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Triangular lattice antiferromagnet $RbFe(MoO_4)_2$ in an applied magnetic field

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Abstract

$RbFe(MoO_4)_2$ is a rare example of a nearly two-dimensional Heisenberg antiferromagnet on a triangular lattice. We have studied its low-temperature magnetic properties by means of single crystal ESR spectroscopy and magnetization measurements. On the basis of the obtained results an $H-T$ phase diagram, containing at least five different magnetic phases is constructed. In zero field, $RbFe(MoO_4)_2$ undergoes a phase transition at $T_N = 3.8$ K into a noncollinear spin structure with all the spins confined in the hexagonal plane. The application of an external magnetic field in the plane induces a collinear spin state ($H_{c1} = 47$ kOe, $H_{c2} = 71$ kOe) and produces magnetization plateaux at one-third of the saturation moment. Both the ESR and magnetization measurements also clearly indicate an additional first-order phase transition in a field of 35 kOe. The exact nature of this phase transition is unclear.

Keywords: Two-dimensional antiferromagnet; Triangular spin structure

1. Introduction

Antiferromagnetic exchange systems on a triangular planar lattice (AFMT) have been intensively studied theoretically [1,2]. In the molecular field approximation their ground state is a "triangular" planar spin structure with the three magnetic sublattices arranged 120° apart. The orientation of the spin plane remains arbitrary in the exchange approximation.

For a classical antiferromagnet in a weak magnetic field there are two degenerate spin configurations: the umbrellalike structure (U), where all the three magnetization sublattices are equally tilted towards the field and the planar structure (P), where all the spins lie in the plane parallel to the magnetic field. The P-configuration undergoes a transition to a collinear "up-up-down" phase at $H_c = H_{sat}/3$, where H_{sat} is a saturation field. In the magnetic fields above H_c the structure is again noncollinear, although two of the three sublattices remain parallel to each other forming the so-called canted phase.

Finally, at the saturation field $H = H_{sat}$ there is a spin–flip transition. Quantum fluctuations play an important role in the formation of the ground state of an antiferromagnet on a triangular lattice [2]. They reduce the free energy of the planar structure in a magnetic field and stabilize the collinear phase in a range of magnetic fields $H_{c1} \leqslant H \leqslant H_{c2}$ including H_c.

For the AFMT with the easy-plane-type anisotropy a similar field dependence is expected when a magnetic field is applied in the easy-plane, while the U-structure takes place for magnetic fields applied along the C_3-axis, provided that the easy-plane anisotropy is sufficiently strong.

There are three resonance modes for purely two-dimensional (2D) triangular spin structure [2]. Two of them are degenerate in zero field but have

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different field dependencies, the third mode remains zero in any field.

The introduction of a weak interplane antiferromagnetic exchange does not alter significantly the evolution of the triangular structure in a magnetic field described above. It may, however, result in the appearance of additional field-induced phase transitions where the mutual orientation of spins in neighboring planes is changed. It may also modify the values of the critical fields H_{c1}, H_{c2} [3]. The influence of the interplane exchange on the resonance spectrum is likely to be seen in a splitting of the main resonance frequencies and also by the appearance of new resonance modes due to the increase in the number of sublattices.

$RbFe(MoO_4)_2$ is a rare example of a nearly 2D Heisenberg antiferromagnet on a triangular lattice. We have verified experimentally the above theoretical concepts using single crystal samples of $RbFe(MoO_4)_2$.

2. Samples and static magnetic properties

Single crystal samples of $RbFe(MoO_4)_2$ were synthesized by means of the spontaneous crystallization from a flux melt. The single crystals have the form of thin hexagonal plates with the in-plane size of about 3 mm.

The field and temperature dependencies of the magnetization were measured using a vibrating sample magnetometer in a field up to 120 kOe (the reported value of the saturation field of $RbFe(MoO_4)_2$ is 186 kOe [4] and lies well above our experimental field limit). For $H \parallel C_3$ the magnetization increases linearly in the field for all values of the applied fields. For $H \perp C_3$, the field dependence of the magnetization is much more complicated (Fig. 1).

There are abrupt changes in the magnetization slope at 47 and 71 kOe, with the magnetic susceptibility being significantly reduced in the region between these fields. This behavior is consistent with the concept of a collinear phase stabilization by thermal or quantum fluctuations [2]. We believe that $H_{c1} = 47$ kOe and $H_{c2} = 71$ kOe correspond to the lower and upper critical fields for the formation of a collinear spin structure. In addition to these two transitions, a first-order phase transformation was detected at $H_{c3} = 35$ kOe accompanied by a well pronounced hysteresis in the **M(H)** magnetization curve (see inset in Fig. 1).

3. Antiferromagnetic resonance

Magnetic resonance spectra were recorded using the transmission-type ESR spectrometer with a set of microwave cavities having their resonant frequencies in the range 18–110 GHz. For $H \parallel C_3$ two resonance branches were found. The frequency of the first branch rises with the field, while the frequency of the second branch decreases (see Fig. 2). The frequencies of these two branches are monotonic functions of the applied magnetic field as expected for the umbrella-like structure.

For $H \perp C_3$ a complicated nonmonotonic field dependence of all the resonant frequencies were observed (see Fig. 3). The values of the phase transition fields are in a good agreement with the results of static magnetization measurements. For the triangular system with an antiferromagnetic interplane exchange interaction there are five nonzero resonance modes (the sixth mode remains zero in any field because of the absence of any in-plane anisotropy). We have observed all five of them. The resonance frequencies were calculated by V.I. Marchenko for weak magnetic fields when the exchange triangular spin configuration is only slightly distorted. This calculation follows the macroscopic theory based on a classical Lagrange function [5]. The calculated frequencies are presented in Fig. 3 by the dashed lines.

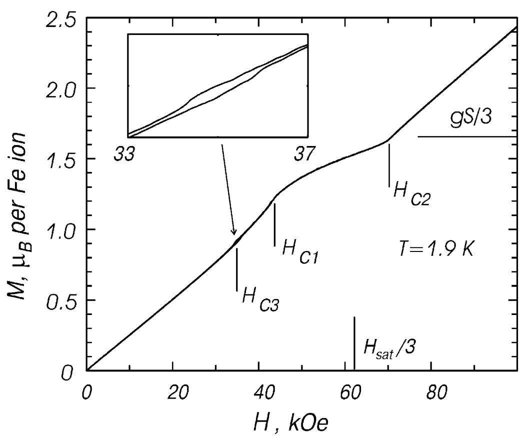

Fig. 1. Magnetization vs. magnetic field at $H \perp C_3$.

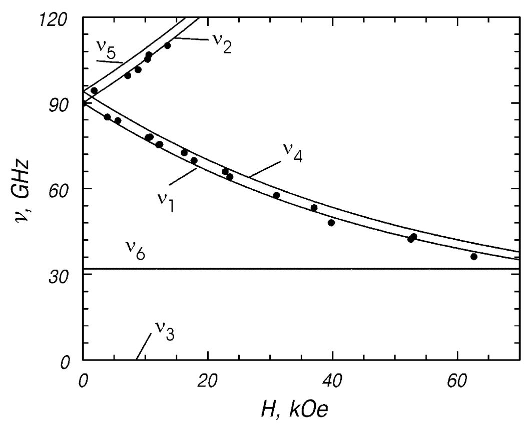

Fig. 2. ESR frequency vs. magnetic field at $H \parallel C_3$, $T = 1.3$ K.

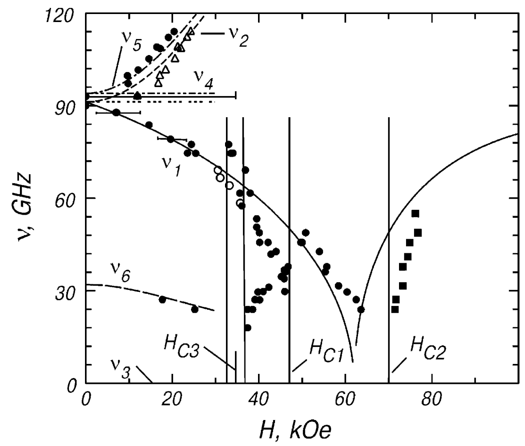

Fig. 3. ESR frequency vs. magnetic field at $H \perp C_3$, $T = 1.3$ K.

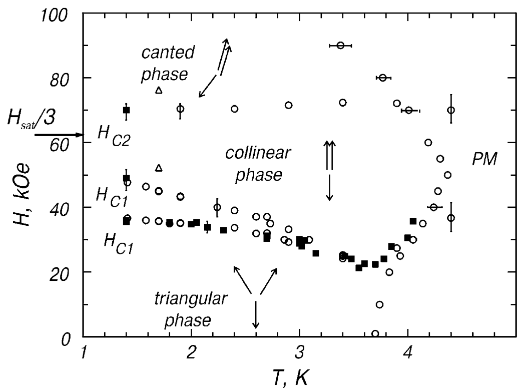

Fig. 4. $H-T$ phase diagram derived from magnetization and ESR measurements, data marked by triangles are taken from powder measurements [4].

The frequencies marked in Fig. 3 as $\nu_{1,2,4,5}$ correspond to the resonance modes of spin oscillations perpendicular to the basal plane. For the modes $\nu_{3,6}$ the oscillating components lie within the plane. The modes with frequencies $\nu_{1,2,3}$ correspond to the in-phase oscillations of spins in the neighboring planes and for the modes with frequencies $\nu_{4,5,6}$ the spins in the neighboring planes oscillate out of phase. Note that for a purely 2D AFMT only the modes $\nu_{1,2,3}$ are present.

From the observed values of the resonance gaps and of the critical fields one can evaluate the effective spin-Hamiltonian parameters (we use the notations of Ref. [2]). The five spin resonance branches shown in the low-field range in Fig. 3 and the magnetization curves may be reasonably described by using only three fitting parameters: effective exchange and anisotropy fields $H_E = 6JS/(g\mu_B) = 62$ kOe, $H_A = DS/(g\mu_B) = 5.7$ kOe, and the ratio of the interplane to intraplane exchange integrals $J'/J = 0.045$. Here $S = \frac{5}{2}$ is the spin value of Fe^{3+} ions. For $H \perp C_3$ the triangular spin structure is already strongly distorted in a field of about 10 kOe. Therefore, the macroscopic theory of magnetic dynamics [5] cannot be adequately used. In order to find at least an approximate description of the resonant frequencies over the whole range of applied fields, we used the calculations of Ref. [2], which are valid for the three sublattice 2D model in a classical approximation. The results of these calculations are shown in Fig. 3 by the solid lines.

A phase transformation near the critical field H_{c3} is accompanied by a hysteresis in the microwave absorption and by a distortion of the branche ν_6. This mode has a nonzero frequency due to the interplane exchange interaction, so the dramatic transformation of this mode corroborates the suggestion that this phase transition involves a change in the mutual orientation of the spins in the neighboring planes predicted in Ref. [3]. Another indication of the interplane exchange is the splitting of the rising ESR mode (the difference in resonance fields of the modes ν_2 and ν_5.

Recent neutron scattering experiments [6] revealed the magnetic structure of $RbFe(MoO_4)_2$ to be indeed triangular in the basal plane, but along the *c*-axis it is incommensurate rather than simply antiferromagnetic. According to this observation the spins in the neighboring planes are not aligned antiparallel to each other but form an angle of approximately 163°. Thus, another possible interpretation of the first-order phase transition at 35 kOe may be the transition from the incommensurate state to a commensurate one, which is favorable in the magnetic field. The resulting magnetic phase diagram of $RbFe(MoO_4)_2$ for $H \perp C_3$ is plotted in Fig. 4.

4. Conclusion

We have observed a sequence of spin-reorientation transitions in a planar antiferromagnet on the triangular lattice. The magnetization curves and the spin resonance frequencies are consistent with a mode involving strong in-plane exchange interactions and a weak antiferromagnetic exchange between the planes. All five nonzero resonance modes of this noncollinear quasi-2D magnetic system were observed experimentally.

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