Reinvestigation of the homogeneous spin model in YbMgGaO₄

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Motivated by a recent inelastic neutron scattering experiment on YbMgGaO₄ [1], we reinvestigate the homogeneous spin model on the triangular lattice. Using the cluster mean-field theory, we study the phase diagram and the magnetic-field-induced phase transition. We find that the phase boundary between the stripe state and the 120° antiferromagnetic state is broadened by the magnetic field, leading to a field-induced phase transition. This phase transition is suppressed by the next-nearest neighbor exchange interaction $J_2 / J_1$ and vanishes as $J_2 / J_1 > 0.13$. We find a parameter space at $J_2 / J_1 = 0.1$, in which the field-induce transition can be achieved and the deviation of theoretical spin excitation energies from experimental data is only 5.4%. Our results imply that an effective homogeneous spin model still works in YbMgGaO₄.

Introduction — Quantum spin liquids refer to a novel state where spins do not form an ordered pattern down to zero temperature in spite of strong spin-spin interactions [2]. A variety of materials and models with frustrated geometries, including the triangular, kagome, and pyrochlore lattices, have been suggested to achieve this state [3–8].

Recently, YbMgGaO₄ has been attracting much attention as a candidate for the quantum spin liquid state [9–19]. In YbMgGaO₄, the magnetic ions, Yb³⁺, form a perfect triangular lattice. The spin-orbit coupling (SOC) and the crystal electric field lead to a Kramers’ doublet for the Yb³⁺ ion, which is described by an effective spin-1/2 local moment [15]. The interaction between the effective spin-1/2 moments is anisotropic because of the spin-orbit-entangled nature of the Kramers’ doublet for the Yb³⁺ ion [19]. In YbMgGaO₄, the magnetic ions, Yb³⁺, form a perfect triangular lattice. The spin-orbit coupling (SOC) and the crystal electric field lead to a Kramers’ doublet for the Yb³⁺ ion, which is described by an effective spin-1/2 local moment [15]. The interaction between the effective spin-1/2 moments is anisotropic because of the spin-orbit-entangled nature of the Kramers’ doublets [20]. Besides, in YbMgGaO₄, Mg and Ga are mixing [11], leading to a disordered effect. Even though this disordered effect has been discussed in several studies [21], the mainstream view still supports that an effective homogeneous model can capture the spin properties in YbMgGaO₄ [15, 23–25, 26] because the extensively used A [16], B [15], and C [27] models [see 1] reproduce inelastic neutron scattering (INS) data at high fields very well.

This idea is being challenged by a new INS experiment, which reported a magnetic-field-induced phase transition in YbMgGaO₄ [1]. As the magnetic field increases, the peak of the magnetic susceptibility moves from the M point to the K point, as shown in Figs. 1 (a) and 1 (b). In the homogeneous model, this change implies a phase transition from the stripe state to the 120° antiferromagnetic state [see Figs. 1 (c) and 1 (d)]. Ref. 1 showed that the C model cannot produce this phase transition. Then a natural question arises. Can the other two models (A and B) produce this phase transition? If not, can we find another homogeneous model to simultaneously describe the phase transition at low fields and the spin excitations at high fields?

To answer these questions, we carefully study the homogeneous spin Hamiltonian, which is extensively used to describe YbMgGaO₄. We first performed classical Monte Carlo simulations for A, B, and C models at different magnetic fields and found that these three models cannot produce the field-induced phase transition [see the supplement [28]]. Next, we study the phase diagram using the cluster mean-field theory. We find a parameter space for the field-induced phase transition and calculate the spin excitation in that region using the linear spin-wave theory. Comparing to INS data, we find that the homogeneous model in that parameter space can simultaneously describe the spin properties of YbMgGaO₄ at both low and high fields.

Model — We study a homogeneous spin Hamiltonian [23, 25, 26, 30], which reads

$$H = \sum_{\langle ij \rangle} \left\{ J_{ij}^z S_i^z S_j^z + J_{ij}^\pm \left( S_i^+ S_j^- + S_i^- S_j^+ \right) \right\}$$

$$+ J_{ij}^{\pm \pm} \left( \gamma_{ij} S_i^+ S_j^- + \gamma_{ij}^* S_i^- S_j^+ \right)$$

$$- \frac{i}{2} J_{ij}^{\pm \pm} \left( \left[ \left( \gamma_{ij} S_i^+ S_j^- - \gamma_{ij}^* S_i^- S_j^+ \right) S_j^+ + S_i^+ \left( \gamma_{ij}^* S_j^+ - \gamma_{ij} S_j^- \right) \right] \right)$$

$$+ \sum_{\langle ij \rangle} J_{ij}^{zz} S_i^z S_j^z + J_{ij}^{xy} \left( S_i^+ S_j^+ + S_i^- S_j^- \right)$$

$$- \sum_{\langle ij \rangle} \left[ g_{\perp} \mu_0 \mu_B (H_z S_i^z + H_y S_i^y) + g_{\parallel} \mu_0 \mu_B H_z S_i^z \right],$$

where $\langle \cdots \rangle$ runs over the nearest neighbors, and $\langle \langle \cdots \rangle \rangle$ runs over the next-nearest neighbors. $S_i^\pm = S_i^x \pm i S_i^y$ and

| Model | A | B | C |
|-------|---|---|---|
| $J_{ij}^{zz}$ (meV) | 0.126 | 0.164 | 0.149 |
| $J_{ij}^{xy}$ (meV) | 0.109 | 0.108 | 0.085 |
| $J_{ij}^{zz}$ (meV) | 0.013 | 0.066 | 0.07 |
| $J_{ij}^{xy}$ (meV) | 0 | 0.098 | 0.1 |
| $J_2 / J_1$ | 0.22 | 0 | 0.18 |
| $g_{\perp}$ | 3.72 | 3.72 | 3.81 |
| $g_{\parallel}$ | 3.06 | 3.06 | 3.53 |

TABLE I: Exchange parameters for different models derived from fitting the spin-wave dispersions. Models A, B and C are from Ref. [16], Ref. [15], and Ref. [27], respectively.
\[ \gamma_{ij} = \gamma_{ji} = 1, e^{i2\pi/3}, e^{-i2\pi/3} \] are the phase factors for the bond \( ij \) along the three principal directions. \( J^z_1 \) and \( J^z_2 \) terms constitute the XXZ model, \( J^\pm_1 \) and \( J^\pm_2 \) are the spin-orbit interactions, and \( J^{zz}_1 \) and \( J^{zz}_2 \) are the next-nearest neighbor exchange interactions. To reduce the number of variables, we set \( J^{zz}_2/J^z_1 = J^z_2/J^z_1 = J_2/J_1 \), following Ref. \[27\] made. \( H_x \), \( H_y \), and \( H_z \) are the magnetic field along the \( x \), \( y \), and \( z \) directions, respectively.

The cluster mean-field theory is a self-consistent approach, which exactly treats interactions inside the cluster and treats interactions between clusters at the mean-field level \[31–33\]. This theory can produce surprisingly accurate boundaries between various phases compared to results from more sophisticated methods \[34\].

For both the 120\(^\circ\) antiferromagnetic (AFM) and stripe states, the lattice can be divided into small identical clusters \( c \) with \( 6 \times 6 \) sites. To reduce the tremendous computational cost of the exact diagonalization of 36 sites, we divide the cluster \( c \) into two sub-clusters \( C_1 \) and \( C_2 \). Each sub-cluster has \( 3 \times 6 \) sites. Then the lattice Hamiltonian is rewritten as

\[
H = \sum_{C_i} H_{C_i} + \sum_{C_i,C_j} H_{C_i,C_j},
\]

where \( H_{C_i} \) describes interactions in the sub-cluster \( C_i \), and \( H_{C_i,C_j} \) describes interactions between two neighboring sub-clusters \( C_i \) and \( C_j \). In the cluster mean-field theory, we treat the interaction term \( J_{ij} S_i S_j \) in \( H_{C_i,C_j} \) at the mean-field level with \( J_{ij} S_i S_j = J_{ij} \langle S_i \rangle \langle S_j \rangle - J_{ij} \langle S_i \rangle \langle S_j \rangle \). Then the total energy of the cluster \( c \) is given by \( E_c = E_1 - E_1^1/2 + E_2 - E_2^1/2 \), where \( E_1 \) and \( E_2 \) are the energies of the sub-cluster \( C_1 \) and \( C_2 \); \( E_1^1 \) and \( E_2^1 \) are the energies of interactions between two sub-clusters, which are given by \( \sum_{(i,j)} J_{ij} \langle S_i \rangle \langle S_j \rangle \).

The 120\(^\circ\) AFM state has three sites in one unit cell [see Fig. 1(d)], and two sub-clusters \( C_1 \) and \( C_2 \) are therefore identical. The stripe state has two sites in one unit cell [see Fig. 1(c)], and these two sub-clusters are not identical. Then a self-consistency between two sub-clusters is required for the stripe state. For each parameter set, we perform simulations for both two states at zero temperature, and the ground state is determined by which one gives a smaller \( E_c \). Due to the finite size, we neglect the multi-\( Q \) state proposed in Ref. \[35\]. However, we believe that our results are reasonable since Ref. \[35\] showed that these multi-\( Q \) states only occupy a very small region in the phase diagram.

Results — We first study the phase transition as a function of \( J^{\pm}_1 \) and \( J^{\pm}_1 \). Figures 2(a) - 2(c) plot phase diagrams for \( J_2/J_1 = 0, 0.05, \) and \( 0.1 \), respectively. The blue and green dotted lines represent the phase boundary between the stripe and 120\(^\circ\) AFM states at \( H = 0 \) and \( H = 2 \) T. The stripe state is inside the boundary and the 120\(^\circ\) AFM state is outside the boundary. The white star shows the location of the B model in the phase diagram. Here, \( J^{\pm}_1 = 0.164 \text{ meV} \) and \( J^{\pm}_1 = 0.108 \text{ meV} \).

![FIG. 1. Schematic of the phase transition induced by the magnetic field in YbMgGaO\(_4\). In panel (a) and panel (b), red dots show the peak position of the static magnetic susceptibility in the Brillouin zone at the magnetic field \( H = 0 \) and \( H = 2 \) T. Panel (c) and panel (d) show the corresponding spin configurations in the real space. The number represents the index of atoms in one unit cell.](image-url)

![FIG. 2. The phase diagram in the plane of \( J^\pm_1 \) and \( J^\pm_1 \). Panels (a), (b), and (c) show results for \( J_2/J_1 = 0, 0.05, \) and \( 0.1 \), respectively. The z-axis represents the uniform magnetization (\( S^z \)) at \( H = 2 \) T. The blue and green dotted lines represent the phase boundary between the stripe and 120\(^\circ\) AFM states at \( H = 0 \) and \( H = 2 \) T. The stripe state is inside the boundary and the 120\(^\circ\) AFM state is outside the boundary. The white star shows the location of the B model in the phase diagram. Here, \( J^{\pm}_1 = 0.164 \text{ meV} \) and \( J^{\pm}_1 = 0.108 \text{ meV} \).](image-url)
uniform magnetization $\langle S^z \rangle$ at $H_z = 2$ T. Figures 2 (a) and 2 (c) show $\langle S^z \rangle < 0.3$, indicating that the ground state is not fully polarized at $H_z = 2$ T.

At $J_2/J_1 = 0$ and $H_z = 0$, the $120^\circ$ AFM state resides in an enclosed region (blue dotted-line) centered at $J_1^{z\pm} = 0$ and $J_1^{z\pm} = 0$. The phase boundary is asymmetric about $J_1^{z\pm} = 0$ as predicted in previous classical Monte Carlo and linear spin-wave theory studies [24]. At $H_z = 2$ T, the boundary for the $120^\circ$ AFM state is slightly broadened, and an overlap region (II) forms. In both I and III regions, the ground state does not change as the magnetic field increases. While in the II region, a phase transition from the stripe state to the $120^\circ$ AFM state occurs as the magnetic field increases. We label the B model as a white star in the phase diagram, outside of the green dotted line. The absence of the field-induced phase transition in the B model is because $J_1^{z\pm}$ is too large.

At $J_2/J_1 = 0.05$ and $H_z = 0$, the ground state is a stripe state for all different pairs of $J_1^{z\pm}$ and $J_1^{z\pm}$, and there is no phase transition. This result is consistent with previous DMRG predictions that $J_2$ supports the stripe state [22]. At $H_z = 2$ T, the region of the $120^\circ$ AFM state shrinks compared to that at $J_2/J_1 = 0$. Continuing to increase $J_2/J_1$ to 0.1, this region becomes very small. These results imply that both the next-nearest neighbor interaction and the SOC $J_1^{z\pm}$ suppress the field-induced phase transition.

To further explain the effect of the next-nearest neighbor interaction, we examine the phase transition as a function of $J_2$ and $J_1^{z\pm}$ in Fig. 3 (a). Here, we set $J_1^{z\pm} = 0$ and the other parameters are the same as the B model. At $H_z = 0$ and $J_2/J_1 = 0$, the phase transition occurs at $J_1^{z\pm}/J_1^{z\pm} = 0.5$. As $J_2/J_1$ increases, the phase boundary moves toward $J_1^{z\pm}/J_1^{z\pm} = 0$ from two ends, and an enclosed region forms at $J_2/J_1 = 0.04$. At $H_z = 2$ T, the phase boundary (green dotted line) has a similar behavior, but an enclosed region forms at $J_2/J_1 = 0.11$. There is a maximal value of $J_2/J_1 (\approx 0.11)$, above which the field-induced transition does not occur. We note that the ratio of $J_1^{z\pm}/J_1^{z\pm}$ has a tiny effect on this maximal value. Figure 3 (b) shows the value of $J_2/J_1$ for the phase transition as a function of $J_1^{z\pm}/J_1^{z\pm}$. At $H_z = 2$ T, as $J_1^{z\pm}/J_1^{z\pm}$ increases, $J_2/J_1$ increases and reaches to a maximum value (about 0.13) at $J_1^{z\pm}/J_1^{z\pm} = 0.5$ and then slowly decreases. The change of $J_2/J_1$ is small and the mean value of $J_2/J_1$ is about 0.11. In A and C models, $J_1^{z\pm}/J_1^{z\pm} = 0.865$ and 0.57, and $J_2/J_1 = 0.22$ and 0.18. These two values of $J_2/J_1$ are much larger than 0.11, leading to the absence of the field-induced phase transition.

Next, we analyze the spin excitations using the linear spin-wave theory. At high fields, the spin excitation energy at the $\Gamma$ point is given by $E(H_z) = g_\parallel \mu_B H_z - 2x$ and $E(H_x) = \sqrt{(g_\perp \mu_B H_x + x)^2 - x^2}$, where $x = \frac{1}{2}(J_1^{z\pm} + J_2^{z\pm}) - 3(J_1^{z\pm} + J_2^{z\pm})$. We fit these two equations to the time-domain terahertz spectra obtained from Ref. [27] and get $g_\parallel = 3.8$, $g_\perp = 3.55$, and $x = -0.0414$ [28]. Considering results shown in
Fig. 2 and Fig. 3 we calculate the momentum dependent spin excitation energy $E_{sw}(q)$ in the parameter space of $\{J_{1}^{zz} \in [0.11, 0.21], J_{2}/J_{1} \in [0, 0.4], J_{1}^{zz}/J_{2}^{zz} \in [-0.2, 0], J_{1}^{zz}/J_{1}^{zz} \in [-0.6, 0.6]\}$. The deviation $R$ of $E_{sw}(q)$ from the INS data $E_{exp}(q)$ [27] is defined as $R = \frac{1}{L} \sum_{q} \sqrt{(E_{sw}(q_{i}) - E_{exp}(q_{i}))^{2}}$, where $L$ is the total number of momenta. Figure 3 plots $\log(R)$ as a function of $J_{1}^{zz}$ and $J_{2}/J_{1}$. Here, we choose values of $J_{1}^{zz}$ and $J_{2}/J_{1}$ that produce the smallest $R$ at a given $J_{1}^{zz}$ and $J_{2}/J_{1}$. The minimal $R$ locates at $J_{1}^{zz} = 0.146$ meV and $J_{2}/J_{1} = 0.22$.

Here, we note a paradoxical fact that a small $J_{2}$ contributes to the field-induced phase transition but produces a large deviation $R$. To make $R$ as small as possible, we choose a large allowed value of $J_{2}$ ($J_{2}/J_{1} = 0.1$) and carefully analyze results. Figure 5 (a) plots $R$ as a function of interactions $J_{1}^{zz}$ and $J_{1}^{\pm \pm}$. We set $J_{1}^{zz} = 0.158$ meV because it produces the smallest $R$ at a given $J_{1}^{\pm \pm}$, $J_{1}^{\pm \pm}$, and $J_{2}/J_{1}$. The blue dotted line is the phase boundary at $H_{z} = 2$ T calculated from the cluster mean-field theory. $R$ decreases slowly as $J_{1}^{\pm \pm}$ increases. We chose a point near the boundary (the white star in Fig. 5 (a)) and compute the momentum-dependent spin excitation energy in Figs. 5 (b) and 5 (c). Yellow dots in Fig. 5 (b) and 5 (c) are experimental results obtained from Ref. [27]. The theoretical line and the experimental data are almost overlap with a small deviation $R = 5.432\%$. The uniform magnetization $\langle S^{z} \rangle$ of the chosen point is about 0.25, which is 25% larger than the experimental value (0.175) [16]. We note that all parameters inside the blue line produce similar values of $R$ and $\langle S^{z} \rangle$.

At last, we perform classical Monte Carlo simulations [35] with the chosen parameters: $J_{1}^{zz} = 0.158$ meV, $J_{1}^{\pm \pm} = 0.092$ meV, $J_{1}^{\pm \pm}/J_{1}^{zz} = 0.02$, $J_{1}^{\pm \pm}/J_{1}^{zz} = 0.1$, and $J_{2}/J_{1} = 0.1$. Figures 5 (d) - 5 (g) show the static magnetic susceptibilities at $T = 0.1$ K for $H_{z} = 0, 1$ T, 2 T, and 3 T, respectively. At $H_{z} = 0$, the ground state is the stripe state with a strong intensity at the M point. Increasing the magnetic field, the intensity at the M point is suppressed and the intensity at the K point is enhanced. At $H_{z} = 2$ T, the ground state becomes a 120° AFM state, and its z component is polarized. These results show a clear magnetic-field-induced phase transition, which is consistent with the result of our cluster mean-field theory calculations.

Summary — We have carefully examined the spin Hamiltonian to understand the spin properties in YbMgGaO$_{4}$. We study the phase diagram of this Hamiltonian using the cluster mean-field theory. We find that both the next-nearest neighbor interaction and the SOC $J_{1}^{\pm \pm}$ suppress the magnetic-field-induced phase transi-
tion. We find a maximal value of $J_2/J_1$ (equals to 0.13), above which the magnetic-field-induced phase transition cannot occur. On the other hand, the minimal deviation of the theoretical spin excitation energy from experimental data locates at $J_2/J_1 = 0.2$. To minimize the deviation $R$, we use a large allowed $J_2/J_1$ and find a parameter region, in which the INS data can be reproduced with a deviation of 5.432%.

The INS data of YbMgGaO$_4$ has a broad spectrum, and the energy of the peak is not well defined. The 5.432% deviation produced by our model is therefore acceptable. We find a parameter space at $J_2/J_1 = 0.11$ and $J_2/J_1 = 0.12$. To shrink a parameter space to one point, it is necessary to further study this spin model.

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