Magnetism of two-dimensional magnets in the presence of random fields

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In low-dimensional magnets, thermal agitation and spatial disorders generate strong spin fluctuations that suppress the long-range magnetic ordering. We develop an analytical equation for the equilibrium magnetization of two-dimensional magnets at finite temperatures in the presence of the random magnetic field. We find that the random field induces a first-order phase transition in addition to the reduced Curie temperature. The first-order phase transition persists even in the presence of the moderate external magnetic field.

In this paper, we establish an analytical theory of the temperature dependence of the magnetization in the presence of the random field. We show that, by using the self-consistent random phase approximation, the analytical form of the magnetization is simple enough and yet it has properly included the 2D spin fluctuation. We find that the random field not only reduces the ordering temperature through the reduced anisotropic gap, it also makes the phase transition from the second order to the first order. Even with a moderate external magnetic field, the first order transition persists.

We start with the generic anisotropic Heisenberg Hamiltonian on a 2D square lattice,

$$\hat{H} = -J_{ex} \sum_{<i,j>} \hat{S}_i \hat{S}_j - A \sum_{<i,j>} \hat{S}_i^z \hat{S}_j^z - \sum_i (H + h_i) \hat{S}_i^z \tag{1}$$

Where $\hat{S}_i$ and $\hat{S}_i^z$ are respectively the spin and the z-component (taken as perpendicular to the two-dimensional plane) of the spin operators at lattice site $\mathbf{R}_i$, $J_{ex}$ is the isotropic exchange integral, $A$ is the anisotropic exchange integral, $<i,j>$ indicates the sum over nearest neighbors, and $H$ and $h_i$ are the external and the random magnetic field in the z-direction. The random field is assumed uncorrelated, i.e.,

$$<h_i>_c = 0; \quad <h_i h_j>_c = \delta_{ij}\gamma^2 \tag{2}$$

where $<>_{c}$ represents the configuration average over the distribution of the random field. The physical origins of the random fields may come from the spin-orbit coupling at the imperfect surface or interface in which the local electronic potential is no longer periodic. For simplicity, we assume the strength of the random field, $\gamma$, is independent of the spin states and temperature. As the model Hamiltonian, Eq. (1), has no exact solution even without the random field, one usually relies on numerical methods such as quantum Monte Carlo simulation to determine the equilibrium magnetization and critical phenomena. As the analytical formulation for the magnetization is extremely useful for studying various spin transport properties, we will use a self-consistent random phase approximation (RPA) to determine the temperature de-
dependence of the magnetization. Although the RPA is an approximate method, the physics of the spin fluctuation from the low energy excitations has been taken into account.

To use the RPA, we first define the retarded Green’s function of spin operators,

\[ G_{ij}^R(t) = \langle \langle \hat{S}_i^+(t)\hat{S}_j^-(t) \rangle \rangle = i\theta(t) - \langle \langle \hat{S}_i^+(t)\hat{S}_j^-(t) \rangle \rangle \]

\[ \equiv i\epsilon_S^+ - \langle \langle \hat{S}_i^+(t)\hat{S}_j^-(t) \rangle \rangle \]

(3)

where \( \hat{S}_i^\pm = \hat{S}_i \pm \hat{S}_\mp \) is lowering and raising spin operator, \( \Theta(t) \) is the Heaviside step function and \( \langle \langle \cdot \rangle \rangle \) denotes the thermal average. The equation of motion for the above Green function is then

\[ i\frac{dG_{ij}^R(t)}{dt} = \langle \langle \hat{S}_i^+, \hat{S}_j^- \rangle \rangle = \delta_{ij} + \langle \langle \hat{S}_i^+(t), \hat{H}, \hat{S}_j^- \rangle \rangle \]

When we substitute Eq. (1) into the commutator \( [\hat{S}_i^+(t), \hat{H}] \), the result involves the terms involving the product of the three spin operators, e.g., \( \hat{S}_i^+\hat{S}_j^+\hat{S}_l^- \). To obtain a closed form for the Green’s function, we use the RPA in which the longitudinal spin \( \hat{S}_i^z \) and the transverse spin fluctuation \( \hat{S}_i^+\hat{S}_j^- \) at the different sites \( i \neq j \), are uncorrelated, i.e.,

\[ \langle \langle \hat{S}_i^+\hat{S}_j^+, \hat{S}_l^- \rangle \rangle = \langle \langle \hat{S}_i^+ \rangle \rangle \langle \langle \hat{S}_j^- \rangle \rangle \]

(4)

Defining the site-independent magnetization \( M(T) \equiv \langle \langle \hat{S}_i^z \rangle \rangle \), and making the Fourier transformation in space and time, \( G_{kk'}^R(E) = (2\pi)^{-1}\sum_{ij} e^{ik\cdot R_i} e^{i\epsilon_k R_j} \int dt e^{-iEt} G_{ij}^R(t) \), we find

\[ EG_{kk'}^R(E) = 2M\delta_{kk'} + E_k^0 G_{kk'}^R(E) - \sum_q h_q g_{kk'}^R(E) \]

(5)

where \( E_k^0 = 2\varepsilon M[J(1 - \gamma_k) + A] + H \) is the energy spectrum without the random field, \( z \) is the number of the nearest neighbors and \( \gamma_k = 1 \frac{1}{z} \sum e^{ik\cdot R} \) and the summation is over the nearest-neighbor sites. The above retarded Green’s function can also be written in a compact form,

\[ G_{kk'}^R(E) = \frac{2M\delta_{kk'}}{E - E_k^0 - \Sigma(E, k)} \]

(6)

where \( \Sigma(E, k) \) is the self-energy of the random field which can be expressed in terms of the series summation over the orders of the random field. If we keep the random field up to the second order, the self-energy is

\[ \Sigma(E, k) = \gamma^2 \int g(\epsilon) de \]

(7)

where we have used \( \hbar k\epsilon_k = \gamma^2 \) and \( g(\epsilon) \) is the density of states. To further simplify the analytical expression, we approximate the unperturbed dispersion by \( E_k^0 = M(8A + 2Jk^2a_0^2) + H \) for the square lattice such that the density of states is a constant for the energy within the magnon band, i.e., \( g(\epsilon) = (8\pi JM)^{-1} \) for \( \Delta_0 < \epsilon < \Delta_0 + W_0 \) where the energy gap is \( \Delta_0 = 8MA + H \) and the bandwidth \( W_0 = 8JM \). The energy dispersion in the presence of the random field is given by the poles of Green’s function, Eq. (7). By explicitly integrating the constant density of state in Eq. (8), we obtain the energy dispersion with the random field,

\[ E_k = E_k^0 + \frac{\gamma^2}{8\pi JM} \ln \left| \frac{E_k - \Delta_0}{\Delta_0 + W_0 - E_k} \right| \]

(8)

The above equation is an implicit equation that determines \( E_k \) for a given magnetization \( M \). However, \( M \) is unknown a priori, and must be determined self-consistently. Recall the spin operator identity, \( \hat{S}_i^z = S(S + 1) - \langle \langle \hat{S}_i^+ \rangle \rangle^2 \). For spin-1/2, the identity becomes \( \hat{S}_i^z = 1/2 - \langle \langle \hat{S}_i^+ \rangle \rangle \) and thus, \( M = 1/2 - \langle \langle \hat{S}_i^+ \rangle \rangle \). By taking the thermal averaging of the above identity, we have

\[ M = \frac{1}{2} - \sum_{kk'} \int dE \frac{2\Im(G_{kk'}^R(E + i0^+))}{2\pi} \frac{e^{\beta E} - 1}{e^{\beta E} + 1} \]

(9)

By replacing

\[ \Im G_{kk'}^R(E + i0^+) = 2\pi M\delta \left( E - E_k^0 - \text{Re}\Sigma(E, k) \right) \]

\[ = 2\pi M\delta(E - E_k) Z_k \]

where \( Z_k = (1 - \frac{\partial E_k}{\partial k})^{-1} \) into Eq. (8), we have

\[ M = \frac{1}{2} - \int \frac{d^2k}{(2\pi)^2} \frac{2MZ_k}{e^{\beta E_k} - 1} \]

(10)

Since we have used quadratic dispersion in the energy \( E_k^0 \propto k^2 \), we can change the integration over \( d^2k \) to \( dE_k \), i.e., replacing \( d^2k = 2\pi k dk \) by \( \frac{2\pi}{\Delta_0} dE_k = \frac{2\pi}{\Delta_0} (1 - \frac{\partial E_k}{\partial k}) dE_k \) in Eq. (9), we find

\[ M = \frac{1}{2} - \frac{1}{4\pi J} \left( \frac{1}{\beta} \ln \left| \frac{e^{\beta(\Delta_0 + W_0)} - 1}{e^{\beta\Delta} - 1} \right| - W_0 \right) \]

(11)

where the effective energy gap \( \Delta = \Delta_0 + (E_k - E_k^0)|_{k=0} \) and we have set the bandwidth \( W_0 \) unchanged since we assume the density of states remains unperturbed by the disorder. By using Eq. (7), we may explicitly write the effective gap,

\[ \Delta = \Delta_0 - \frac{\gamma^2}{8\pi JM} \ln \left| 1 + \frac{8\pi JM}{\Delta_0 - \Delta} \right| \]

Equations (10) and (11) are our main results. The role of the random field is the reduction of the anisotropic gap from \( \Delta_0 \) to \( \Delta \). The limiting values of the gap reduction can be readily obtained from Eq. (11). At the temperature well below the Curie temperature, \( \gamma \ll 8\pi JM \), the above gap reduction \( \Delta_0 - \Delta \) is negligible. As temperature increases, \( M \) decreases and thus \( \Delta_0 - \Delta \) increases.
FIG. 1. The complete solutions of $M$ as a function of temperature are calculated from Eq. (10) and (11). The section with the blue line is the equilibrium ferromagnetic state while the other two sections are not stable states. The section with the yellow color has higher free energy compared to the blue line and the section with the orange color has a negative effective energy gap. We have used $J = 1$, $A = 0.2$, $H = 0$ and $\gamma = 0.1$.

When $M$ becomes very small such that $8\pi J M \ll \gamma$, the gap reduction reaches its maximum value of $\gamma$. In the absence of the magnetic field, $\Delta_0 = 8MA$ decreases with temperature while the gap correction from the random field increases with the temperature. Thus, at a certain temperature, the effective gap becomes too small to support long-range ordering since the spin fluctuations at finite temperature destabilizes the magnetization and long range order is destroyed. More quantitatively, we shall numerically solve Eq. (10), along with Eq. (11), to determine the temperature dependence of the magnetization.

We show general features of the mathematical solution of Eq. (10) in Fig. 1. For a given anisotropy constant $A$ and a random field strength $\gamma$, there are three solutions for the magnetization at the low temperature. The upper curve represents the physically meaningful solution. The bottom curve is unphysical since it represents the case where the effective gap $\Delta$ becomes negative. Clearly, the ground state is no longer in the $z$-direction when the gap is negative, and thus the magnon excitation along the $z$-axis becomes invalid. The middle curve in Fig. 1 is also the solution of Eq. (10), but the free energy is higher than the upper curve at the same temperature. Therefore, we will take the upper curve as the physical solution of the Eq. (10) and we will only show the upper curve in the following numerical results.

We show the magnetization curves as a function of the strength of the disorder in Fig. 2. Without disorders, the magnetization undergoes the second-order phase transition at the Curie temperature where the anisotropy gap can no longer stabilize the magnetization against thermal fluctuations. The magnetization approaches zero at the critical temperature of the second-order phase transition, as shown in the blue line of Fig. 2. Two distinct features are seen as we increase the strength of the disorders. First, the reduction of the transition temperature scales
as the strength of the random field; this is expected since the effective gap, $\Delta$ of Eq. (11), decreases as the random field increases. At low temperature, however, the effect of the random field is negligible since the gap $\Delta$ is not significantly different from $\Delta_0$. The second feature is more interesting: the phase transition becomes first order with the random field. When the temperature reaches a critical value, Eq. (10) does not have a solution anymore, indicating that the ferromagnetic phase we have assumed in deriving Eq. (10) does not exist, i.e., the phase transition occurs at a finite value of the magnetization $M_c$ whose magnitude scales with the strength of the random field, as shown in the insert of the Fig.2b. Since there are no solutions of Eq. (10) for $T > T_c$, the magnetization is no longer uniform. Instead, the magnetization breaks into domains by the random field with the magnetization of each domain fluctuating at high temperature, known as superparamagnetic (SPM) states.

We now discuss the effects of an external magnetic field on magnetization. Any external magnetic field breaks the time-reversal symmetry and thus the second-order phase transition which characterizes the transition between the time-reversal symmetry-broken and symmetry-conserving does not exist. With the random field, however, we find the first-order phase transition persists. If the magnetic field is smaller or comparable to the strength of the random field, the solution of Eq. (10) shows a similar first-order phase transition at a critical temperature. The explanation is as follows. The random field leads an effective anisotropy gap as small as $\Delta = \Delta_0 - \gamma = 8AM + H - \gamma$ when $M$ is small (or temperature is high). If $H$ is smaller than $\gamma$, the effective gap would be small or negative at the high temperature which leads to the collapse of the magnetization due to thermal fluctuation at a critical value of the temperature. In Fig. 3, we show the magnetization at several different external fields. When the external field is much larger than $\gamma$, the magnetization is essentially identical to that without the random field, i.e., there is no phase transition.

![Diagram](image)

FIG. 3. (a) Magnetization as a function of temperature for several different external magnetic fields. The dotted lines mark the critical points where the solutions end and the first-order phase transition occurs. $A = 0.2$ and $\gamma = 0.1$. (b) and (c) The critical magnetization and the critical temperature as functions of the external magnetic field for several different random fields. $A = 0.2$.

Taken together, we construct a temperature-magnetic field phase diagram in the presence of the random field, shown in Fig. 4. Since there is no second-order phase transition in the presence of the magnetic field, we denote the region as the (Ferro-Para)magnetic in which the solution of a uniform magnetization exists. When the magnetic field is small, there is a region in which there is no solution with a uniform magnetization. While our theory cannot address the details of this non-uniform SPM, we postulate that the magnetization breaks into domains whose local directions are dictated by the net random field within the domains; this similar picture has been described by Imry and Ma [24] from the viewpoint of thermal dynamics, and further confirmed by Fisher and co-workers [25]. When the magnetic field is large, the magnetic state is a single phase, however, we can artificially separate the regions with the ferromagnetic state FM and the paramagnetic state PM by defining the boundary line as the maxima of the magnetic susceptibility.

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FIG. 4. The phase diagram of a two-dimensional magnetic system with a random field. The solid black line represents the first-order phase transition from the (Ferro/Para)magnetic phase to the Super-paramagnetic (SPM) phase. The black dotted line represents the states of the largest susceptibility for a given temperature. One may define this dotted line as a “boundary” between ferromagnetic and paramagnetic regions.

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[2] Gong, C. et al., Nature 546, 265–269 (2017).