Existence of a tricritical point in the antiferromagnet $KFe_3(OH)_6(SO_4)_2$ on a kagome lattice

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We study the phase diagram in the H-T plane of the potassium jarosite compound $KFe_3(OH)_6(SO_4)_2$ for the antiferromagnetic XY model with Dzyaloshinskii-Moriya (DM) interaction using the mean-field theory for different values of DM. In our approach, we obtain the tricritical point in the H-T plane and the adjustment has a strong correlation with experimental data.

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Recent experimental studies of the compounds $KFe_3(OH)_6(SO_4)_2$ and $AgFe_3(OH)_6(SO_4)_2$, have indentified the existence of a tricritical point (TCP) at temperatures of around $T = 50$ K [1,2]. Such systems have been researched as a prototype of the antiferromagnet on a kagome lattice using the Heisenberg model [1,3–9]. The behavior of this transition has been investigated by analysis of the M-H phase diagram and TCP has also been obtained in the H-T plane for the K-jarosite system [1]. Different theoretical approaches have been made to such compounds but none has been able to detect the existence of TCP.

In K-jarosites, above a certain critical value of the field perpendicular to the kagome plane, H_c, a phenomenon known as spin canting emerges [10], in which the spins are tilted at small angles on their axis rather than being coparallel. This phenomenon causes some antiferromagnetic materials to exhibit a nonzero magnetic moment at temperatures above absolute zero. Therefore resulting, for these systems, in a transition from an antiferromagnetic state to a long-range order ferromagnetic (LRO) state by spin canting in the kagome lattice [1,2]. Such a transition happens to relatively large fields ($H \gtrsim 10T$); however, the presence of tricritical behavior has not been observed in the literature [11,12]. In particular, the studies of Fujita *et al.* [3,4] and Elhajal *et al.* [13], and references therein, suggest an interaction of type Dzyaloshinskii-Moriya (DM) which has encouraged the theoretical study of this work.

Here, adopting the antiferromagnetic XY model with DM interaction in the presence of a magnetic field for K-jarosite compounds and following the same ideas of Lee *et al.* [14], the Hamiltonian considered is given by

$$\mathcal{H} = J\sum_{\langle i,j\rangle}\vec{s}_i\cdot\vec{s}_j - \sum_{\langle i,j\rangle}\vec{D}_{ij}\cdot(\vec{s}_i\times\vec{s}_j) - \sum_i \vec{H}\cdot\vec{s}_i. \quad (1)$$

The sum $\langle i,j\rangle$ is restricted to the nearest neighbors, $\vec{D}_{ij} = D\vec{z}$ is the DM interaction, $\vec{H}$ is the magnetic field, and the spins $\vec{s}_i = (\cos\theta_i, \sin\theta_i)$ are unitary classical vectors confined in the kagome lattice. So, the reduced Hamiltonian can be written as

$$\beta\mathcal{H} = K\sum_{\langle i,j\rangle}\cos(\theta_i-\theta_j) - D_0\sum_{\langle i,j\rangle}\sin(\theta_i-\theta_j) - h\sum_i\cos\theta_i, \quad (2)$$

where $K \equiv J/k_BT$, $D_0 \equiv D/k_BT$, $h \equiv \mu_BH/k_BT$, μ_B is the Bohr magneton, and k_B is the Boltzmann constant. On the other hand, the free energy per spin is given as

$$\begin{aligned}\psi(a_\alpha,\phi_\alpha) = {}& \frac{q}{2n}K\sum_{\alpha=1}^{n}\sum_{\beta=1,\beta\neq\alpha}^{n} R(a_\alpha)R(a_\beta)\cos(\phi_\alpha-\phi_\beta)\\ & - \frac{q}{2n}D_0\sum_{\alpha=1}^{n}\sum_{\beta=1,\beta\neq\alpha}^{n} R(a_\alpha)R(a_\beta)\sin(\phi_\alpha-\phi_\beta)\\ & - \frac{h}{n}\sum_{\alpha=1}^{n}R(a_\alpha)\cos\phi_\alpha\\ & + \frac{1}{n}\sum_{\alpha=1}^{n}\{a_\alpha R(a_\alpha) - \ln[2\pi I_0(a_\alpha)]\},\end{aligned} \quad (3)$$

where n is the number of sublattices for the antiferromagnetic structure (for kagome lattice, $z = 4$ and $n = 3$; see Ref. [14] for more details), $q \equiv z/(n-1)$, z is the coordination number, $R(a_\kappa) = I_1(a_\kappa)/I_0(a_\kappa)$ (with $\kappa = \alpha,\beta$), and $I_\nu(x)$ is the modified Bessel function of the first kind of integer order ν. The parameters a_κ and ϕ_κ are, respectively, the magnitude and the angle of the local mean field on site κ [14], and are obtained for the solution of the coupled equations:

$$\begin{aligned} Kq\sum_{\beta=1,\beta\neq\alpha}^{n}R(a_\beta)\cos\phi_\beta + D_0q\sum_{\beta=1,\beta\neq\alpha}^{n}R(a_\beta)\sin\phi_\beta + a_\alpha\cos\phi_\alpha = h,\\ Kq\sum_{\beta=1,\beta\neq\alpha}^{n}R(a_\beta)\sin\phi_\beta - D_0q\sum_{\beta=1,\beta\neq\alpha}^{n}R(a_\beta)\cos\phi_\beta + a_\alpha\sin\phi_\alpha = 0.\end{aligned} \quad (4)$$

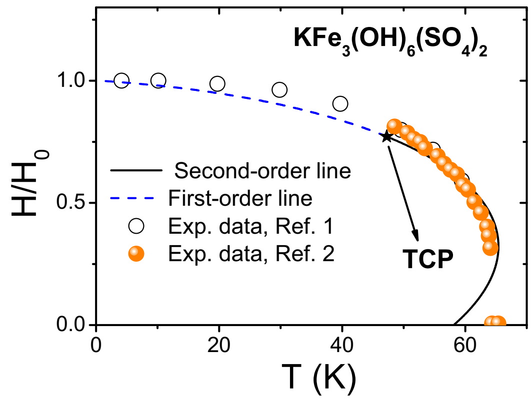


FIG. 1. (Color online) Phase diagram for the compound potassium jarosite. Solid lines represent second-order transition, while dotted lines represent first-order transition. Open circles and solid circles are the experimental data from Refs. [1] and [2], respectively. TCP is obtained by solving numerically Eqs. (6) and $H_0 = 16.8T$ [1].

Through minimizing the free energy, Eq. (3), with respect to a_κ and ϕ_κ we get a line of second-order transition able to build the phase diagram in the H-T plane, as shown in Fig. 1. The first-order line is obtained by equating the equations for the free energy of antiferro- and ferromagnetic phases [with $\phi'_\kappa s = 0$, $a_\kappa = a_0$, and $K < 0$ in Eq. (2)]. With these data, we observe a strong correlation with the experimental data, particularly in the second-order transition region except for 58 K $< T <$ 65.4 K ($T_N \approx 65$ K for potassium jarosites [2,15]), where a marked reentrance appears possibly by the mean-field approximation adopted in the present work. In a recent paper, Matan and collaborators [1] obtained hysteresis behavior in the H-T plane, with $H = H_0 = 16.8T$ when T goes to zero. This behavior identifies first-order line transition with a change to second-order line transition in the vicinity of $T \approx 50$ K. This characterizes the existence of TCP. In our approach, we expand the equation for the free energy in terms of the absolute value of the magnetization per site, $\vec{M}$, given by

$$\psi = \psi_0 + A(T,h,a_\alpha,\phi_\alpha)|\vec{M}|^2 + B(T,h,a_\alpha,\phi_\alpha)|\vec{M}|^4 + \cdots , \quad (5)$$

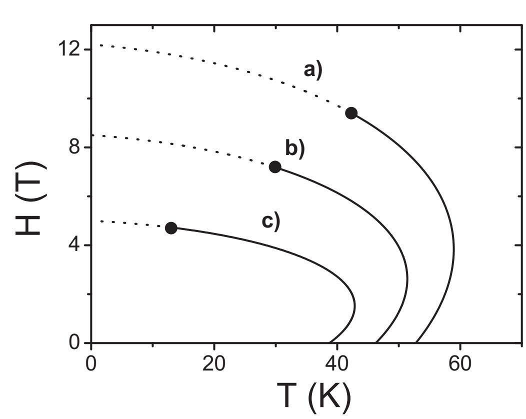


FIG. 2. Phase diagram in the H-T plane for kagome lattice: (a) $D/k_B = 0.8$ K, (b) $D/k_B = 0.5$ K, and (c) $D/k_B = 0.1$ K.

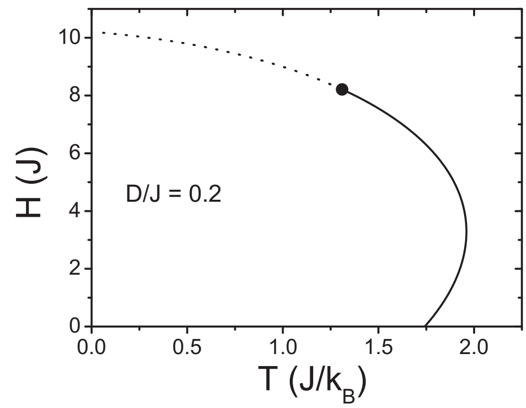


FIG. 3. Diagram H-T for the square lattice, with $D/J = 0.2$. Solid lines represent continuous transition from the determined phase (helical phase) to a paramagnetic phase and the dotted line represents the transition of the first order. The field H is given in units of exchange interaction, J.

where $\vec{M}$ can be decomposed in two components, M_x and M_y, proportional to $R(a_\alpha)\cos\phi_\alpha$ and $R(a_\alpha)\sin\phi_\alpha$, respectively, and a_α,ϕ_α are solutions of Eqs. (4). The TCP is obtained by numerically solving the coupled equations [16]

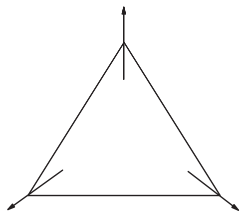
$$A(T,h,a_\alpha,\phi_\alpha) = 1 \quad \text{and} \quad B(T,h,a_\alpha,\phi_\alpha) = 0. \quad (6)$$

By solving Eqs. (6), with $J/k_B = 42$ K and $D/k_B = 1$ K (see Ref. [4] for comparison), we get $(T_{\mathrm{TCP}},H_{\mathrm{TCP}}) = (47.3\ \mathrm{K}, 12.9T)$. These values are quantitatively very good when compared with data experimentally obtained, i.e., $(T_{\mathrm{TCP}},H_{\mathrm{TCP}}) \approx (50\ \mathrm{K}, 13.4T)$ [1,2]. Although, by using different values of DM interaction for kagome lattice, we get the TCP even for small values of D/k_B (Fig. 2). In the absence of such interaction, the phase diagram is qualitatively the same as shown in Fig. 2 obtained at Ref. [14].

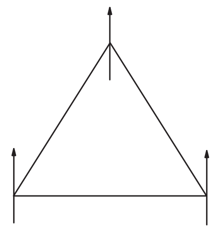
It is interesting to note that this configuration is identical to the diagram obtained for the square lattice, as shown in the Fig. 3, despite the kagome (in its antiferromagnetic configuration) tripartite lattice. Contrastingly the square lattice is bipartite and shows no transition from helical-nonhelicoidal, as presented by the kagome and triangular lattices even with $D = 0$. This transition occurs only on lattices with nonzero helicity [13], such as the kagome lattice, an example of which can be seen in Figs. 4(a) and 4(b), and the values obtained for TCP are shown in Table I for different D/k_B. As D goes to zero, the curve of continuous transition extends to the whole range of T in accordance with Ref. [14]. Likewise, for $D = 0$ (or very small D, as shown in Fig. 5), a transition from the

TABLE I. TCPs obtained for some values of interaction DM in a kagome lattice, shown in the graph of Fig. 2.

D/k_B (K)	TCP (T,H)
1	(47.3 K, 12.9 T)
0.8	(42.3 K, 9.4 T)
0.5	(29.9 K, 7.2 T)
0.1	(13 K, 4.7 T)



(a) Helicoidal configuration (helical) known as $q = 0$.



(b) No-helicoidal configuration(nonhelical).

FIG. 4. Spin configurations in triangular and kagome tripartite lattices.

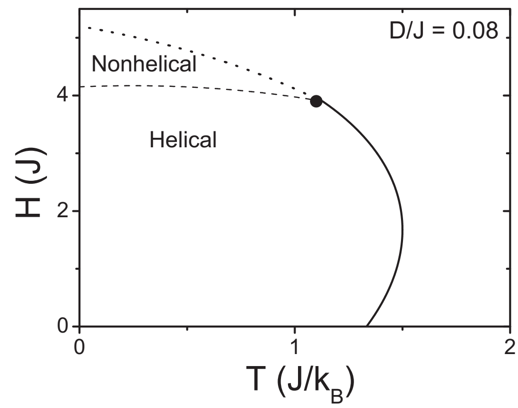


FIG. 5. Diagram H-T for triangular lattice, with $D/J = 0.08$. The solid line represents the continuous transition from the determined phase (helical phase) to the paramagnetic phase; the trajectory line represents the helical-nonhelical transition [14] and the dotted line represents the transition of the first order. The field of H is given in exchange interaction units, J.

helical phase to another nonhelical phase arises, as discussed in Ref. [14], and higher values of D lead to higher values of the ordering temperature to the zero field. In particular, for kagome lattices, small values of DM interaction imply a breaking of degeneracy of the system so that there is no transition from the helical type to nonhelical for any D.

The phase diagram in the H-T plane was obtained for the square lattice using the same procedure adopted for the kagome/jarosite lattice, with $z = 4$ and $n = 2$. For relatively high values of D there is no qualitative difference between the phase diagrams in the H-T plane of kagome and square lattices and this, we suppose, is due to the fact that both have the same coordination number, an important factor in mean-field approximation. In the absence of DM interaction, the phase diagram shows a continuous transition line that separates a determined phase from an undetermined phase without the degeneracy presented by the triangular lattice phase diagram, and TCP obtained for $D/J = 0.2$ is $(k_B T/J, H) = (1.31, 8.21)$, with H in units of J. Similarly the phase diagram for the triangular lattice is shown in Fig. 5 and we can notice the existence of the first- and second-order lines separated by a TCP located at $(k_B T/J, H) = (1.1, 3.9)$, even to a very small value of D.

In summary, this study investigates the existence of TCP in the phase diagram in the H-T plane by using the antiferromagnetic XY model in the presence of an external magnetic field and DM interaction, and a strong correlation exists between the theoretical results and the experimental data. The presence of the phenomenon of reentrance is considered in the light of the mean-field approximation adopted for cluster with one spin and it has been obtained in many other works known in the literature with some magnetic anisotropy (see, for example, Ref. [17], and references therein). As is well known, within such kind of mean-field framework, the strict criticality of the system is lost, and the real dimensionality of the system is only partially incorporated through the coordination number of the lattice. However, for tridimensional lattices, to which we devoted our calculations, we believe that the present results are, to a certain extent, of qualitative and quantitative relevance. However, it shows that this mean-field approach leads to the conclusion that the tricritical point at which the phase transition changes from second to first order may exist in the system under consideration, and the use of clusters with a greater number of spins can significantly improve this theoretical fit.

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