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Search for Novel Quantum States in Highly-Frustrated Magnets

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A simple Néel order can be destroyed by geometrical cancellation of magnetic interactions between nearest-neighbor spins on triangle-based networks or by competitions among two or more kinds of interactions. Stabilized alternatively are quantum disordered states called the spin liquid or unusual long-range orders having emergent degrees of freedom such as chirality or topological excitations. Extensive theoretical studies have proposed fascinating states of matter in various classes of models with frustration, and experimental studies have been undertaken to search for related exotic phenomena in real candidate compounds. However, there are numerous obstacles to clarify the frustration physics: the selection of a ground state from macroscopically large number of states existing next to it within a small energy range is subtle and is rather difficult to predict even by the state-of-the-art calculation techniques. Moreover, the ground state is often severely influenced by perturbations like anisotropy or additional interactions to the simplest Heisenberg model. Furthermore to experiments, the presence of crystallographic disorder, which does exist more or less in actual compounds, tends to disturb and mask the intrinsic properties of frustrated systems. Therefore, one has to be careful to enjoy the interesting frustration physics.

In my presentation, I will focus on some frustrated spin systems from materials point of view. A particular emphasis will be on two kagome-type copper minerals: volborthite [1] and Cd-kapellasite (CdK) [2]. The others are so-called J_1–J_2 magnets with first and second-neighbor interactions in 1D chains [$NaCuMoO_4(OH)$, 3] and 2D square lattices [$AMoOPO_4Cl$ (A = K, Rb), 4]. In addition, I would like to address our recent trials searching for novel magnets in 5*d* transition metal compounds.

Volborthite was seemingly considered as a distorted kagome antiferromagnet, but is now approximated as an effective spin-1/2 J_1–J_2 square-lattice magnet [1]. An extremely wide 1/3 magnetization plateau appears above 28 T and continues over 74 T at 1.4 K, in which a simple up-up-down spin configuration in a trimer may be stabilized. Interestingly, a novel phase called phase N appears between the SDW phase below 22 T and the 1/3 plateau phase. Moreover, very recent heat capacity and magnetocalorimetry measurements by Kohama et al. revealed the presence of another phase between the N and plateau phases. It seems that a series of unconventional magnetic states occur in volborthite under high magnetic fields.

CdK [$CdCu_3(OH)_6(NO_3)_2 \cdot H_2O$] is a structurally perfect kagome antiferromagnet crystallizing in the kapellasite-type [2]. An antiferromagnetic order accompanied by a small spontaneous magnetization that surprisingly is confined in the kagome plane sets in at $T_N \sim 4$ K, well below the nearest-neighbor exchange interaction $J/k_B = 45$ K. This suggests that a unique “q = 0” type 120º spin structure with “negative” (downward) vector chirality, which breaks the underlying threefold rotational symmetry of the kagome lattice and thus allows a spin canting within the plane, is exceptionally realized in this compound rather than a common one with “positive” (upward) vector chirality. The origin is discussed in terms of the Dzyaloshinskii-Moriya interaction. Very recent faraday-rotation measurements at ultrahigh fields up to 160 T by Okuma and Takeyama et al. found the whole magnetization curve of CdK, where a series of magnetization plateaux occur.

In a frustrated J_1–J_2 chain with the nearest-neighbor ferromagnetic interaction J_1 and the next-nearest-neighbor antiferromagnetic interaction J_1, novel magnetic states such as a spin-nematic state are theoretically expected. However, they have been rarely examined in experiments because of the difficulty in obtaining suitable model compounds. Nawa et al. show that the quasi-1D antiferromagnet $NaCuMoO_4(OH)$, which comprises edge-sharing CuO_2 chains, is a good candidate with J_1 = 51 K and J_2 = 36 K [3]. We are now looking for evidence of a spin nematic state expected just below the saturation field of 26 T. It is noted that there are some similarities between this compound and volborthite.

$AMoOPO_4Cl$ (A = K, Rb) with Mo^{5+} ions in the $4d^1$ electronic configuration are good model compounds for the spin-1/2 J_1–J_2 square-lattice magnet [4]. Magnetic transitions are observed at around 6 and 8 K in the K and Rb compounds, respectively. In contrast to the normal Néel-type antiferromagnetic order, the NMR and neutron diffraction experiments find a columnar antiferromagnetic order for each compound, which is stabilized by a dominant antiferromagnetic J_2. Both compounds realize the unusual case of two interpenetrating J_2 square lattices weakly coupled to each other by J_1.

A new oxychloride $Ca_3ReO_5Cl_2$ was recently discovered by Hirai et al. [5]. This compound shows unusually distinct pleochroism; that is, the material exhibits different colors depending on viewing directions. It is a consequence of the fact that a complex crystal field splitting of the 5*d* orbitals of the Re^{6+} ion in a square-pyramidal coordination of low-symmetry occurs accidentally in the energy range of the visible light spectrum. Thanks to the specific arrangement of the lowest d_{xy} orbitals in an anisotropic triangular lattice, this $5d^1$ antiferromagnet behaves as a unique 1D magnet with strong couplings.

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