Pinwheel valence-bond solid in the deformed kagome compound Rb2Cu3SnF12

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Determining ground states of the s=1/2 quantum kagome antiferromagnet is a long-standing unsolved issue. Theoretically suggested are several intriguing ground states, such as the resonating-valence-bond (RVB) or its solidified version, valence-bond-solid state. However, lack of good model materials prohibits the experimental verifications of such intriguing ground states. Recently, Tanaka and co-workers at Tokyo Tech. have successfully grown large single crystals of the Rb2Cu3SnF12 compound [1]. In this compound the quantum s=1/2 spins of the Cu3+ ions form a slightly-deformed two-dimensional (2D) kagome lattice with dominant antiferromagnetic exchange interactions. Although the slight deformation from the ideal kagome lattice complicates the situation, this model system provides the first experimental opportunity to study the ground state and low-energy excitations in the quantum kagome antiferromagnet by single crystal neutron scattering.

Large single crystals of Rb2Cu3SnF12 were ground by a method described in Ref. [1]. Neutron scattering experiments were performed using the thermal and cold neutron triple-axis spectrometers GPTAS and HER, both installed at JRR-3, Tokai, Japan. Several spectrometer conditions were chosen depending on the contradicting necessity of the intensity and Q- or E-resolution. The sample was set either in the 4K GM refrigerator or the vertical field superconducting magnet with mostly the c-aixs being vertical to the scattering plane.

First, the magnetic excitations in the zero external field were investigated. We found that the two excitation peaks were observed in an energy range up to 15 meV; one peak is at 2 meV, and the other at 7 meV, at the antiferromagnetic zone center. These peaks appear below 30 K, and thus are surely magnetic. The excitations are highly dispersive in the 2D plane, merging into single peak at 5 meV at the M and K points, whereas it hardly shows the dispersion along the c-axis. The lower energy peak splits into two separate peaks at high magnetic field with the splitting being linearly dependent on H. On the other hand, the higher energy peak shows neither the splitting nor peak shift under the external field up to 5 T. From these observations we have confirmed that they are excitations from the singlet ground state to the $|Sz| = 1$ states (lower energy), and to the $|Sz|=0$ state (higher energy) of the triplet levels. These splitting of the $|Sz|=1$ and $|Sz|=0$ states clearly indicate the significance of the Dzyaloshinskii-Moriya interactions between the Cu3+ ions. The experimental results, as well as theoretical calculations, strongly support the formation of the pinwheel VBS state as the ground state in this deformed kagome compound, clearly observed for the first time [2].

[1] K. Morita et al., J. Phys. Soc. Jpn. 77 (2008) 043707.
[2] K. Matan et al., Nature Phys. 6 (2010) 865-869.