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Spin nematics in frustrated magnets and their electron spin resonance

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Geometrically frustrated quantum magnets offer a center stage of the search for an exotic quantum state of matter. Recent theoretical researches [1, 2, 3] found that a class of systems—frustrated ferromagnets—that features both the nearest-neighbor ferromagnetic and competing further-neighbor antiferromagnetic interactions can show a spin nematic phase—a novel state of matter—at low temperatures. The spin nematic state is a type of spin liquid-crystal, wherein spins neither have any vector order nor break translational symmetry. However the spins have a directional order that partially breaks the spin-rotational symmetry. The directional order parameter is given by a rank-2 symmetric spin tensor, which has a close analogy to the nematic order parameter in liquid crystals. The presence of the spin nematic order was theoretically understood in the magnon picture near the saturation point in an applied magnetic field. As is well known, spin vectors in usual antiferromagnets have an antiferromagnetic order with the transverse spin components being perpendicular to the applied field, when they are not fully polarized. This canted antiferromagnetic order arises as a consequence of magnon condensation. However, in frustrated ferromagnets, magnons form stable two-magnon bound states known as bimagnons. The bimagnon formation prevents spin vector ordering and, furthermore, the condensation of bimagnons leads to nematic ordering in the spin transverse components [2].

In the first half of my talk, I would like to explain our recent theoretical study of a natural mineral volborthite $Cu_3V_2O_7(OH)_2 \cdot 2H_2O$, showing that volborthite is a plausible candidate for realization a spin nematic phase [4]. Volborthite contains a frustrated kagome plane formed by S=1/2 spins. Magnetization measurements showed a wide 1/3-plateau, which is much wider than expected in the pure kagome antiferromagnet. In addition, a novel spin-disordered phase was observed just below the 1/3 magnetization. Motivated by these experiments on volborthite single crystals, we performed microscopic modeling by means of density functional theory (DFT) with the single-crystal structural data as a starting point. Comparing with exact diagonalization simulations, we concluded that the microscopic magnetic model of volborthite contains four exchanges (antiferromagnetic J and J_2, as well as ferromagnetic J ' and J_1). See Fig. 1(a). Simulations of the derived spin Hamiltonian showed good agreement with the experimental magnetic susceptibility and magnetization process data. The 1/3-plateau phase pertains to polarized magnetic trimers formed by strong J bonds. An effective $J \to \infty$ model shows a tendency towards condensation of magnon bound states preceding the onset of the plateau phase, which leads to an emergence of a bond nematic phase.

Though some theoretical proposals have been presented for the family of frustrated ferromagnets, they have not been experimentally verified. This is partly because the spin nematic order parameter is not directly accessible by conventional measurement techniques. It has been discussed that dynamical observables can capture the characteristics of

quadrupolar ordering [5, 6]. In the second half of my talk I would like to explain how electron spin resonance (ESR) measurements enable us to identify spin nematic order [7]. We show that the frequency of the paramagnetic resonance peak of the ESR spectrum is shifted by an amount proportional to the ferroquadrupolar order parameter. In contrast, the antiferroquadrupolar order does not induce any frequency shift. Instead, the antiferroquadrupolar order yields a characteristic resonance peak in the ESR spectrum, which corresponds to a magnon-pair resonance. Reflecting the condensation of the bound magnon pair, the magnon-pair resonance frequency shows a singular upturn at the saturation field (See Fig. 2). Moreover, the amplitude of the magnon-pair resonance peak shows an interesting angular dependence. We confirmed these properties in the spin nematic phase by taking an example of an $S = 1$ bilinear-biquadratic model.

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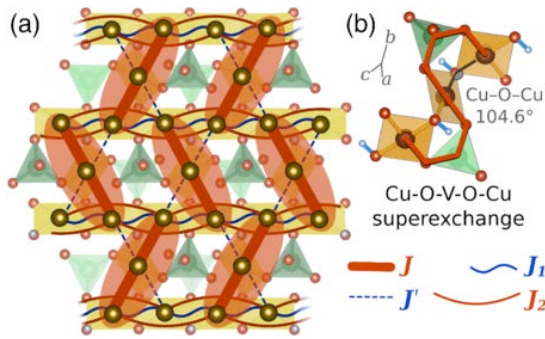

Fig. 1: Four relevant exchange couplings of volborthite: AF J and J_2, as well as FM J' and J_1. Magnetic trimers formed by J exchanges are highlighted.
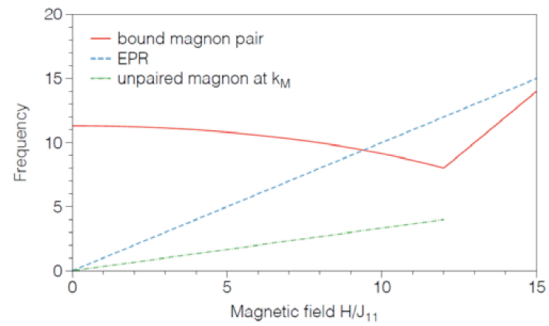
Fig. 2: Resonance frequencies of ESR in the antiferroquadrupoler phase ($J_{11}=1$). The saturation field is given by $H_S/J_{11}=12$. The magnon-pair resonance (the solid curve) and the unpaired-magnon resonance are found in addition to the electron paramagnetic resonance.

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