absence of long-range ferromagnetic order in the spin system.

At the same time, various studies,[1,2] including the Mössbauer experiment just discussed, established that in monolayer Fe films, uniaxial anisotropy with the easy axis normal to the film is present. Theoretical studies of an isolated Fe monolayer yield spin-orbit-induced anisotropies with magnitudes similar to those observed.[5,6] A question then arises. Is the presence of such anisotropy, possibly very weak compared to the effective exchange energies in a material such as Fe, able to induce a phase transition in two dimensions to a ferromagnetic state with true long-range order? This question was answered in the affirmative by Khokhlachev,[7] within a mean-field approach. This issue was also discussed by Pelcovits and Nelson,[8] within the framework of a renormalization-group treatment that outlines the nature of the phase transition, upon assuming that a fixed point exists. The present paper explores the renormalization-group analysis, and then proves that a phase transition of Ising character does indeed exist. We address the experimental work discussed above in an explicit manner.

We proceed as follows. If H is the Hamiltonian, and $\beta=1/k_BT$ with k_B Boltzman's constant, and T the temperature, we begin with

$$\beta H=\frac{J}{2T}\int d^2x(\nabla\mathbf{S})^2-\frac{K}{2Ta_0^2}\int d^2x S_z^2(x)\,, \tag{1}$$

with J a measure of the strength of the exchange coupling, K the anisotropy constant, and a_0 the lattice constant. (Recall that many years ago, it was argued that this form applies to long-wavelength spin fluctuations in both itinerant and localized spin systems.[9]) While our interest is in the case where the order parameter $\mathbf{S}$ has three components, we generalize to the case where it has N components, with the Hamiltonian still given by Eq. (1).

In a continuum theory such as described by Eq. (1), the values assumed by J and K depend on the choice of the underlying microscopic length scale a_0. Thus, these two parameters may be viewed as functions of a_0. In the critical region, a rescaling of a_0 can lead to unchanged physics if we modify T/J and K/T appropriately. Following Polyakov,[10] we obtain the renormalization-group equations

$$\frac{J}{T(\tilde{a}_0)}=\frac{J}{T(a_0)}+\frac{N-2}{2\pi}\ln\left[\frac{a_0}{\tilde{a}_0}\right], \tag{2a}$$

and

$$\frac{K(\tilde{a}_0)}{\tilde{a}_0^2T(\tilde{a}_0)}=\frac{K(a_0)}{a_0^2T(a_0)}\left[1+\frac{N}{2\pi}\frac{T(a_0)}{J}\ln\left[\frac{a_0}{\tilde{a}_0}\right]\right]. \tag{2b}$$

From these, we show below that the combinations

$$\frac{J}{T(a_0)}+\frac{(N-2)}{2\pi}\ln(a_0)$$

and

$$[K(a_0)/Ja_0^2][T(a_0)/J]^{2/(N-2)}$$

are invariant under changes of a_0.

Requiring that the expression for the correlation length $\lambda(T)$ also be invariant in form under such rescaling leads to

$$\frac{1}{\lambda(T)}=\frac{1}{a_0}e^{-[2\pi J/(N-2)T]}\times F\left[e^{[4\pi J/(N-2)T]}\frac{K}{J}\left[\frac{T}{J}\right]^{2/(N-2)}\right]. \tag{3}$$

Our method is not powerful enough to yield the form of

the function F. A critical point will occur at a temperature at which the correlation length becomes infinite; thus, at the critical temperature, F has a zero. It follows that this critical temperature T_2 for the two-dimensional system obeys the following scaling law:

$$\frac{K}{J}\left(\frac{T_2}{J}\right)^{2/(N-2)}\exp\left(\frac{4\pi J}{(N-2)T_2}\right)=\text{const}. \tag{4}$$

The result in Eq. (4) is obtained as follows: Equation (2a) may be rearranged to read

$$\frac{1}{\tilde{a}_0-a_0}\left(\frac{J}{T(\tilde{a}_0)}-\frac{J}{T(a_0)}\right)=\frac{N-2}{2\pi}\left(\frac{\ln a_0-\ln\tilde{a}_0}{\tilde{a}_0-a_0}\right), \tag{5a}$$

or in the limit $\tilde{a}_0\rightarrow a_0$, we have

$$\frac{\partial}{\partial a_0}\frac{J}{T(a_0)}=-\frac{N-2}{2\pi}\frac{\partial}{\partial a_0}\ln a_0, \tag{5b}$$

which can be arranged to read

$$\frac{J}{T^2}a_0\frac{\partial T}{\partial a_0}=\frac{N-2}{2\pi}. \tag{5c}$$

A similar treatment of Eq. (2b) gives

$$a_0\frac{\partial}{\partial a_0}\left(\frac{K(a_0)}{a_0^2T(a_0)}\right)=-\frac{N}{2\pi J}\frac{K(a_0)}{a_0^2}, \tag{6}$$

which may be combined with Eq (5c) to give

$$a_0\frac{\partial}{\partial a_0}K(a_0)=2K(a_0)-\frac{1}{\pi J}K(a_0)T(a_0). \tag{7}$$

The results in Eqs. (5c) and (7) are identical to the first and last of Eq. (4) of Pelcovits and Nelson.[8(a)]

From Eqs. (5a) and (7), one may show that the derivative of the combinations $(K/Ja_0^2)(T/J)^{2/(N-2)}$ vanishes, so this quantity is independent of a_0. Using the fact that $J/T(a_0)+(N-2/2\pi)\ln a_0=\text{const}$, as follows from Eq. (5b), we may replace a_0 by $\exp[-2\pi J/(N-2)T]$, and the result in Eq. (4) then follows. Thus, the correlation length must obey a relation such as that given in Eq. (3). We note two limits: the physically interesting case of $N=3$,

$$\frac{K}{J}\left(\frac{T_2}{J}\right)^2\exp\left(\frac{4\pi J}{T_2}\right)=\text{const}, \tag{8}$$

and the large-N result

$$\frac{K}{J}\exp\left(\frac{4\pi J}{NT_2}\right)=\text{const}. \tag{9}$$

Further on, in the large-N limit, we evaluate the constant on the right-hand side of Eq. (9).

The renormalization-group calculation presented here and by Pelcovits and Nelson is a "one-loop" calculation, and is thus valid for a limited range of parameters; Polyakov[10] [his Eq. (17)] shows the region of validity is

$$\frac{1}{T}\gg\frac{N-2}{\pi}\ln\left(\frac{\tilde{a}_0}{a_0}\right). \tag{10}$$

The argument presented above, as many other renormalization-group analyses, cannot determine whether there is a phase transition or not; one can use such an approach to decide if there is a phase transition, *then* the scaling relations obtained must hold. This is the point of view put forward by Pelcovits and Nelson.

We now establish that there is indeed a phase transition. Within this limit, we establish that the character of the phase transition is Ising-like, and in the large-N limit we obtain an explicit expression for the temperature T_2 of the phase transition.

To see this, we rewrite the partition function corresponding to Eq. (1) as

$$Z=\int\prod[dS(\mathbf{x})d\alpha(\mathbf{x})]\exp\left[-\frac{J}{2T}\int(\nabla\mathbf{S})^2d^2x-\frac{1}{2a_0^2}\int\alpha^2(\mathbf{x})d^2x-\left(\frac{K}{T}\right)^{1/2}\frac{1}{a_0^2}\int\alpha(\mathbf{x})S_z(\mathbf{x})d^2x\right]. \tag{11}$$

The Gaussian integration over α gives back the original Hamiltonian. For $\alpha(\mathbf{x})$ fixed, we may view Eq. (7) as the partition function of an isotropic Heisenberg spin system in an external-external magnetic field $\mathbf{h}(\mathbf{x})=\sqrt{TK}\,\alpha(\mathbf{x})\hat{z}$. As, for any finite T, this system has no critical point, the free energy may be expanded as a power series in the external field

$$Z=\int\prod d\alpha(\mathbf{x})\exp\left[\int F_0(T/J)\frac{d^2x}{a_0^2}-\frac{1}{2a_0^2}\int d^2x\alpha^2(\mathbf{x})+\frac{K}{2}\int\frac{d^2xd^2y}{a_0^4}\alpha(\mathbf{x})G(\mathbf{x}-\mathbf{y})\alpha(\mathbf{y})+[\text{terms quartic in }\alpha(\mathbf{x})]+\cdots\right]. \tag{12}$$

Here $G(\mathbf{x}-\mathbf{y})$ is the two-point correlation function of the Heisenberg system.

The form in Eq. (12) may be viewed as a ϕ^4 field theory for α with the interaction range governed by the correlation length of the Heisenberg system. *The system will thus have a critical point corresponding to Eq. (4), with Ising model critical exponents.*

One may also use Eq. (11) as a starting point for a large-N expansion, where the quadratic term in α is of higher order in N than the quartic and higher terms. For $T>T_2$, we may keep only this term. The large-N correlation function and correlation length are also known. Thus, the effective theory for the field $\alpha(\mathbf{x})$ (in momentum space) is

$$Z=\int\prod d\alpha(\mathbf{q})\exp\left[-\tfrac{1}{2}\int d^2q\,\alpha(-\mathbf{q})\times\left[1-\frac{K}{a_0^2}G(\mathbf{q})\right]\alpha(\mathbf{q})\right],$$

with

$$JG(\mathbf{q}) = (q^2 + m^2)^{-1}, \tag{13a}$$

and

$$m^2 = \frac{\pi^2}{a_0^2} e^{-4\pi J/NT}. \tag{13b}$$

The effective correlation length for the α field then satisfies

$$\frac{1}{\lambda^2} = m^2 - \frac{K}{Ja_0^2} = \frac{1}{a_0^2}\left[\pi^2 e^{-4\pi J/NT} - \frac{K}{J}\right], \tag{14}$$

so we have

$$\lim_{N\to\infty} T_2 = \frac{4\pi J}{N} \frac{1}{\ln(\pi^2 J/K)} \tag{15}$$

in agreement with Eq. (9).

In the same limit, $T_3 = 4\pi J/N$ is the transition temperature to the ferromagnetic state of the *three*-dimensional N component system. Using this result for $N = 3$, we obtain an estimate for the transition temperature T_2 of the two-dimensional system

$$T_2 = T_3 / \ln\left(\frac{3\pi}{4}\frac{T_3}{K}\right). \tag{16}$$

The presence of uniaxial anisotropy thus always produces a phase transition to ferromagnetism of Ising character at finite temperatures, no matter how weak the anisotropy (provided, of course, that $K > 0$). Furthermore, the logarithmic dependence of T_2 on K given by Eq. (13) shows T_2 can be quite substantial, even for very weak anisotropy. Khokhlachev's mean-field treatment provides an expression for T_2 similar to that in Eq. (15), but with N replaced by $N-2$; he confines his attention only to the case $N = 3$. We prefer to extrapolate our large-N expression down to $N = 3$, viewing this as a procedure more reliable than mean-field theory.

We may estimate K for a monolayer as follows, when the easy axis is normal to the surface. For the monolayer, assume K is dominated by the spin-orbit-coupling contribution $K_{\text{s.o.}}$. Then for a film of finite thickness, with d layers, an effective dipolar anisotropy $-4\pi M_s$ develops, which acts on spins in each layer. Thus, with increasing film thickness, the film is described by an effective anisotropy constant K_{eff} which decreases, eventually to become negative at a certain number of layers d_c. Indeed, the data show that for films thicker than four or five layers, the magnetization is parallel to the surface.[1,2] Thus, $d_c \cong 5$, and $2K_{\text{s.o.}} \cong 4\pi M_s d_c$, assuming the spin-orbit contribution to act only in the surfaces and that it has the same magnitude on each. With $4\pi M_s \simeq 2\times 10^4$ G, we have $K_{\text{s.o.}} \cong 5\times 10^4$ G, or in temperature units, $K_{\text{s.o.}} \sim 10$ K. Hence, with $T_3 = 10^3$ K, $\ln(3\pi T_3/4\text{ K}) \sim 6$, and for the monolayer we estimate $T_2 \sim T_3/6 \cong 150$ K. This is well below room temperature but substantial, considering the modest value of K/J. Ordering of the two-dimensional Heisenberg ferromagnet is "triggered" easily by a weak, spatially uniform perturbation.

Our estimate of T_2 is consistent with the Mössbauer data of Ref. 2, which show that in monolayer films of Fe fluctuations in the spin system slow dramatically below room temperature. Unfortunately, such a localized probe has difficulty sensing the onset of a true phase transition. Measurements of the susceptibility of the spin system would prove of great interest.

As remarked above, when the easy axis is normal to the surface, the dipolar contribution to the anisotropy constant will cause K_{eff} to decrease with increasing thickness d and to change sign as d passes d_c. Thus, we expect T_2 to decrease initially as the film thickness increases, and when K_{eff} becomes negative there will be a crossover to XY behavior at low temperatures; long-range order is then absent at $T = 0$, since the correlation length remains finite there [Eq. (14), with $K < 0$].

The above comments on the variation of K_{eff} with d apply only to the case where the easy axis is normal to the surface. An easy axis in the plane will cause K_{eff} to be roughly proportional to d itself, assuming the spin-orbit anisotropy to dominate and be the same for each plane of spins. Then T_2 will increase with d. The present theory does not describe the crossover to three-dimensional behavior as d increases. Also, at this time the physical origin of the in-plane anisotropy is not clear in our view, so it is difficult to be precise about the dependence of T_2 on d when the magnetization lies in plane in the ordered state.

It would be of great interest to measure K_{eff} as a function of film thickness, along with T_2. In principle, K_{eff} could be measured by the Brillouin scattering of light from spin waves, excited at temperatures $T \ll T_2$. This technique has been applied to the study of ultrathin films by Hillebrands and co-workers, who report spectra taken from films with thicknesses in the few-monolayer range.[11]

The purpose of this paper has been to outline, on theoretical grounds, the behavior expected for a very thin two-dimensional Heisenberg ferromagnet, subject to the uniaxial anisotropy elucidated in recent experiments. It is our aim to provide a framework within which subsequent data may be discussed.

The research of one of us (M.B.) is supported by National Science Foundation Grant No. PHY-86-05552, and that of the other (D.L.M.) is supported by the Army Research Office through Contract No. CS0010238.

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[6]An elementary argument produces anisotropy energies comparable to those observed in the monolayers. In Fe, the anisot-

ropy energy may be obtained by expanding the ground-state energy in powers of the spin-orbit coupling constant λ. Thus, the single-site quartic-anisotropy energies in the bulk of this cubic material are roughly λ^4/W^3, with W a typical electron energy denominator, here taken as the width of the $3d$ band ($\sim$4 eV). In the monolayer, where the z axis normal to the monolayer is inequivalent to the x and y directions, the single-site anisotropy constant K is of order λ^2/W, where $\lambda \sim 2\times 10^{-2}$ eV for Fe. This estimate is in rough accord with the data; of course, the simple argument fails to give the sign of K.

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