Dipole forces and phase transition in a two-dimensional planar ferromagnet

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The effect of dipole forces on the Kosterlitz-Thouless phase transition is considered. It is shown that dipole forces are responsible for the existence, in the vicinity of T_c, of a narrow critical region within which they play a dominant role. In this region, self-consistent field theory is valid to within logarithmic corrections; the spontaneous moment varies by an amount of the order of the saturation moment. The results are also applicable to layered magnetic materials with sufficiently weak interplane coupling, and to films of ferroelectric smectic C.

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1. INTRODUCTION

In recent years, intensive theoretical investigations have been carried out on two-dimensional (2D) degenerate systems. As is well known,[1] in such systems, with local interaction, the order parameter $\langle \mathbf{m} \rangle$ is zero at nonzero temperature. The formal reason for this is divergence of the integral that determines the mean square of the fluctuation of the order parameter:

$$\langle \delta \mathbf{m}_\perp^2 \rangle \sim T \int \frac{d^2k}{k^2}.$$

Therefore the method of the self-consistent field and the 4-ε expansion, which has it as a starting point, are completely inapplicable to such systems.

A 2D XY model was studied by Berezinskii.[2] The existence of a phase transition was demonstrated; the low-temperature phase is characterized by the presence of a stiffness ρ_s with respect to transverse fluctuations of the moment (an analog of the superfluid density in He II) and by a gradual decrease of the spin-spin correlator with distance. Pokrovskii and Uimin[3] investigated the behavior of a system in various weak fields from the point of view of scale invariance. Kosterlitz and Thouless[4,5] investigated the phase transition in this model (see also Refs. 6 and 7). On the other hand, experimental studies are now being made on systems which, in the "zeroth" approximation, are described by the 2D XY model. These include thin superfluid films of ^{4}He,[8] a two-dimensional crystal of atoms adsorbed on a surface,[9] and an easy-plane (planar) layered ferromagnet with weak interplane exchange.[10-12] Communications have recently appeared on the synthesis of a two-dimensional ferromagnet[13] and on experiments with thin films (a few molecular layers thick) of ferroelectric smectic C[14] (on the properties of bulk smectic C, see Ref. 15).

Ferromagnetic and ferroelectric systems are especially suitable for investigation, since there exist external fields (magnetic and electric) that are directly related to the order parameter. But in these systems there also exists the nonlocal dipole-dipole interaction (DDI). Strongly developed fluctuations produce an anomalously large susceptibility in degenerate 2D systems. Therefore DDI, because of its long range, strongly influences the properties of the system, despite its small intensity as regards local forces. The low-temperature phase of a 2D Heisenberg ferromagnet, with allowance for DDI, was investigated by Maleev[16] and by Pokrovskii and the author.[17] It was shown that DDI leads to an effective anisotropy of the "easy plane" type and to the appearance of a term in the energy of the fluctuations that is linear in the momentum; therefore a spontaneous moment appears at sufficiently low temperature.

The present paper studies the phase transition in a 2D planar ferromagnet. It is shown that in this system a single phase transition occurs, at which the stiffness ρ_s and the spontaneous moment disappear simultaneously, and that DDI leads to a shift of the transition temperature as compared with the purely exchange XY model:

$$(T_c - T_{c0})/T_c \sim \ln^{-2}(1/\mu^2)$$

(μ^2 is the ratio of the intensity of the dipole to that of the exchange interaction). In the vicinity of T_c there is a critical region, of width $\sim T_c \ln^{-3}(1/\mu^2)$, where DDI plays a dominant role, provided such large distance scales $r \sim k^{-1}$ are important that the dipole term $\sim \mu^2 k$ in the energy of the fluctuations becomes larger than the exchange term $\sim k^2$. Here a "parquet" situation arises in the two-dimensional theory, as in four-dimensional phase transitions with local interaction and in three-dimensional with participation of dipole forces.[18] The applicability of the results to layered magnets[10-12] and to films of ferroelectric smectic C[14] is discussed.

2. TWO-DIMENSIONAL HEISENBERG FERROMAGNET

It was shown earlier[17] that in the study of large-scale fluctuations, the Hamiltonian of a 2D ferromagnet reduces to the effective Hamiltonian of the 2D XY model with DDI:

$$\mathscr{H} = \frac{1}{2} \int (\nabla \mathbf{m})^2 d^2x - \frac{\mu^2}{2} \iint \frac{\mathbf{m}(\mathbf{x})\mathbf{m}(\mathbf{x}')}{|\mathbf{x}-\mathbf{x}'|^3} d^2x\, d^2x' - \int \mathbf{h}\mathbf{m}(\mathbf{x}) d^2x. \qquad (1)$$

Here $\mathbf{m}$ is a plane vector of unit length. A system of units is adopted in which the lattice constant a, the exchange integral J, and the length of the spin vector $\mathbf{S}$

are unity.

The important small parameter of the problem is $\mu^2 \sim 10^{-2}$. The seed correlator corresponding to the Hamiltonian (1) is

$$D_0(\mathbf{k}) = \frac{1}{T}\langle|\omega_\mathbf{k}|^2\rangle = \frac{1}{k^2+R_0^{-1}k+h}, \quad R_0^{-1}=4\pi\mu^2, \tag{2}$$

where the angular variable $\omega_\mathbf{x}$ has been introduced,

$$\mathbf{m}(\mathbf{x}) = (\cos\omega_\mathbf{x}, \sin\omega_\mathbf{x}),$$

and $\omega_\mathbf{k}$ is a Fourier component of $\omega_\mathbf{x}$.

On scales less than the dipole radius[17] $R = R_0^{1/(1-2\Delta)}$, the behavior of the system is scale-invariant:[2,3]

$$K(\mathbf{x}) = \langle\mathbf{m}(0)\mathbf{m}(\mathbf{x})\rangle = x^{-2\Delta}, \quad \Delta = T/4\pi\rho_s(T). \tag{3}$$

Besides small fluctuations, there exist in the system topological excitations-vortices.[2,4,5,7] They form a two-dimensional Coulomb gas with logarithmic interaction. In the low-temperature phase of the *XY* model, the vortices are connected with neutral quasimolecules and do not substantially affect the properties of the system. With rise of temperature, there occurs a phase transition due to dissociation of quasimolecules. The vortex gas goes over to a plasma state; the Coulomb interaction is screened; the spin system finds itself in a disordered phase.

Kosterlitz[5] constructed for this phase transition a renormalization procedure that enables one to determine exactly the index of the correlator $K(\mathbf{x}) \sim x^{-\eta}$ at the transition point: $\eta = 2\Delta(T_c) = \frac{1}{4}$ [this implies finitness of $\rho_s(T)$ at $T \to T_c - 0$]. It also enables one to find the basic dependence of the correlation radius ξ_0 on the closeness to the transition point in the high-temperature phase, $\tau_0 = (T - T_{c0})/T_{c0}$:

$$\xi_0(\tau) \sim \exp(C/\tau_0^{1/2}), \quad C\sim 1.$$

Since the singular part of the free energy is

$$F_{sing}^{(0)}(\tau_0) \sim \xi_0^{-2} \sim \exp(-2C/\tau_0^{1/2}),$$

the specific heat and all its derivatives are continuous at $\tau_0 = 0$.

To avoid misunderstanding, we remark that the jump in ρ_s at the transition point does not imply a transition of the first kind, since ρ_s is not an order parameter. In fact, the system can be described by a function $\rho_s(k^2)$ that plays the role of dielectric susceptibility in the vortex gas. The interaction of the vortices in momentum space is $V(k) = \rho_s(k^2)/k^2$. In the low-temperature phase, $\rho_s = \rho_s(0) \neq 0$. Above the transition point, $\rho_s(k^2) \sim k^2/(k^2 + \xi_0^{-2})$; that is, in a broad range of scales $1 \ll r \ll \exp(C/\tau_0^{1/2})$ the behavior of the system does not differ from that of the low-temperature phase.

We now consider the effect of DDI on the phase transition. It is easy to show that when $\rho_s \neq 0$, DDI leads to a linear increase of the energy of attraction of vortices at distances larger than the dipole radius R. Therefore a phase transition can occur as soon as the vortices will form a plasma on scales of the order of R. In other words, the correlation radius ξ_0 without allowance for DDI must become of the order of the dipole radius R. When $\xi_0 \gtrsim R$, the system is in an order phase; when $\xi_0 \lesssim R$, in a disordered. Therefore the width of the critical region δ is determined by the conditions $\xi_0 \sim R$ and $\delta d\xi_0/d\tau_0 \sim R$. Hence we obtain an estimate of the shift of the transition temperature,

$$\tau_0 = \frac{T_c - T_{c0}}{T_{c0}} \approx \frac{1}{\ln^2 R} \tag{4}$$

and the width of the critical region,

$$\delta \approx \tau_0^{3/2} \approx 1/\ln^3 R \ll \tau_0. \tag{5}$$

We note an important difference from the usual transitional effects (crossover) (for example in 3D ferromagnets[19]), where weak fields become important in a narrow fluctuational neighborhood of the transition point. In the 2D *XY* model, the whole low-temperature region is, in essence, fluctuational; the spontaneous moment, for example, is determined by DDI at all $T < T_c$. Therefore as soon as $T \lesssim T_c - \delta T_c$, the spontaneous moment S is determined as at low temperatures,

$$S = |\langle\mathbf{m}\rangle| \approx (\mu^2)^{\Delta_c/(1-2\Delta_c)} = (\mu^2)^{1/6} \sim 1,$$

the change of S from zero to a value of the order of the saturation moment occurs over a narrow interval $|T - |T - T_c| \sim T_c \ln^{-3}(1/\mu^2)$.

3. EFFECTIVE HAMILTONIAN IN THE CRITICAL REGION

In the critical region $|\tau| = |T - T_c|/T_c \ll \delta$, the correlation distance $\xi \gg R$; therefore in the Hamiltonian (1), the second term is larger than the first. Since the phase transition is due (in contrast to the *XY* model without DDI) to the appearance of a spontaneous moment $\langle\mathbf{m}\rangle \neq 0$, the critical behavior of the system is described by a functional of the Ginzburg-Landau type:

$$\mathcal{H}_c(\mathbf{m}) = -\frac{(\mu^2)^{1/(1-2\Delta)}}{2}\iint \frac{\mathbf{m}(\mathbf{x})\mathbf{m}(\mathbf{x}')}{|\mathbf{x}-\mathbf{x}'|^3}\,d^2x\,d^2x' + \int\left[\frac{a\tau}{2}\mathbf{m}^2(\mathbf{x}) + \frac{\lambda}{4}\mathbf{m}^4(\mathbf{x}) - \mathbf{h}\mathbf{m}(\mathbf{x})\right]d^2x.$$

Here already $\mathbf{m}^2 \neq 1$, and the modulus of the order parameter fluctuates strongly. The coefficient of the first term is written, with allowance for renormalization, at distances $r < R$:[17]

$$\mu_{eff}^2 = (\mu^2)^{1/(1-2\Delta)}.$$

The parameters a and λ are determined by the conditions

$$a\tau = \xi^{-2} \sim R^{-2} \text{ when } \tau\sim\delta,$$
$$|\langle\mathbf{m}\rangle|^2 \sim (\mu^2)^{1/3} \text{ when } -\tau\sim\delta.$$

Therefore

$$a = AR^{-2}\delta^{-1}, \quad A\sim 1; \quad \lambda = g_0R^{-2}, \quad g_0 \sim (\mu^2)^{-1/3}.$$

On introducing the new variable $\varphi = R^{-1/2}\mathbf{m}$ and transforming to the momentum representation, we get

$$\mathcal{H}_c(\varphi) = \frac{1}{2}\sum_\mathbf{k}(k+t_0)\varphi_\mathbf{k}\varphi_{-\mathbf{k}} + \frac{g_0}{4}\sum_{\mathbf{k}_1+\mathbf{k}_2+\mathbf{k}_3+\mathbf{k}_4=0}(\varphi_{\mathbf{k}_1}\varphi_{\mathbf{k}_2})(\varphi_{\mathbf{k}_3}\varphi_{\mathbf{k}_4}) - R^{1/2}\sum_\mathbf{k}\mathbf{h}_\mathbf{k}\varphi_\mathbf{k}, \tag{6}$$
$$t_0 = A\tau/R\delta.$$

The seed correlator at $h = 0$ is

$$G_0(\mathbf{k}) = \frac{1}{T}\langle\varphi_\mathbf{k}\varphi_{-\mathbf{k}}\rangle = \frac{1}{k+t_0}. \tag{7}$$

The momenta $k \lesssim \Lambda \sim R^{-1}$.

It is easily seen that the physical dimensions of the field φ are $(\text{length})^{-1/2}$; therefore the coupling constant g_0 is dimensionless. A calculation of the corrections for the interaction leads to parquet equations analogous to the equations of 4D local theory:[18,20]

$$\frac{dg(l)}{dl}=-\frac{10}{2\pi}g^2(l), \quad l=\ln\frac{r}{R}, \tag{8a}$$

$$\frac{dt(l)}{dt_0}=\mathcal{T}(l), \tag{8b}$$

$$\frac{d\mathcal{T}(l)}{dl}=-g(l)\mathcal{T}(l)\frac{4}{2\pi}, \tag{8c}$$

where $g(l)$ and $t(l)$ are the renormalized coupling constant and gap on a scale $r \sim \mathrm{R}e^l$.

We solve (8) with logarithmic accuracy:

$$g(l)=\frac{g_0}{1+g_0\cdot 10l/2\pi}, \tag{9a}$$

$$t(l)=t_0(1+g_0\cdot 10l/2\pi)^{-2/5}. \tag{9b}$$

Equations (8) are valid when $g(l) \ll 1$. In our case $g_0 \approx (\mu^2)^{-1/3} \sim 5$. Therefore the theory set forth may be applied, if we assume that, starting with such g_0's, we drop immediately through renormalizations to the "zero-charge" trajectory (9). In this case one can at once take for the solutions (9) the asymptotic expressions for $l \gg 1$.

Returning to the previous variables, we get, as in Ref. 18, expressions for the correlation distance ξ, the magnetic susceptibility $\chi = \partial S/\partial h$, and the singular part of the specific heat C_{sing} in the critical region:

$$\xi(\tau) \sim R\frac{\delta}{|\tau|}\ln^{2/5}\frac{\delta}{|\tau|}, \tag{10}$$

$$\chi(\tau) \sim R^2\frac{\delta}{|\tau|}\ln^{2/5}\frac{\delta}{|\tau|}, \tag{11}$$

$$C_{\text{sing}}(\tau) \sim \frac{1}{R^2\delta^2}\ln^{1/5}\frac{\delta}{|\tau|}. \tag{12}$$

Because of the small factor $(R\delta)^{-2} \sim (\mu^2)^{4/3}\ln^6(1/\mu^2)$, detection of the singularity in the specific heat would be very complicated. The magnetic susceptibility, on the contrary, is anomalously large even at the boundaries of the critical region, since because of exchange interaction the spins are correlated on scales $R \gg 1$. Below the transition point, calculations similar to those of Ref. 18 give the behavior of the spontaneous moment:

$$S \sim \left(\frac{-\tau}{\delta}\right)^{1/2}\ln^{1/10}\frac{\delta}{-\tau}. \tag{13}$$

It is easy to see from (11) and (13) that magnetic fields $h \ll R^2(\tau/\delta)^{3/2} = h(\tau)$ are weak; that is, the thermodynamics in such fields is described by a linear response $\chi(\tau)$. When $h(\tau) \ll h \ll R^{-2}$, the behavior of the system depends primarily on h and not on τ; it is determined as in Ref. 18:

$$S^3 \sim hR^2\ln\frac{1}{(hR^2)^{2/3}}, \tag{14}$$

$$\chi \sim \frac{R^{2/3}}{h^{2/3}}\ln^{1/3}\frac{1}{(hR^2)^{2/3}}. \tag{15}$$

When $h \gg R^2$, DDI is not important, and the system behaves like the XY model in an external field.

4. DISCUSSION OF RESULTS

Formulas (10)–(15) determine the behavior of a two-dimensional planar ferromagnet in the critical region $|T-T_c| \lesssim T_c/\ln^3(1/\mu^2)$. At lower temperatures, the system is in an ordered phase; its properties were determined earlier[17] and depend substantially on DDI. In particular, the spontaneous moment is $S \sim (\mu^2)^{\Delta/(1-2\Delta)}$. In the high-temperature phase, DDI is not important; the system can be described by the 2D XY model.

We shall now explain the applicability of our results to real systems. Experiments on two-dimensional magnets[13] are so far lacking; but there are data on layered compounds of graphite (LCG) with the salts $NiCl_2$ and $CoCl_2$, in which several planes of graphite are located between planes of the salt.[11,12] By introduction of a sufficient number of graphite planes, the direct interplane exchange can be made arbitrarily weak. The dipole interaction falls off slowly and therefore must be taken into account both between spins in a plane and between planes. A calculation of the dipole tensor for a layered system was carried out by Maleev.[16] He showed that in the limit of small momenta, the DDI from other planes completely cancels the linear term in the energy of a fluctuation, so that there remains only a weak interplane antiferromagnetic interaction, decreasing exponentially with the distance between planes. But when the interplane interaction is weak, the spins in different planes are uncorrelated, therefore there is no basis for restricting ourselves to small transverse momenta; and when their magnitude is arbitrary, the cancellation of the term linear in the longitudinal momentum does not occur. Therefore the magnetic properties of LCG may be made as nearly two-dimensional as one pleases, even with allowance for DDI.

In practice, the experimental results cease to depend on the number of graphite layers n as soon as $n \geq 2$ for $NiCl_2$ and as soon as $n \geq 1$ for $CoCl_2$.[12] In the papers of Bragin *et al.*,[12] the magnetization was measured by the NMR method. It was found that the spontaneous moment decreases linearly with temperature at least to $(T_c - T)/T_c \sim 0.03$, reaching 65% of the saturation moment. This is apparently a qualitative confirmation of our results. We remark, however, that such behavior of the moment should also be observed in layered compounds in which the interplane exchange is stronger than the DDI. Our analysis of the critical region is inappropriate in this case; but the simple estimates of the correlation radius, which determine the width of the critical region, remain in force. It is necessary only to take instead of the dipole radius a radius determined by the interplane coupling. In the critical region, the system should then be described by the three-dimensional theory of phase transitions.

Another physical system to which our results are pertinent is a freely floating film of ferroelectric smectic C.[14] The ordering in this system is described by a two-dimensional vector **n** that determines the direction of the projections of the long axis of the molecules on the plane of the smectic layers. The effective Hamiltonian has the form

$$\mathscr{H}=\int d^2x\left\{\frac{1}{2}K_s(\nabla\mathbf{n})^2+\frac{1}{2}K_b[\nabla\mathbf{n}]^2-\mathbf{P}_0\mathbf{E}+\int d^2x'\frac{(\nabla\mathbf{P}_0)(\nabla\mathbf{P}_0')}{2|\mathbf{x}-\mathbf{x}'|}\right\},$$

$$\mathbf{P}_0\perp\mathbf{n},$$

and differs from (1) only by the anisotropy of the elastic moduli $K_s \neq K_b$. In a bulk smectic, K_s and K_b may differ very greatly; but in a 2D system, fluctuations tend to make the situation isotropic.[21] Furthermore, the properties of disclinations (the analog of vortices) do not change qualitatively even when $K_s \neq K_b$.[22] Therefore the phase-transition mechanism that we have investigated is to be expected in this system too.

Note added in proof (November 28, 1978). The author has become aware of an inaccuracy in the conclusion drawn in Ref. 17 regarding the reduction of DDI to isotropic form in consequence of renormalizations. Actually, the quadratic part of the Hamiltonian in the critical region has the form (see R. A. Pelcovits and B. I. Halperin, preprint, Harvard University, 1978)

$$\frac{1}{2}\sum_{\mathbf{k}}\left(k^2R\delta_{\alpha\beta}+\frac{k_\alpha k_\beta}{k}+t_0\delta_{\alpha\beta}\right)\varphi_{\mathbf{k}}{}^\alpha\varphi_{-\mathbf{k}}{}^\beta;$$

a "parquet" situation does not occur, and the fluctuations turn out to be considerably stronger. Therefore the analysis of the critical region ($|\tau| \ll \delta$) is not pertinent to a two-dimensional ferromagnet. All results for $|\tau| \gtrsim \delta$ retain their validity.

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