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Spin-flop transition accompanied with changing the type of magnetic ordering

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ABSTRACT

We discuss theoretically rather rear example of spin-flop transition which is accompanied with changing the type of magnetic ordering and which seemingly has not been addressed yet. We demonstrate that changing the type of magnetic ordering can manifest itself in antiferromagnetic (AF) resonance experiments as an apparent peculiar switching of the anisotropy at the transition from the easy-axis type to the easy-plane one. We argue that this kind of spin-flop transition is observed recently by Povarov et al. (2013) [12] in $Cu(pz)_2(ClO_4)_2$ (where pz denotes pyrazine), one of the best realizations of spin-$\frac{1}{2}$ Heisenberg AFs on square lattice having a very small anisotropy. We show that the magnetic ordering changes at the spin-flop transition in this material in the direction perpendicular to AF square planes. We examine the microscopic mechanism of such behavior in $Cu(pz)_2(ClO_4)_2$ and find that dipolar forces and extremely small exchange coupling between spins from neighboring planes are responsible for it.

1. Introduction

Spin-$\frac{1}{2}$ Heisenberg antiferromagnet (AF) on square lattice has been one of the most extensively discussed models of quantum magnetism in recent three decades. Apart from its relevance to parent compounds of high temperature cuprate superconductors [1], it provides a convenient playground for investigation of novel types of many-body phenomena, among which are quantum spin-liquid and nematic phases, novel universality classes of phase transitions, and order-by-disorder phenomena. In the most simple variant of this model with exchange coupling between only nearest-neighbor spins, the Néel order arises at T=0 which is destroyed by thermal fluctuations at any finite T according to Mermin-Wagner theorem [2]. However all practical three-dimensional (3D) realizations of this model contain weak low-symmetry interactions of relativistic nature and exchange interaction between spins from different planes which lead to a finite transition temperature T_N to the Néel phase. In particular, the role is well known of unavoidable long-range dipolar interaction in stabilization of magnetically ordered phases in low-dimensional magnetic systems [3,4].

$Cu(pz)_2(ClO_4)_2$ (where pz denotes pyrazine) has been found recently to be one of the most perfect realizations of spin-$\frac{1}{2}$ Heisenberg AFs on square lattice with nearest-neighbor exchange coupling constant $J \approx 18.1$ K, $T_N \approx 4.21$ K, and a very small easy-plane anisotropy forcing magnetic moments to lie within the square planes [5–11]. Exchange coupling constant between spins from neighboring planes does not exceed 9 mK and dipolar forces are expected to play a significant role in the interlayer coupling [8]. Recent antiferromagnetic resonance (AFR) experiment [12] reveals also an in-plane easy-axis anisotropy (that is an order of magnitude smaller than the easy-plane one) and a related spin-flop transition at magnetic field $H = H_{sf} \approx 0.42$ T having anomalous properties. According to the common wisdom [13], one expects four possible scenarios summarized in Fig. 1 for AFR frequencies dependence on H in a two-sublattice AF in the field directed along easy/hard axis. However non of them is realized in $Cu(pz)_2(ClO_4)_2$ as it is seen in Fig. 2: $Cu(pz)_2(ClO_4)_2$ behaves as an easy-axis AF (see Fig. 1(a)) and as easy-plane AF (see Fig. 1(c)) at $H < H_{sf}$ and $H > H_{sf}$, respectively. Then, the experimentally obtained picture looks as if the in-plane anisotropy changes at the spin-flop transition from the easy-axis type to the easy-plane one. The origin of this peculiar behavior has not been clarified yet. As it is argued in Ref. [12], a magnetoelastic mechanism cannot be responsible for it.

In the present paper, we propose a microscopic model and describe quantitatively low-temperature experimental data reported before for $Cu(pz)_2(ClO_4)_2$. The model Hamiltonian we discuss contains Heisenberg spin coupling, anisotropic easy-plane exchange interaction, and unavoidable dipolar forces. In particular, we demonstrate in Section 2, where the classical ground state energy is analyzed, that the in-plane easy-axis anisotropy obtained in Ref. [12] originates from dipolar interaction and a small departure of the crystal structure from

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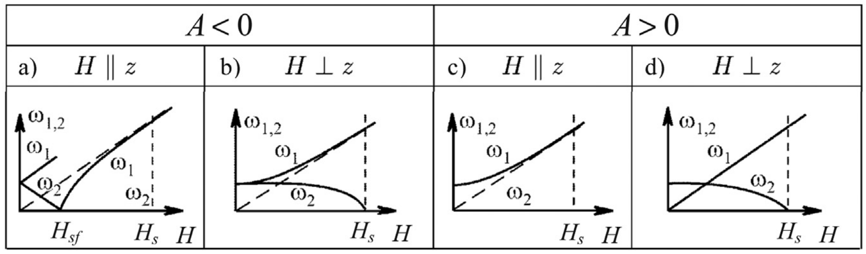

Fig. 1. Sketch of normal behavior of AFR frequencies $\omega_{1,2}$ in easy-axis ($A < 0$) and easy-plane ($A > 0$) two-sublattice antiferromagnets in magnetic field H, where A is the anisotropy value and z is easy/hard axis [13]. H_{sf} and H_s are spin-flop and saturation (spin-flip) fields, respectively.

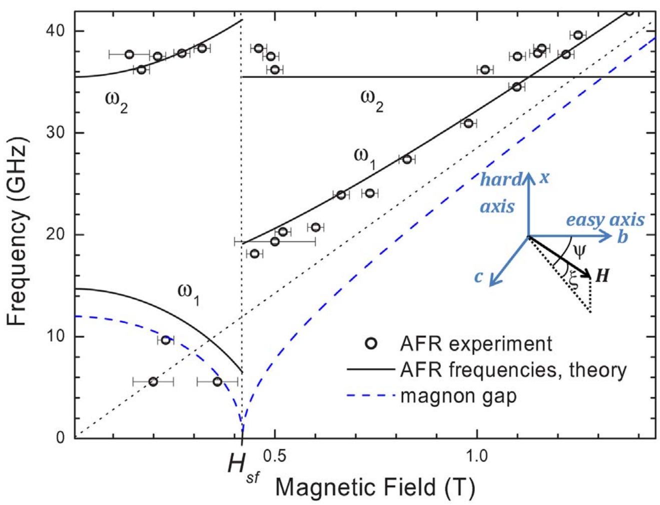

Fig. 2. (Color online.) AFR data obtained in Ref. [12] in $Cu(pz)_2(ClO_4)_2$ at T=1.3 K for magnetic field directed along the easy axis inside the square planes ($\psi = \xi = 0$ in the inset). It is seen from Fig. 1 that the system behaves according to Fig. 1(a) and (c) at $H < H_{sf}$ and $H > H_{sf}$, respectively. Solid lines are theoretical results obtained in the present paper using model 1 in the first order in $1/S$. Magnon gap is also shown which is derived theoretically in the present work.

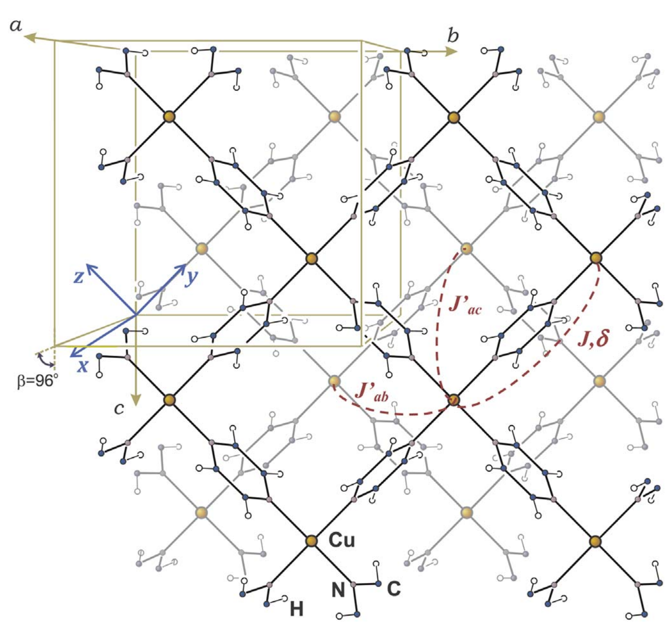

Fig. 3. (Color online.) $Cu(pz)_2(ClO_4)_2$ structure obtained in crystallographic measurements [9] (this figure is adopted from Ref. [12]). Two layers are shown each containing a square magnetic lattice. ClO_4 complexes are not shown for clarity. All symbols are faded out corresponding to the lower layer. Cartesian coordinate system xyz is also shown which is used in our consideration. Parameters of model (1) (exchange coupling constants J, J'_{ab}, J'_{ac}, and exchange anisotropy δ) are also shown.

the tetragonal one (in particular, from small deviation of the angle β from 90° depicted in Fig. 3). We show that dipolar forces lead to changing at the spin-flop transition of the magnetic ordering in the direction perpendicular to square planes if inter-plane exchange coupling is sufficiently small. This changing of the magnetic ordering takes place at $\xi < \xi_c$ and $\psi < \psi_c$, where ξ and ψ are angles determining the field orientation (see inset in Fig. 2). We find expressions for critical angles ξ_c and ψ_c.

Magnon spectrum, AFR frequencies and ground state energy are derived in Section 3 in the first order in $1/S$. We demonstrate that due to the changing of the magnetic ordering at the spin-flop transition, the lower AFR frequency does not coincide with the gap Δ in lower magnon branch (as usual, Δ vanishes at the spin-flop transition) and AFR frequencies behave in this model as those experimentally obtained in $Cu(pz)_2(ClO_4)_2$ (see Fig. 2). In Section 4, we use analytical expressions obtained to fit available low-temperature experimental data and to extract parameters of the microscopic model. In particular, we conclude that the inter-plane exchange coupling cannot exceed a value of the order of 10^{-3} K in $Cu(pz)_2(ClO_4)_2$. Then, it is the tiny inter-plane exchange coupling that makes possible the peculiar behavior of $Cu(pz)_2(ClO_4)_2$ in magnetic field which is governed by dipolar forces.

We summarize our results in Section 5. To provide an intuitively clear example of a system showing the anomalous spin-flop transition similar to that obtained in $Cu(pz)_2(ClO_4)_2$, we discuss also in Section 5 a phenomenological model of a layered two-sublattice AF having a hierarchy of small anisotropic spin interactions. These interactions lead to changing the type of magnetic ordering at the spin-flop transition in the direction perpendicular to AF planes. Then, the effect of long-range dipolar interaction in the microscopic model is simulated by the hierarchy of anisotropic short-range spin interactions in the phenomenological model.

Our consideration can be relevant also to members of a family of recently synthesized [8] two-dimensional (2D) spin-$\frac{1}{2}$ Heisenberg AFs to which $Cu(pz)_2(ClO_4)_2$ is a prototype.

2. Microscopic model for $Cu(pz)_2(ClO_4)_2$. Classical ground state analysis

In this section, we discuss classical ground-state properties of a model which we use in Section 4 to describe quantitatively low-temperature experimental data reported before for $Cu(pz)_2(ClO_4)_2$. The model Hamiltonian has the form

$$\mathcal{H} = \frac{1}{2}\sum_{l\neq m}(J_{lm}\delta_{\alpha\gamma} - Q_{lm}^{\alpha\gamma})S_l^\alpha S_m^\gamma - \delta\sum_{\langle l,m\rangle} S_l^x S_m^x - \mathbf{H}\sum_l \mathbf{S}_l, \tag{1}$$

where the summation over repeated Greek letters is implied, $\langle l, m\rangle$ denote nearest-neighbor couples of spins in square planes, $\delta > 0$ is the value of easy-plane anisotropy, x is the hard axis (see Fig. 3), the first term describes the short-range exchange and long-range dipolar interaction between spins, dipolar tensor Q has the form

$$Q_{lm}^{\alpha\gamma} = \frac{\omega_0}{4\pi}\frac{3R_{lm}^\alpha R_{lm}^\gamma - \delta_{\alpha\gamma}R_{lm}^2}{R_{lm}^5}, \tag{2}$$