Long-Time Variation of Magnetic Structure in Multistep Metamagnet Ca3Co2O6

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Recently, a long-time variation in magnetic structure has been observed in the nondiluted magnet CeIr3Si2. This is the first example of a uniform magnet showing long-time variation of magnetic structure [1]. It exhibits successive magnetic transitions and multistep metamagnetic transitions at low magnetic field. These results indicate the presence of frustrated magnetic interactions and suggest that the long-time variation of magnetic structure is caused by frustrated magnetic interactions. We examined other materials that show these magnetic transitions. We have found a long-time variation in magnetic structure in a geometrically frustrated magnet Ca3Co2O6 [2].

Ca3Co2O6 crystallizes in a hexagonal structure. It consists of Co chains along the c-axis and these chains are arranged in a triangular lattice in the c-plane. This compound shows two magnetic transitions at Tc1=25 K and Tc2=13 K and 5-step metamagnetic transitions below 7 T at 1.9 K.

Neutron scattering experiments were carried out using triple-axis spectrometers 4G, 5G, and T1-1 installed at the JRR-3M reactor of JAEA in Tokai. Magnetic scattering peaks were observed at incommensurate positions (1, 0, $\pm\delta$). We found that the magnetic Bragg peaks observed at (1, 0, $\pm\delta$) immediately after cooled below Tc1 moved towards δ=0 with time. Figure 1 shows the time variation of δ measured at representative temperatures. These δ-t curves can be represented by the function $\delta = a + b\exp(-t/\tau)$, where τ is the characteristic time. Temperature variation of τ follows the Arrhenius law and the activation energy Ea has been determined as Ea/kB=57 K.

We also measured the intensities of many magnetic and nuclear Bragg reflections at 10.5 K and determined the magnetic structure of Ca3Co2O6. To avoid the influence of the time variation of magnetic structure, measurements were started 5 hours after the sample was cooled to 10.5 K. In this model, the magnetic moments form a series +++...+++/- - -...- - -/+++...+++ along a Co chain where + and - mean the moment direction parallel and antiparallel to the c direction. On the basis of this magnetic structure model, the shift of the peak position with time corresponds to the increase of the ferromagnetically coupled length of the Co chain.

We stress that the time variation behavior in Ca3Co2O6 is different from that in other materials. Ca3Co2O6 shows a continuous change in the period of the magnetic structure. On the other hand, CeIr3Si2, PrCo2Si2, and TbNi2Si2 show time variation of the volume fractions of their two distinct magnetic structures [1, 3]. Therefore, the time variation observed in Ca3Co2O6 is a new type.

[1] K. Motoya et al., J. Phys. Conf. Series 200 (2010) 032048.
[2] T. Moyoshi and K. Motoya, J. Phys. Soc. Jpn. 80 (2011) 034701.
[3] K. Motoya et al., J. Phys. Conf. Series 273 (2011) 012124.

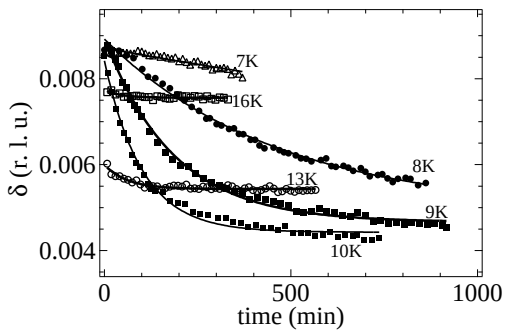

Fig. 1. Time variation of the magnetic peak position, δ.