Successive phase transitions in a magnetic semiconductor $Yb_2Cu_{2n}S_{n+3}$ with a deficient Yb triangle lattice

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In rare-earth insulating or semiconducting compounds, a bond-dependent super-exchange interaction, mediated by nonmagnetic anions between the magnetic cations, plays the leading role in forming a magnetic ordered ground state. When the magnetic cations are located in a geometrically frustrated configuration, the magnetic order is suppressed, and the quantum fluctuations are enhanced.

Magnetic semiconductor $Yb_2Cu_{2n}S_{n+3}$ (n = 3.0 and 3.6) crystallizes in a trigonal $ErCu_3Si_3$-type crystal structure with the space group of *P–31c* [1]. Since the Yb ions form a deficient triangle lattice, the geometrical frustration effect must be suppressed to induce an ordered state. The Curie–Weiss behavior of the magnetic susceptibility between 100 and 300 K indicates the trivalent Yb states, and the negative value of the paramagnetic Curie temperature of θ_p indicates antiferromagnetic interaction. The specific heat divided by temperature, C/T, for n = 3.0, exhibits a cusp-type peak at T_1 = 4.2 K. For n = 3.6, C/T shows a peak at T_1 and a sharp peak at T_2 = 2.3 K. The magnetic entropy S_m at T_1 is only 15% of Rln2. The reduced S_m at T_1 indicates strong fluctuations of the effective spin-1/2 in the triangular lattice.

In the present work, we conducted powder neutron diffraction experiments with samples of n = 3.0 and 3.6, performed with the high-efficiency and resolution powder diffractometer (HERMES) installed at JRR-3M in the Japan Atomic Energy Agency. The powdered samples were sealed in a cylindrical vanadium capsule and cooled by a 1 K refrigerator down to the lowest temperature of 0.7 K.

Figure 1 shows the powder neutron diffraction patterns of $Yb_2Cu_{2n}S_{n+3}$ for (a) n = 3.0 and (b) n = 3.6. The patterns were measured at two different temperatures, 0.7 K (shown in blue) and 6.0 K (red), respectively below and above the transition temperatures of T_1 and T_2. The patterns of 0.7 K (blue) are vertically shifted by 500 cnts. The differences between the patterns measured at 0.7 and 6.0 K are depicted by the (green) solid lines with offsets. For n = 3.0, we observed no magnetic reflections for $T < T_1$. On the other hand, for n = 3.6 in Fig. 1(b), a small super-lattice reflection was observed at around 35 deg. The inset shows the temperature variation of the integrated intensity at 0.7 K within the angle range of $33 \leq 2\theta \leq 38$ deg. The intensity increases on cooling for $T < T_2$. This is evidence of an antiferromagnetic transition with reduced Yb moments.

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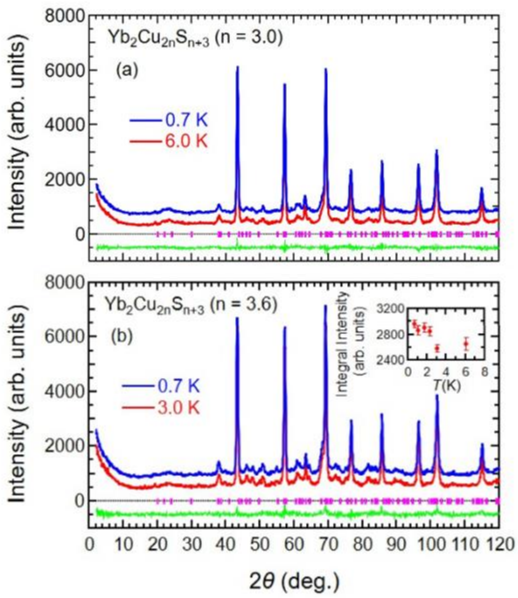

Fig. 1. Powder neutron diffraction patterns of $Yb_2Cu_{2n}S_{n+3}$ for (a) n = 3.0 and (b) n = 3.6.