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Coexistance of spiral and commensurate structures in a triangular antiferromagnet KFe(MoO$_4$)$_2$

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Abstract. Magnetization, specific heat, magnetic resonance and neutron diffraction measurements are used to study the magnetic structure of KFe(MoO$_4$)$_2$. This stacked triangular antiferromagnet (T$_N$=2.5 K) demonstrates an unusual breaking of the spin system into two intercalated and almost independent 2D subsystems. One is a collinear antiferromagnet with a simple spin-flop behavior. The other is a spiral magnet. The spin structure may be explained assuming two types of inequivalent magnetic planes with distorted triangular lattices of Fe^{3+} (S=5/2) ions.

1. Introduction

Frustrated magnets are sensitive to weak interactions or small distortions of crystal lattice. For example, a 2D triangular lattice antiferromagnet should demonstrate a transition from the three sublattice structure with magnetizations at 120° apart to the incommensurate spiral structure at a tiny distortion, which breaks the 3-fold axis symmetry [1]. The in-plane spiral structures on distorted triangular lattices were found, e.g. in the ABX$_3$ family of triangular lattice antiferromagnets see, e.g. [2], and in the S=1/2 quasi-2D triangular lattice antiferromagnet Cs$_2$CuCl$_4$ [3]. A quasi 2D antiferromagnet with a regular triangular lattice RbFe(MoO$_4$)$_2$ has 120°-structure in stacked magnetic layers, while the mutual orientation of spins in the adjacent layers corresponds to a spiral structure with a wavevector perpendicular to magnetic layers [4, 5]. In this paper we describe a relative compound, triangular antiferromagnet KFe(MoO$_4$)$_2$ which has the quasi-2D magnetic structure with stacked magnetic layers of $S = 5/2$ Fe^{3+} ions placed on a lattice of isosceles triangles. This distorted lattice appears from a regular triangular lattice due to a structural transition at T =311 K, when equilateral triangles become stretched [6]. The high temperature symmetry group is D^3_{3d}, the lattice parameters are $a = 5.66\mathring{A}$ and $c = 7.12\mathring{A}$. At the structural phase transition, in addition to the distortion of "triangular" layers, the size of the elementary cell along the c-axis is doubled. There is a second structure transition at T=139 K [6, 7] at which the space group remains unchanged. The low temperature symmetry is, according to Ref[6] monoclinic with the space group C^3_{2h}. The actual lattice distortion is too small to be detected with the resolution of x-ray and neutron experiments, therefore a hexagonal lattice notation will be used further. Nevertheless, the distortion, appearing at 311 K is clearly seen by observation of segnetoelastic domains in a polarized light. The anticipated displacement of K-ions, resulting in the period doubling and symmetry lowering is presented following Ref.[8] in Figure1. The group C^3_{2h} enables nonequivalent adjacent magnetic layers. Thus, due to this structural transition, two different exchange integrals J_1 and J_2 within a magnetic layer might be there, and, besides, the exchange integrals in the adjacent layer should take other values J'_1 and J'_2 because Fe^{3+}-layers become crystallographically inequivalent. The anticipated exchange paths between magnetic ions in the low temperature crystal lattice of $KFe(MoO_4)_2$ are marked on Figure 1.

The early study of the magnetization curves and electron spin resonance [9] revealed a multi-branch ESR spectrum and a magnetization curve with three anomalies before saturation. It was suggested that these macroscopic features correspond to a superposition of two types of magnetic structures - of a spiral and of a collinear structures with spins lying in the plane of magnetic layers. Layers carrying spiral structure were denoted as S-layers and collinear structures - as C-layers. This suggestion was verified in elastic neutron scattering experiments, the first report is published in [10]. In the present paper we present the temperature dependence of the magnetic Bragg peaks, compare it to the specific heat data, and consider their correspondence to magnetic resonance data.

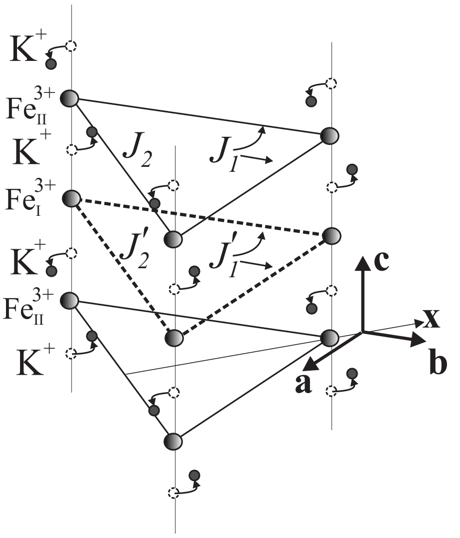

Figure 1. Schematic representation of lattice distortion at T=311 K according to [7].

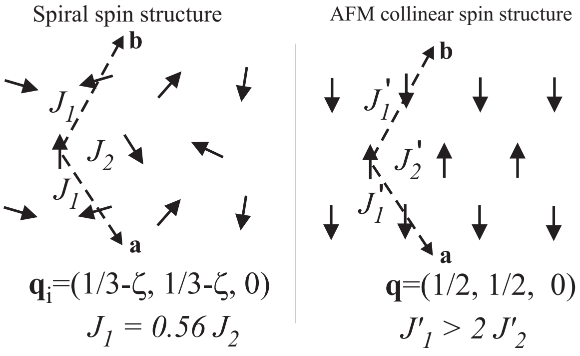

Figure 2. Anticipated spin structures for S and C-layers

2. Experimental results and discussion

The crystal samples of $KFe(MoO_4)_2$ are transparent thin plates of a triangular shape, with the planes perpendicular to the former 3-fold axis. The elastic neutron scattering data were taken on the HB-1 and HB-1A 3-axis spectrometers at Oak Ridge National Laboratory operating in 2-axis mode, using a Pyrolitic Graphite PG(002) monochromator to select λ = 2.46 Å for HB-1

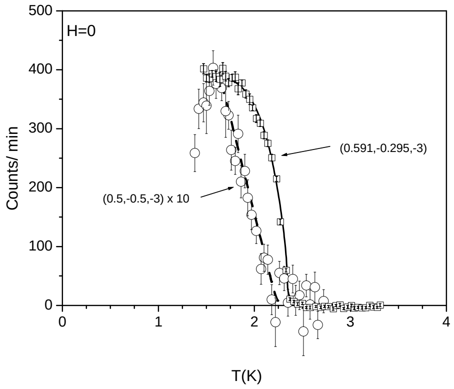

Figure 3. Temperature dependence of the magnetic Bragg peaks in zero field.

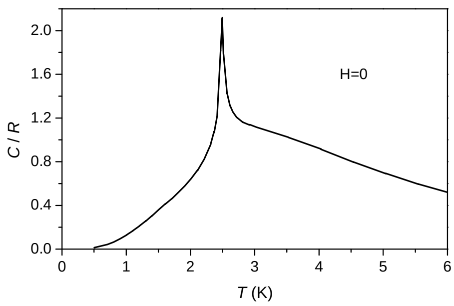

Figure 4. Temperature dependence of the molar specific heat in zero field.

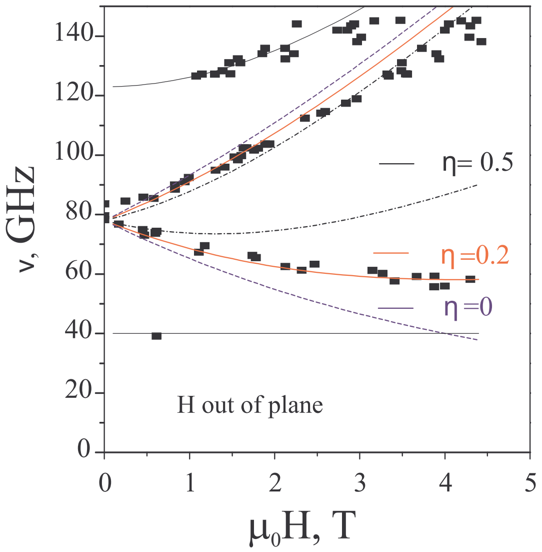

Figure 5. Frequency-field dependence for the electron spin resonance at T =1.4 K.

and $\lambda = 2.37$ Å for HB-1A and on the D23 lifting counter diffractometer at ILL (Grenoble)using $\lambda = 2.38$ Å neutrons. At low temperatures, two types of magnetic Bragg peaks were found, corresponding to the propagation vectors of the magnetic structure $(1/3-\zeta, 1/3-\zeta, 0)$, $\zeta = 0.038$, and $(1/2, 1/2, 0)$, respectively. Both types of reflections emerge at the same temperature $T_{\mathrm{N}} = 2.4$ K within the experimental accuracy. The temperature dependence of the corresponding Bragg peaks are shown in Fig. 3. An analysis of 19 inequivalent Bragg reflections in the $(h, k, 0)$ plane at $T = 1.5$ K revealed that the $(1/3-\zeta, 1/3-\zeta, 0)$ peaks can be entirely accounted for by a planar helimagnetic state, with spins rotating in the (a, b) plane. At the same time, the 14 inequivalent sets of $(1/2, 1/2, 0)$-type Bragg intensities measured in the $(h, k, 0)$ plane are consistent with a collinear antiferromagnetic spin arrangement, with spins in the (a, b) plane and forming a small angle of 15° with the a axis. Thus the diffraction data confirm the two-layer model depicted in Fig. 2. As discussed earlier in Ref. [9], the different spin arrangement in C- and S-layers can be accounted by the difference in the corresponding ratios $R = J_1/J_2$ vs. $R' = J_1'/J_2'$: theory predicts a switch from a helimagnetic to a collinear state at $R > 2$ [1, 2, 11]. The temperature dependence of the specific heat, measured by means of the Quantum Design PPMS at Warwick University is shown in Fig. 4. It demonstrates single sharp peak at the temperature $T_{\mathrm{N}} = 2.5$ K. The field-evolution of the observed structures and the entire phase diagramm are described in Ref. [10].

The low-temperature ESR spectrum taken in the frequency range 25-150 GHz at the magnetic field directed along the c-axis is shown on Figure 5. This orientation of the magnetic field is usefull because all crystallographic domains have the same resonance frequencies. The spectrum has four branches, two of them (the upper and the lower), where ascribed to the collinear structure in Ref.[9], on the base of the analysis of frequency-field dependences for different orientations of the magnetic field. Two branches with a common gap 80 GHz are described to the spiral structure, as it corresponds to a resonance spectrum of a planar noncollinear structure [5]:

$$\nu_{1,2} = \gamma\sqrt{\Delta^2 + (\tfrac{1+\eta}{2}H)^2} \pm \gamma\tfrac{1-\eta}{2}H$$

here γ is the magnetomechanical ratio, Δ is the energy gap and $\eta = (\chi_{\|}-\chi_{\perp})/\chi_{\perp}$ is parameter of the susceptibility anisotropy, indices $\|$ and $\perp$ mark susceptibilities parallel and perpendicular to the spiral plane. The third branch of the spiral structure should have zero frequency in assumption that the in-plane anisotropy does not give contribution to the energy of this mode oscillations. This assumption is in accordance with the coincidence of two branches in zero field. The results of the fitting of the resonance branches with the common gap are shown in Figure 5. From here we get the $\eta = 0.2 \pm 0.1$. Note that the measurement of the magnetization does not allow one to derive this parameter because magnetic moment consists of contributions of both S and C-type planes. Further, we can estimate the ratio of the exchange parameters in the S-planes, using the molecular field consideration of the spiral wavevector (see, e.g.,[1, 2], which gives $\cos(q) = -J_1/2J_2$. From the observed value of q we derive $J_1 = 0.56J_2$. The susceptibilities of the spiral structure may be than expressed in terms of exchange integrals, using the calculation of [11] where the incommensurate behavior of a system of ferromagnetic planes is considered. Incommensurate structures on the triangular lattice may be mapped to this problem, because there are ferromagnetic lines, perpendicular to the exchange bonds J_2. Using the calculation of [11] for perpendicular and parallel susceptibilities with the above ratio of exchange integrals we get $\eta = 0.195$. Thus, there is a good correspondence of the observed spiral modulation within the S-planes and the ESR spectrum.

In conclusion, it may be pointed out, that two coexisting antiferromagnetic structures with incommensurate wavevectors are found in $\mathrm{KFe(MoO_4)_2}$ below 2.5 K. Specific heat measurement fixes only one transition temperature. Magnetic resonance signals of the spiral subsystem is consistent with the wavevector observed in neutron scattering experiment.

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