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Low Temperature Magnetic Phase of Triangular Lattice Antiferromagnet $Ca_3Co_2O_6$

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In the oxide $Ca_3Co_2O_6$, the spin system consists of ferromagnetic Ising-spin chains which form the triangular lattice on the hexagonal c plane.[1-3] Because of the antiferromagnetic interchain interaction, it exhibits a ferrimagnetic ordering of the chains below 25 K, which has been confirmed by a 1/3 plateau in the magnetization curves[1-3] and also by neutron diffraction peaks.[3] Moreover, the Mössbauer effect of $Ca_3(Co_{0.99}{}^{57}Fe_{0.01})_2O_6$ has revealed that the relaxation of the Fe^{3+} spins is extremely slow due to the large difference between magnetic characters of the host (Ising) and impurity (Heisenberg) spins.[4]

Although magnetic properties of $Ca_3Co_2O_6$ have been thus extensively investigated from the viewpoint of the Ising spin magnetism and the geometric frustration on the triangular net, the low temperature magnetic phase especially below 15 K is still an open question; there is a broad maximum around 15 K in the temperature dependence of the neutron diffraction intensity of the magnetic (100) reflection indicating a ferrimagnetic ordering reported by Aasland *et al.*[3] Since this measurement was carried out only above 10 K, the magnetic structure at lower temperature is still not clear. Additionally, a 1/3 plateau of the saturation moment in the M vs. H curves disappears for $T \leq 5$ K.[1,2] Instead, multisteps accompanied with the large hysteresis have been observed.[2] These facts suggest the presence of other magnetic structures at lower temperatures. In this note, we will present the results of powder neutron diffraction measured as a function of temperature and magnetic field.

The sample of $Ca_3Co_2O_6$ was prepared by the conventional solid state reaction method as described in ref. 1. Crystalline powders of $Ca_3Co_2O_6$ were filled into a cylinder of aluminum thin foil, about 25 mm in diameter and about 30 mm long. The neutron diffraction measurements were carried out with a neutron diffractometer installed at the Kyoto University Reactor (KUR-ND) in the temperature range 3 K~30 K and in the magnetic field range 0 T~5.0 T. The monochromatic neutron used has the wave length of 1.006 Å with the second order contamination below 0.2%. The behavior of the (100) magnetic reflection should strongly reflect magnetic properties of the low temperature phase, so that only the field and temperature dependence of this reflection was measured.

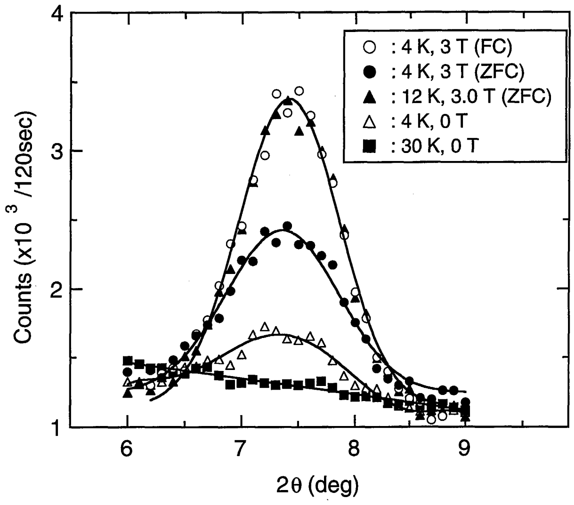

Fig. 1. The neutron diffraction profiles of the magnetic (100) reflection measured under various conditions. Solid curves are guide to the eye.

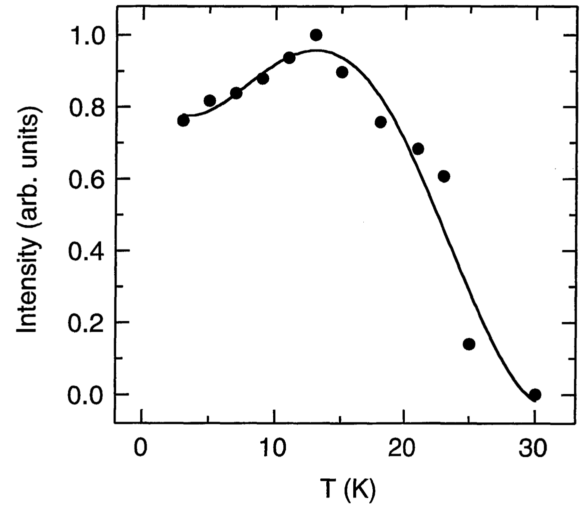

Fig. 2. Temperature dependence of the integrated intensities of the (100) reflection, I_{int} in the absence of magnetic field. The solid curve is just guide to the eye.

Figure 1 demonstrates the representative neutron diffraction patterns of the magnetic (100) reflection, clearly showing that the peak intensity depends on the measured conditions, i.e., temperature, magnetic field and cooling process (field cooled, FC; zero-field cooled, ZFC). The temperature variation of the integrated intensity of the (100) reflection I_{int} in the absence of magnetic field is shown in Fig. 2. The sharp rise in I_{int} at 25 K (onset of the ferrimagnetic ordering) and the maximum of I_{int} around 13 K are almost in agreement with the result of ref. 3. It is worth noting that I_{int}, though it gradually decreases below 13 K, is still non-zero at 3 K, indicating that there is no breakdown of ferrimagnetic correlation down to 3 K in contrast with the spin-glass like feature

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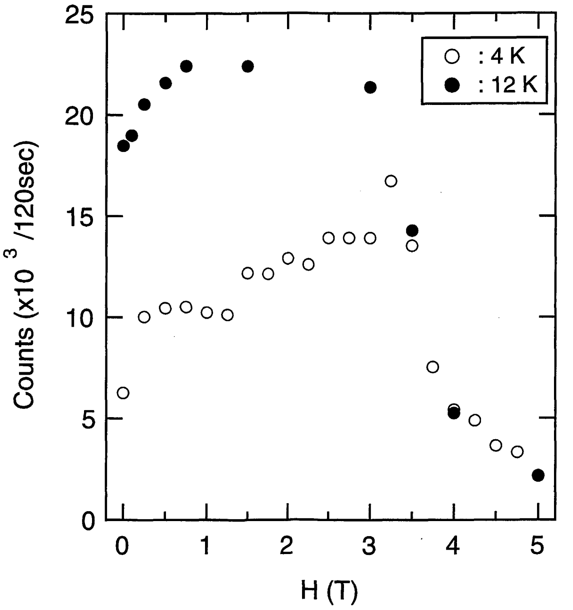

Fig. 3. Magnetic field dependence of the integrated intensities of the (100) reflection, $I_{\rm int}$ obtained at 4 K (open) and at 12 K (solid).

in the macroscopic magnetization measurements. This fact strongly suggests that the ferrimagnetic domain decreases in size below 13 K probably because the interaction which makes the ferrimagnetic phase unstable, say the third or forth next nearest interaction, becomes effective at lower temperatures.

We now discuss the magnetic field dependence of $I_{\rm int}$ measured at 4 K and 12 K (Fig. 3). In a given field below 3 T, $I_{\rm int}$ at 12 K is relatively larger than that at 4 K. At low fields, $I_{\rm int}$ increases with magnetic field, suggesting that, when magnetic field is applied, favorably oriented domains grow in size. However, it should be admitted that the applied field might align the powdered sample along the c axis to some extent, which also causes the increase in $I_{\rm int}$. The abrupt decrease in $I_{\rm int}$ above 3 T for both temperatures is due to the transition from ferrimagnetic to ferromagnetic structure. As seen in Fig. 1, the sample measured at 4 K under the field of 3 T in the FC process has much larger intensity than that in the ZFC case, which can be also attributed to the domain size effect; When the sample is cooled in the presence of external field, it helps the domains to grow.

In summary, our neutron diffraction study reveals an existence of long range ferrimagnetic correlation even below 10 K. $I_{\rm int}$ increases with H up to 3 T, while it decreases abruptly above the field. Furthermore, $I_{\rm int}$ measured in the FC process is much larger than in the ZFC case. These results can be interpreted in terms of the domain size effect of the ferrimagnetic phase.

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