



Research article

Prediction of a first-order phase transition in two-dimensional ferromagnets in the presence of random fields

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ARTICLE INFO

Keywords:

Two dimensions

Ferromagnet

First-order phase transition

Random fields

Random phase approximation

ABSTRACT

Random magnetic fields suppress the long-range magnetic ordering through the formation of the magnetic domains. The size of the domains is determined by the competition among the exchange interaction, the magnetic anisotropy, and the strength of the random field. Here we theoretically investigate the temperature dependence of the magnetization of the two-dimensional domains for the anisotropic Heisenberg model with a random magnetic field. We find that magnetization of the domains displays a first-order phase transition in which the magnetization is discontinuous at a critical temperature. Moreover, the first-order transition persists even in the presence of an external magnetic field whose magnitude is smaller than the strength of the disorder. The above unusual first-order phase transition can be experimentally tested with doped two-dimensional magnets.

1. Introduction

In the last several years, research interests in low dimensional magnetism are getting greatly boosted by experimental identifications of many 2D magnetic materials, particularly, those of 2D van der Waals (vdW) magnetic semiconductors [1–8]. Similar to the well-studied graphenes, the thickness of 2D vdW semiconductors can be precisely controlled down to one monolayer, and thus it is now possible that theoretical predictions on 2D magnetism can be quantitatively compared with the experiments. Several magnetic and transport phenomena have already been reported experimentally, like the giant magnetoresistance [9], spin–orbit torques [10–13], spin Seebeck, and Nernst effect [14–16] including the discovery of large anomalous Nernst effect in Fe_3GeTe_2 by C.L. Chien's group [17]. These experimental advances raise an interesting perspective on 2D magnetic materials for spintronics applications.

One of the most fundamental properties of 2D magnetism is that the long-range ferromagnetic ordering should not exist in principle [18]. Imry and Ma [19] have shown that an arbitrary small random field or disorder would destroy the ferromagnetic phase since the random field favors the break-up of the uniform magnetization into domains as a result of the competition among exchange interactions, magnetic anisotropy, and the random field. Theoretically, such a conclusion is universally valid for Heisenberg and Ising models with or without long-range dipolar interaction [20]. Since the random field is always present in real materials, the experimentally observed 2D ferromagnetism must

not be a true ferromagnetic phase with a uniform magnetization. Instead, one may reasonably argue that the size of the domains induced by the random field is large for weak disorders and these domains can be swiped away by a small magnetic field. Indeed, a theoretical analysis by Malozemoff [21] has estimated the domain size in various model Hamiltonians and has established qualitative relations between the random field and domain size.

In this paper, we investigate how the magnetization of the ferromagnetic domains in 2D films varies with the temperature and the magnetic field. We assume, *a priori*, that these domains are much larger than the domain wall thickness such that we only need to calculate the magnetization within an individual domain. By using the self-consistent random phase approximation, we establish an analytical theory of the temperature dependence of magnetization as a function of the strength of the random field. We have found a novel first-order phase transition that has not been anticipated previously: the magnetization has a discontinuous drop when the temperature increases to a critical value. Even with a moderate external magnetic field, the first-order transition persists. The paper is organized as follows. In Section 2, we define our model Hamiltonian, followed by deriving the analytical expression of the equilibrium magnetization by using the self-consistent random phase approximation (RPA) in Section 3. We discuss the results in Section 4.

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<https://doi.org/10.1016/j.jmmm.2022.169993>

Received 11 July 2022; Received in revised form 20 September 2022; Accepted 21 September 2022

Available online 27 September 2022

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2. Model

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

$$\hat{H} = -J \sum_{\langle i,j \rangle} \hat{S}_i \cdot \hat{S}_j - A \sum_{\langle i,j \rangle} \hat{S}_i^z \cdot \hat{S}_j^z - \sum_i (H + h_i) \hat{S}_i^z \quad (1)$$

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 is the isotropic exchange integral, A is the anisotropic exchange integral, $\langle i,j \rangle$ indicates the sum over nearest neighbors, and H and h_i are the external and the random magnetic field in the z -direction. We take the random field to be an uncorrelated white-noise,

$$\langle h_i \rangle_c = 0; \quad \langle h_i h_j \rangle_c = \delta_{ij} \gamma^2 \quad (2)$$

where $\langle \rangle_c$ represents the configuration average over the distribution of the random field. Without the random field, the above simple anisotropic Heisenberg model has been applied to a number of 2D magnets such as CrI_3 with the exchange constant J and the anisotropy A obtained by the first-principle calculation [22,23]. The random field we introduced here may come from the spin-orbit coupling at the imperfect surface or interface in which the local electronic potential is no longer periodic. One may identify two sources of the random field. First, as 2D magnetic films are usually grown on a substrate or by exfoliation, lattice mismatch with the substrate leads to roughness, which causes the breakdown of the perfect periodicity for the 2D film. The second case is due to the presence of impurities or vacancies in which the impurities could be introduced intentionally or unintentionally. The detailed correlation between the random field and the material structure is complex and we will follow the conventional choice of the random field [19–21] to be uncorrelated at different sites with its strength γ^2 by a single constant, independent of energy 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 extend the self-consistent random phase approximation (RPA) [24–27] to include the random field for the calculation of the temperature dependence of the magnetization. Although the RPA is an approximate method, the physics of the spin fluctuation from the low energy excitations has been explicitly taken into account. The RPA becomes a poor approximation when the magnetization is small, i.e., near the second-order phase transition temperature. In the present case, we find a first-order phase transition occurs where the magnetization remains finite and thus the RPA would be an excellent approximation for the temperature below the first-order phase transition temperature to be determined later in the paper.

3. Equilibrium magnetization in the presence of random fields

Following the conventional method of RPA [24–27], we first define the retarded Green's function of spin operators,

$$G_{ij}(t) = \langle\langle \hat{S}_i^+(t) : \hat{S}_j^- \rangle\rangle \equiv -i\Theta(t) \langle\langle \hat{S}_i^+(t), \hat{S}_j^- \rangle\rangle \quad (3)$$

where $\hat{S}^\pm = \hat{S}_x \pm i\hat{S}_y$ is lowering and raising spin operator, $\Theta(t)$ is the Heaviside step function, $\langle\langle \dots \rangle\rangle$ denotes the thermal average and $\langle\langle : \dots \rangle\rangle$ is Zubarev notation [28]. The equation of motion for the above Green function in the frequency space is then

$$E G_{ij}(E) = \langle[\hat{S}_i^+, \hat{S}_j^-] \delta_{ij} + \langle\langle [\hat{S}_i^+, \hat{H}] : \hat{S}_j^- \rangle\rangle \quad (4)$$

When we substitute Eq. (1) into the commutator $[\hat{S}_i^+, \hat{H}]$, the result contains the terms involving the product of the three spin operators,

e.g., $\hat{S}_i^z \hat{S}_i^+ \hat{S}_j^-$. To obtain a closed form for the Green's function, we use the decoupling scheme known as 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 [24–26], i.e.,

$$\langle\langle \hat{S}_i^z \hat{S}_i^+, \hat{S}_j^- \rangle\rangle \approx \langle\langle \hat{S}_i^z \rangle\rangle \langle\langle \hat{S}_i^+, \hat{S}_j^- \rangle\rangle. \quad (5)$$

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, $G_{\mathbf{k}\mathbf{k}'}(E) = (2\pi)^{-1} \sum_{ij} e^{i\mathbf{k}\cdot\mathbf{R}_i + i\mathbf{k}'\cdot\mathbf{R}_j} \int dt e^{-iEt} G_{ij}(t)$, we find

$$(E - E_k^{(0)}) G_{\mathbf{k}\mathbf{k}'}(E) + \sum_q h_{\mathbf{k}-\mathbf{q}} G_{\mathbf{q}\mathbf{k}'}(E) = 2M \delta_{\mathbf{k}\mathbf{k}'} \quad (6)$$

where $E_k^{(0)} = 2zM[J(1 - \gamma_k) + A] + H$ is the energy spectrum without the random field, z is the number of the nearest neighbors and $\gamma_k = \frac{1}{z} \sum e^{i\mathbf{k}\cdot\mathbf{R}}$ and the summation is over the nearest-neighbor sites. We note that we will neglect the finite size effect of the domain such that the lower bound of the wave-number k is set to zero.

The above retarded Green function may be expanded in terms of the order of the random fields. The zeroth order is $G_{\mathbf{k}\mathbf{k}'}^{(0)} = 2M \delta_{\mathbf{k}\mathbf{k}'} / (E - E_k^{(0)})$, the first order is $G_{\mathbf{k}\mathbf{k}'}^{(1)} = -2M h_{\mathbf{k}-\mathbf{k}'} / (E - E_k^{(0)})(E - E_{\mathbf{k}'}^{(0)})$, and the second order is,

$$G_{\mathbf{k}\mathbf{k}'}^{(2)} = -\frac{2M}{(E - E_k^{(0)})(E - E_{\mathbf{k}'}^{(0)})} \sum_q \frac{h_{\mathbf{q}-\mathbf{k}'} h_{\mathbf{k}-\mathbf{q}}}{E - E_q^{(0)}}. \quad (7)$$

By using the assumption of white-noise random fields, $\langle h_{\mathbf{q}-\mathbf{k}'} h_{\mathbf{k}-\mathbf{q}} \rangle = \gamma^2 \delta_{\mathbf{k}\mathbf{k}'}$, the Green's function, up to the second order, $G = G^{(0)} + G^{(2)}$ (the first order is averaged to zero), is

$$G_{\mathbf{k}\mathbf{k}'}(E) = \frac{2M \delta_{\mathbf{k}\mathbf{k}'}}{E - E_k^{(0)} - \Sigma(E, \mathbf{k})} \quad (8)$$

where we have defined self-energy

$$\Sigma(E, k) = \gamma^2 \sum_q \frac{1}{E - E_q} = \gamma^2 \int \frac{g(\epsilon) d\epsilon}{E - \epsilon} \quad (9)$$

and we have replaced the summation of $\mathbf{q}$ with the integration over the energy in the last identity, with $g(\epsilon)$ being the density of states. To further simplify the analytical expression, we consider a simple dispersion $E_k^{(0)} = zM(2A + Jk^2 a_0^2/2) + H$ such that the density of states is a constant for the energy within the magnon band, i.e., $g(\epsilon) = (2z\pi JM)^{-1}$ for $\Delta_0 < \epsilon < \Delta_0 + W_0$ where the energy gap is $\Delta_0 = 2zM A + H$ and the bandwidth $W_0 = 2\pi zJM$. The energy dispersion in the presence of the random field is given by the poles of Green's function, Eq. (8). By explicitly integrating the constant density of state in Eq. (9), we obtain the energy dispersion with the random field,

$$E_k = E_k^{(0)} + \frac{\gamma^2}{2\pi zJM} \ln \left| \frac{E_k - \Delta_0}{\Delta_0 + W_0 - E_k} \right| \quad (10)$$

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, $\hat{S}_i^z = S(S+1) - (\hat{S}_i^z)^2 - \hat{S}_i^- \hat{S}_i^+$. For spin-1/2, the identity becomes, $\hat{S}_i^z = 1/2 - \hat{S}_i^- \hat{S}_i^+$ and thus, $M = 1/2 - \langle\langle \hat{S}_i^- \hat{S}_i^+ \rangle\rangle$. By taking the thermal averaging of the above identity, we have

$$M = \frac{1}{2} - \sum_{\mathbf{k}\mathbf{k}'} \int \frac{dE}{2\pi} \frac{2\text{Im}(G_{\mathbf{k}\mathbf{k}'}(E + i0^+))}{e^{\beta E} - 1} \quad (11)$$

By substituting

$$\begin{aligned} \text{Im} G_{\mathbf{k}\mathbf{k}'}(E + i0^+) &= 2\pi M \delta(E - E_k^{(0)} - \text{Re} \Sigma(E, k)) \\ &= 2\pi M \delta(E - E_k) Z_k \end{aligned}$$

into Eq. (11), we have

$$M = \frac{1}{2} - \int \frac{d^2 k}{(2\pi)^2} \frac{2M Z_k}{e^{\beta E_k} - 1} \quad (12)$$

where $Z_k = (1 - \frac{\partial \Sigma}{\partial E_k})^{-1}$. Comparing the above equation with the spin wave approximation shown in [22,23], our self-consistent RPA

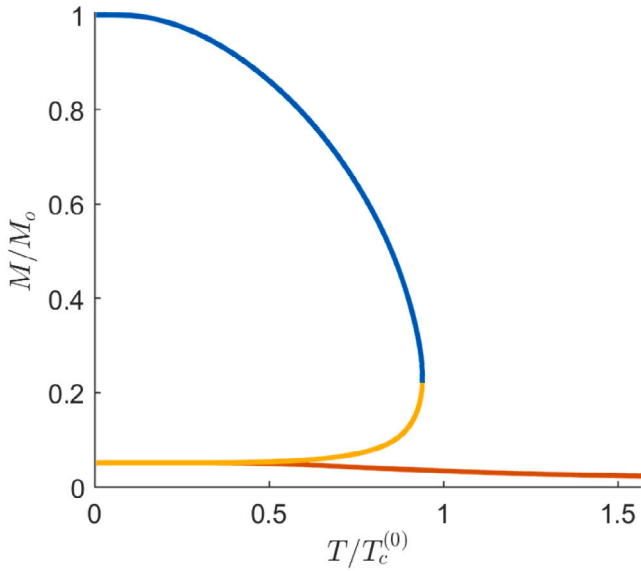


Fig. 1. The complete solutions of M as a function of temperature are calculated from Eqs. (13) and (14). The section with the blue line is the equilibrium ferromagnetic state while the other two sections are not stable states. The section with the yellow color has higher free energy compared to the blue line and the section with the orange color has a negative effective energy gap. We have used $J = 1$, $z = 4$, $A = 0.2$, $H = 0$ and $\gamma = 0.1$.

approach contains the factor of MZ_k in the second term; they represent the renormalization Z_k to the self-energy of the magnon and the reduced magnon angular momentum by M . Without this correction, the magnetization $M = 0$ does not satisfy Eq. (12) at any temperature without the disorder.

The integration in Eq. (12) can be readily done if we use the quadratic dispersion in the energy $E_k^{(0)} \propto k^2$. By changing the integration over d^2k to dE_k , i.e., substituting $d^2k = 2\pi k dk = \frac{2\pi}{zMJ} dE_k^{(0)} = \frac{2\pi}{zMJ} (1 - \frac{\partial \Sigma}{\partial E_k}) dE_k = \frac{2\pi}{zMJ} Z_k^{-1} dE_k$ we find

$$M = \frac{1}{2} - \frac{1}{\pi z J} \left(\frac{1}{\beta} \ln \left| \frac{e^{\beta(\Delta + W_0)} - 1}{e^{\beta\Delta} - 1} \right| - W_0 \right) \quad (13)$$

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 number of states remains unperturbed by the disorder. By using Eq. (10), we may explicitly write the effective gap,

$$\Delta = \Delta_0 - \frac{\gamma^2}{2\pi z J M} \ln \left| 1 + \frac{2\pi z J M}{\Delta_0 - \Delta} \right|. \quad (14)$$

The role of the random field is the reduction of the anisotropic gap from Δ_0 to Δ . At the temperature well below the Curie temperature, $\gamma \ll 2\pi z J M$, the above gap reduction $\Delta_0 - \Delta$ is negligible. As temperature increases, M decreases and thus $\Delta_0 - \Delta$ increases. When M becomes very small such that $2\pi z J M \ll \gamma$, the gap reduction reaches its maximum value of γ . In the absence of the magnetic field, $\Delta_0 = 2zMA$ 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. (13), along with Eq. (14), to determine the temperature dependence of the magnetization.

4. Results and discussions

We show general features of the mathematical solution of Eq. (13) in Fig. 1. For a given anisotropy constant A and a random field strength γ ,

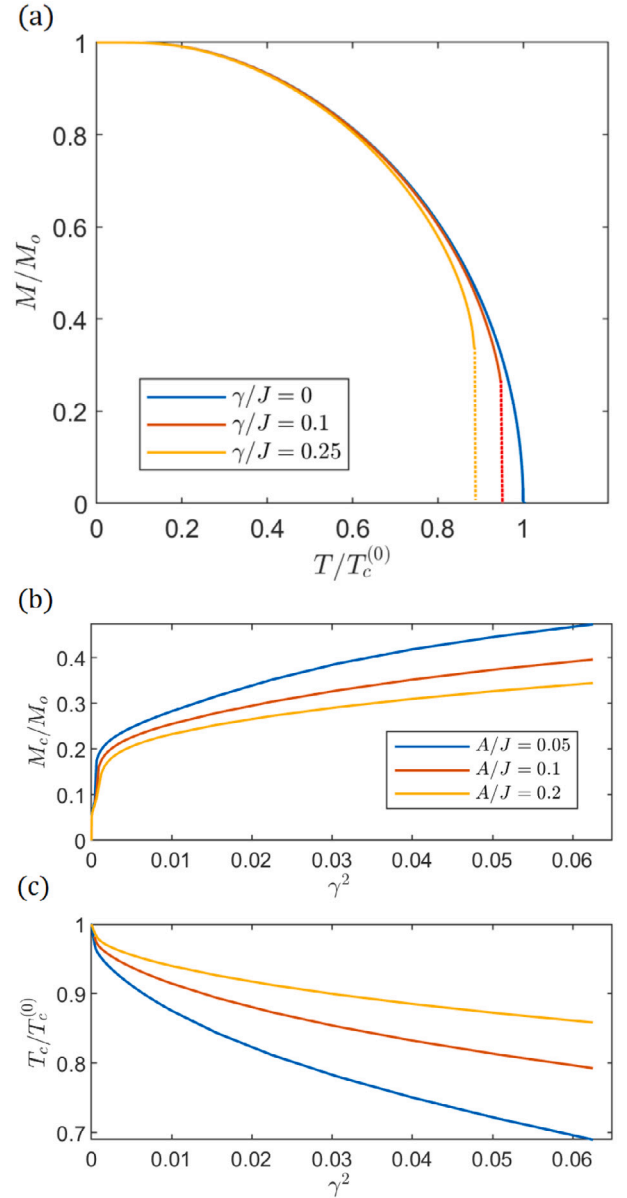


Fig. 2. (a) Magnetization as a function of temperature for several different values of the random fields without an external magnetic field. The dotted lines mark the critical points where the solutions end and the first-order phase transition occurs. We have used $J = 1$ and $A = 0.3$, $z = 4$. (b) and (c) The critical magnetization and the critical temperature as functions of the strength of the random field for several systems with different anisotropy constants.

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 Δ 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. (13), 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 Eq. (13) 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 at zero magnetic field in Fig. 2. Without disorders, the magnetization undergoes the second-order phase transition at the Curie

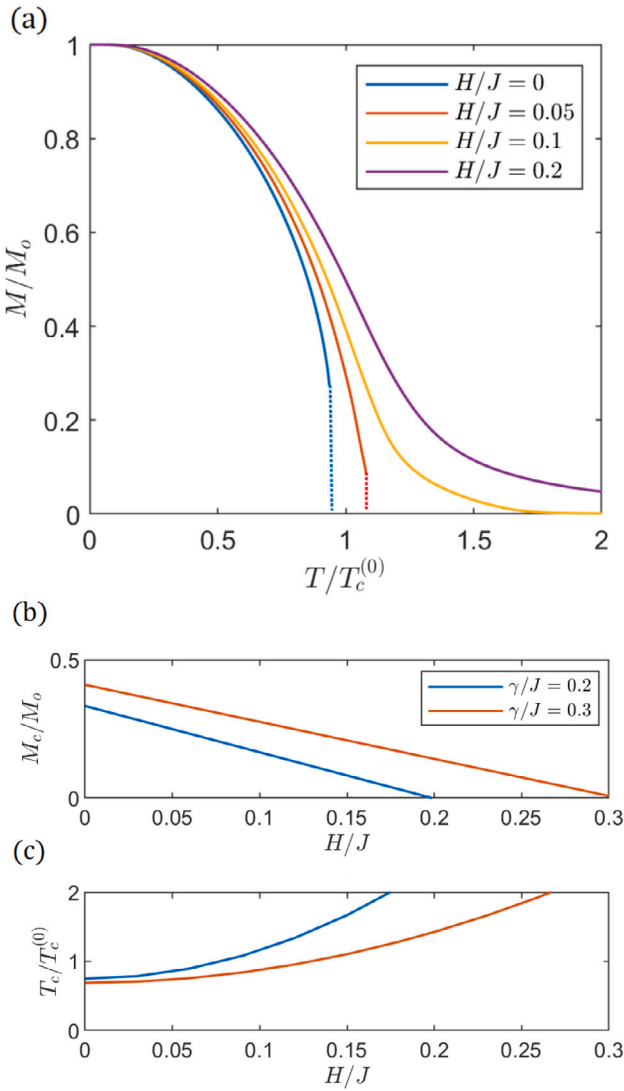


Fig. 3. (a) Magnetization as a function of temperature for several different external magnetic fields. The dotted lines mark the critical points where the solutions end and the first-order phase transition occurs. $A = 0.2$, $z = 4$, and $\gamma = 0.1$. (b) and (c) The critical magnetization and the critical temperature as functions of the external magnetic field for several different random fields. $A = 0.2$, $z = 4$.

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, Δ of Eq. (14), decreases as the random field increases. At low temperature, however, the effect of the random field is negligible since the gap Δ is not significantly different from Δ_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. (13) does not have a solution anymore, indicating that the ferromagnetic phase we have assumed in deriving Eq. (13) 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 Fig. 2b. Since Eq. (13) has no solution for $T > T_c$, the magnetization would be no longer uniform.

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

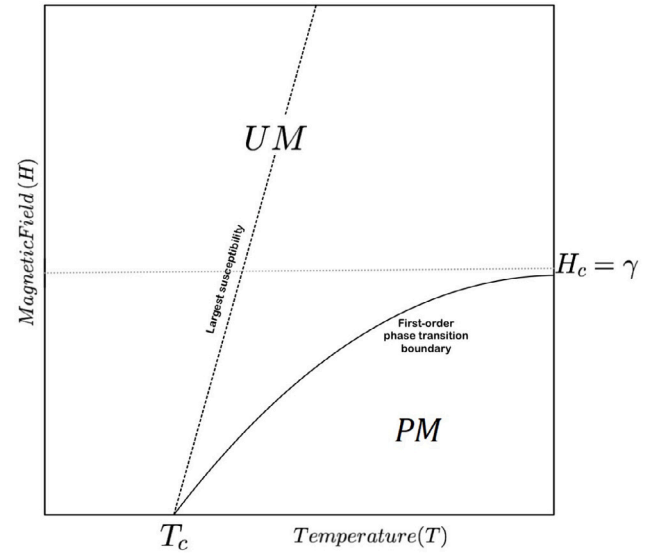


Fig. 4. The phase diagram of a two-dimensional magnetic system with a random field (based on the data presented in Fig. 3c). The solid black line represents the first-order phase transition from the uniform magnetization phase (UM) to a paramagnetic state (PM). The dotted line represents the maximum susceptibility for the uniform magnetization. The critical magnetic field scales with $H_c = \gamma$.

the transition between the time-reversal symmetry-broken phase and symmetry-conserving phase 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. (13) 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 $\Delta = \Delta_0 - \gamma = 2zAM + H - \gamma$ when M is small (or temperature is high). If H is smaller than γ , 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 larger than γ , the magnetization is essentially identical to that without the random field, i.e., there is no phase transition.

Taken together, we may 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 with a uniform magnetization as UM in which the solution of a uniform magnetization exists. For the magnetic field larger than the strength of the random field, the magnetization is at the UM state for all temperatures, i.e., no phase transition. When the magnetic field is smaller than the random field, the first order phase transition appears with the magnetization jump and the transition temperature higher for a larger magnetic field. While our theory cannot address the details after the first order phase transition to a paramagnetic state, we believe such paramagnetic states may contain short-range orders with special topological structures. Since our 2D Heisenberg model does not host Kosterlitz-Thouless (KT) vortices (which are formed in the XY model) [29], the actual domain structure will require further investigation.

CRediT authorship contribution statement

Essa M. Ibrahim: Conceptualization, Methodology, Formal analysis, Software, Validation, Visualization, Writing – original draft. **Ping Tang:** Methodology. **Shufeng Zhang:** Supervision, Project administration, Funding acquisition, Conceptualization, Validation, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

Acknowledgments

This work was partially supported by the U.S. National Science Foundation under Grant No. ECCS-2011331.

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