Perpendicular versus In-Plane Magnetization in a 2D Heisenberg Monolayer at Finite Temperatures

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We show that the 2D Heisenberg monolayer with *perpendicular* ground-state spontaneous magnetization may have a temperature T_R at which the spontaneous magnetization turns into the plane of the monolayer. The reorientation of the magnetization occurs via a phase transition.

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Monolayers of magnetic materials have a broken translational symmetry along the direction perpendicular to the plane defined by the layer. It was originally suggested by Néel[1] that this symmetry breaking could, in principle, remove the degeneracy of the magnetic ground state with respect to the direction of the magnetization: A ground state with spins pointing along the normal or in the plane could have different energies. The possibility of realizing a state with perpendicular magnetization is one of the driving thoughts behind the research on magnetic thin films, aimed at increasing the storage density in magnetic recording devices.[2]

Recently, Gay and Richter gave a rigorous quantitative framework to Néel's original idea.[3] They succeeded in estimating the strength both of the magnetostatic dipole interaction (which favors in-plane magnetization) and of the spin-orbit-coupling–induced magnetic anisotropies (which may favor perpendicular orientation). It turns out[3] that in the monolayer limit of some 3D transition metals the perpendicular anisotropy overcomes the dipole interaction and the ground state is perpendicularly magnetized. In this case a question arises: What happens to the perpendicular configuration when the temperature is raised?

In this Letter we answer this question for the case of a ferromagnetic Heisenberg monolayer. We show that, as a consequence of the strong fluctuations of the 2D Heisenberg model (the very same which lead to the no-long-range-order rule in the Mermin-Wagner theorem[4]), a temperature T_R may exist at which the magnetization of a perpendicularly oriented ground state turns into the plane of the monolayer. In virtue of these fluctuations, in fact, the ground-state anisotropy constant λ and the dipole coupling constant Ω renormalize to temperature-dependent constants $\lambda(T)$ and $\Omega(T)$. We show that $\lambda(T)$ and $\Omega(T)$ renormalize in such a way that the equation $\lambda(T)=\Omega(T)$ has a solution T_R below which the magnetization M is perpendicular $[\lambda(T<T_R)>\Omega(T<T_R)]$ and above which M is in the plane $[\lambda(T>T_R)<\Omega(T>T_R)]$. At T_R a phase transition occurs. The value of T_R is positive (and therefore physical) only if the ground-state values λ and Ω satisfy the inequality $\lambda>\Omega$. Depending on the exact values of λ and Ω, T_R can be smaller or even larger than the Curie temperature T_C of the system (the temperature at which the magnetization vanishes). In the case $T_R>T_C$ no reorientation transition occurs over the whole temperature range where $M(T)\neq 0$. We were driven to search for this transition by experimental observations on the test-case system Fe/Ag(100) obtained by Krebs, Jonker, and Prinz[5] and by Stampanoni *et al.*[6] They reported the disappearance of the ground-state perpendicular remanence at temperatures ≤ 100 K, i.e., well below the Curie temperature of the system. Our argument offers a reasonable explanation of this important experimental finding.

The aim of the present paper is the determination of the anisotropic part of the *free energy*, which is responsible for the existence of easy magnetization directions. We start with the Hamiltonian

$$H=\tfrac{1}{2}\Gamma\int[\nabla\cdot\mathbf{n}(\mathbf{x})]^2d^2x-\lambda\int n_z^2(\mathbf{x})d^2\left(\frac{x}{a}\right)+\frac{1}{4\pi}\Omega\int\int\frac{\mathbf{n}(\mathbf{x})\cdot\mathbf{n}(\mathbf{x}')-3[\nu\cdot\mathbf{n}(\mathbf{x})][\nu\cdot\mathbf{n}(\mathbf{x}')]}{|\mathbf{x}-\mathbf{x}'|^3}d^2\left(\frac{x}{a}\right)d^2\left(\frac{x'}{a}\right),\qquad(1)$$

where $\Gamma=0.5rS^2J$ is the exchange energy (r denotes the number of nearest neighbors, J the exchange constant, and S the spin per atom), λ is the single-ion anisotropy constant favoring perpendicular orientation, and $\Omega=2\pi(g\mu_BS)^2/a^3$ (g denotes the Landé factor, μ_B the Bohr magneton, and a the lattice constant) is the strength of the dipole interaction. ν is a unit vector pointing in the direction of $\mathbf{x}-\mathbf{x}'$ and the integrals in (1) run over the x-y plane defined by the monolayer (z being perpendicular to this plane). $\mathbf{n}(\mathbf{x})$ is a classical vector (field) of unit length with three components. The use of a continuum approximation and of classical vectors to represent the spins is justified by the length scales entering

our problem being much larger than the lattice constant. The net spin measured over such length scales is much larger than 1, so that the quantum nature of the magnetic moment is not resolvable.

When $\lambda=\Omega=0$ we recover the isotropic Heisenberg model, the thermodynamics of which is determined by the strong fluctuations which destroy long-range order as soon as the temperature is raised above $T=0$ K.[4,7] To account for such fluctuations we decompose, using a renormalization procedure devised by Polyakov[7] for the isotropic 2D Heisenberg model, the field $\mathbf{n}(\mathbf{x})$ into a slowly varying component $\mathbf{n}_0(x)$—which, without loss of generality, we chose to be in the z-x plane—and a rapidly varying component $\varphi(x)$ orthogonal to $\mathbf{n}_0$. By slowly [rapidly] varying component we mean that the length scale over which $\mathbf{n}_0(\mathbf{x})$ [$\varphi(\mathbf{x})$] changes is much larger [smaller] than a length L, L being the characteristic length over which the fluctuations of φ "die down." Following Polyakov,[7] the Hamiltonian $H_L(\mathbf{n}_0(\mathbf{x}))$ for the field $\mathbf{n}_0$ can be found by averaging H over the component φ. It can be shown that $H_L(\mathbf{n}_0(\mathbf{x}))$ takes the same form as $H(\mathbf{n}(\mathbf{x}))$, the effect of averaging over φ being to transform the coupling constants from Γ, λ, Ω to

$$\Gamma\left(1-\frac{T}{2\pi\Gamma}\ln\frac{L}{a}\right),\quad \lambda\left(1-3\frac{T}{2\pi\Gamma}\ln\frac{L}{a}\right),\quad \Omega\left(1-2\frac{T}{2\pi\Gamma}\ln\frac{L}{a}\right). \tag{2}$$

From these transformation rules we construct the renormalization-group equations for a k-component field $\mathbf{n}(\mathbf{x})$ ($\zeta=\ln L/a$):

$$\frac{d\Gamma}{d\zeta}=-(k-2)\frac{T}{2\pi},\quad \frac{d\ln\lambda}{d\zeta}=-k\frac{T}{2\pi\Gamma},\quad \frac{d\ln\Omega}{d\zeta}=-(k-1)\frac{T}{2\pi\Gamma}, \tag{3}$$

whose solutions are $\Gamma_L=\Gamma Z$, $\lambda_L=\lambda Z^{k/(k-2)}$, $\Omega_L=\Omega Z^{(k-1)/(k-2)}$, where $Z=1-T\zeta/2\pi\Gamma$. For the isotropic Heisenberg model the characteristic length L over which the fluctuations of φ die down is effectively bounded only by the physical lateral dimensions of the system (and ideally grows to infinity, in the thermodynamic limit). This leads to Γ_L becoming arbitrarily small as L grows and thus to a vanishing Curie temperature. For the model Hamiltonian (1), instead, L is bounded by $L_0=(\Gamma/\lambda)^{1/2}$, after which the fluctuations have died out allowing "conventional" long-range order (corresponding to the two-component field $\mathbf{n}_0$) to set in up to a Curie temperature given by the Kosterlitz-Thouless value $T_C=2\pi\Gamma/(4+\zeta_0)$.[8] The anisotropic part of the Landau free energy per atom is obtained as usual by substituting in $H_{L_0}(\mathbf{n}_0(\mathbf{x}))$ the field $\mathbf{n}_0(\mathbf{x})$ with its mean value $\langle\mathbf{n}_0(\mathbf{x})\rangle=(\cos\vartheta,\sin\vartheta)$

$$F(\vartheta)=-\lambda_{L_0}\cos^2\vartheta-\frac{3\Omega_{L_0}}{4\pi}\int d^2\left(\frac{x}{a}\right)\frac{(\nu\cdot\langle\mathbf{n}_0\rangle)^2}{|\mathbf{x}/a|^3}$$
$$=-\lambda_{L_0}\cos^2\vartheta-\frac{3\Omega_{L_0}}{4\pi}\sin^2\vartheta\int d\omega\frac{r}{a}d\left(\frac{r}{a}\right)\frac{\cos^2\omega}{(r/a)^3}, \tag{4}$$

where ϑ is the angle between the magnetization and z. Equation (4) contains a diverging integral, resulting from the continuum approximation unphysically including the origin as an integration limit. This unphysical divergence can be avoided by introducing a cutoff of the order of the lattice constant. The value of the integral depends on the choice of the cutoff length, so that the integral is best estimated by performing the (convergent) corresponding sum over discrete lattice sites, which gives $8\pi/6$. Finally, after some trigonometric manipulations, we obtain

$$F(\vartheta)=\tfrac{1}{2}(\Omega_{L_0}-\lambda_{L_0})\cos2\vartheta. \tag{5}$$

Since Z_0 is smaller than 1, the equation $\lambda_{L_0}(T)=\Omega_{L_0}(T)$ will have a solution $T_R=\lambda^{-1}(\lambda-\Omega)2\pi\Gamma/\zeta_0$ with a positive (and therefore physical) value of T_R for a perpendicularly magnetized ground state ($\lambda>\Omega$). The free energy $F(\vartheta)$ below, at, and above T_R is given graphically in Fig. 1. As expected, below T_R, $F(\vartheta)$ has a stable minimum for $\vartheta=0$; i.e., the system is perpendicularly magnetized. Above T_R the minimum is at $\vartheta=\pi/2$; i.e., the magnetization is in the plane. At T_R the free energy has a singularity, which in this simple picture is manifested by all angles ϑ between 0 and $\pi/2$ being equally

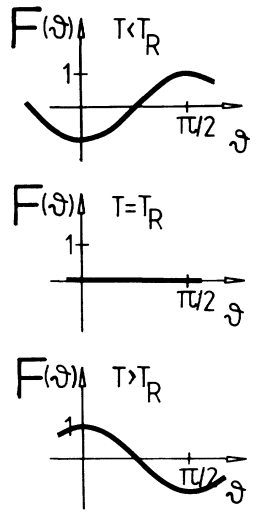

FIG. 1. Free energy $F(\vartheta)$ [in units of $\frac{1}{2}(\lambda_{L_0}-\Omega_{L_0})$] below ($T<T_R$), at ($T=T_R$), and above ($T>T_R$) the reorientation temperature T_R. ϑ is the angle between M and the z axis perpendicular to the monolayer plane.

possible. This degeneracy is probably lifted by the higher-order terms in the anisotropy energy. In this case, fluctuations of the direction of $\mathbf{n}_0$ in the vicinity of T_R [neglected in the Landau approach, Eq. (4)] should be taken into account for a precise treatment of the phase transition, which is beyond the scope of this paper.

The size of T_R can be best appreciated by considering that $T_R/T_C=\lambda^{-1}(\lambda-\Omega)(4+\zeta_0)/\zeta_0$. We obtain the physically plausible result, in line with experimental observations on the test case of Fe films on Ag(100), that if λ is sufficiently close to Ω then T_R is a fraction of the Curie temperature. For instance, for the values of $\lambda=0.38$ meV, $\Omega=0.3$ meV calculated for the Fe monolayer in the original work of Gay and Richter[3] and a bulk value of $\Gamma=40$ meV,[9] we obtain $T_R/T_C\cong 50\%$. On the other hand, T_R/T_C can be larger than 1 if λ is sufficiently large; i.e., in this case the system remains perpendicularly magnetized up to its Curie temperature. This seems to occur in a similar system—Fe/Cu(100)—where some authors[10] reported the persistence of a perpendicular magnetization up to room temperature, which is approximately the Curie temperature of this system.[11] In any case, T_R is not of the order of $\lambda-\Omega$, which is typically in the 1-K range, as one might be induced to think. After submitting this paper we became aware of a theoretical analysis of the same problem by Jensen and Bennemann.[12] Their idea of the entropy of disorder leading to a reorientation of the magnetization coincides with the spirit of our paper. However, by neglecting the fluctuations—which are crucial in two dimensions—they obtain a much lower transition temperature than the one obtained by our renormalization-group approach.

The above results can be easily generalized to the case where a magnetic field $h=g\mu_B H$ is applied, taking into account that h renormalizes according to $h_{L_0}=hZ_0$. For the free energies with field applied parallel or perpendicular to the plane we obtain

$$F_{\parallel}(\vartheta)=\tfrac{1}{2}(\Omega_{L_0}-\lambda_{L_0})\cos 2\vartheta-h_{L_0}\sin\vartheta\,,$$
$$F_{\perp}(\vartheta)=\tfrac{1}{2}(\Omega_{L_0}-\lambda_{L_0})\cos 2\vartheta-h_{L_0}\cos\vartheta\,. \tag{6}$$

The main result of an applied field parallel to the film is that the equilibrium angle ϑ_e in the low-temperature phase $T<T_R$ is not exactly 0 but assumes the field- and temperature-dependent value given by $\sin\vartheta_e=h_{L_0}/2(\lambda_{L_0}-\Omega_{L_0})$. In particular, when $h_{L_0}=2(\lambda_{L_0}-\Omega_{L_0})$, i.e., for sufficiently high fields, a spin-flop phase transition proceeds, with the perpendicular component going to zero. Similarly, a perpendicular applied field modifies the angle assumed by the magnetization after the transition to a value given by $\cos\vartheta_e=\frac{1}{2}h_{L_0}/(\Omega_{L_0}-\lambda_{L_0})$. When $h_{L_0}=2(\Omega_{L_0}-\lambda_{L_0})$ the system undergoes a phase transition characterized by the parallel component of M vanishing. It has been suggested that this transition should be of the Berezhinskii-Kosterlitz-Thouless type.[13]

In conclusion, we predict a reorientation phase transition to occur in the 2D Heisenberg monolayer with perpendicular magnetization, as a consequence of the strong fluctuations of this model. This reorientation, which is essentially the result of the entropy of disorder prevailing over the energy part of the free energy, has apparently already been observed experimentally in Fe thin films on Ag(100). The generality of our argument, however, should intrigue experimentalists to search for such a transition in similar systems and study more closely its features.

Nota bene: The existence of a reversible transition between perpendicular and in-plane magnetization in ultrathin films has been experimentally confirmed by Pappas, Kämper, and Hopster[14] on Fe films on Cu(100).

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