Magnetic Structure and the Anomalous Hall Effect of $Cu_{1-x}Zn_xCr_2Se_4$

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Abstract

To study what roles spin structures of ferromagnets play in determining their Hall resistivity ρ_H or to see if the spin chiral order really contributes to the unusual behavior of anomalous Hall resistivity, we have carried out transport and neutron diffraction studies on the spinel type system $Cu_{1-x}Zn_xCr_2Se_4$, which is in a collinear ferromagnet at $x \sim 0$ and in the helimagnetic state at $x = 1.0$. It is established that the sign change of ρ_H takes place, with decreasing temperature, along with the appearance of the conical spin structure in the region $0 < x < 1.0$.

Key words: anomalous Hall effect; spinel type system $Cu_{1-x}Zn_xCr_2Se_4$; conical spin structure;spin chiral order

The anomalous Hall effect of ferromagnets is one of interesting problems, which seem not to be fully understood. In refs. [1] and [2], we have reported that the Hall resistivity ρ_H observed for the system $Nd_2Mo_2O_7$ cannot be described by the equation $\rho_H = R_0H + 4\pi R_s M$ usually used for ordinary ferromagnets, where H and M are the applied magnetic field and the uniform magnetization, respectively, and R_0 and R_s are the ordinary and anomalous Hall coefficients, respectively, Instead, an equation $\rho_H = R_0H + 4\pi R_s M_{Mo} + 4\pi R'_s M_{Nd}$, which has two individual contributions from the Mo- and Nd-moments, M_{Mo}, M_{Nd}, respectively, can describe the T- and H-dependences of ρ_H. Although the equation seems to be a simple extension of the ordinary equation, the T-dependences of the anomalous Hall coefficients R_s and R'_s obtained for the present system cannot still be understood. Then, a question arises how this unusual behavior of ρ_H is related with its non-collinear magnetic structure reported by the present authors' group [3]. More generally, it is interesting to study effects of the ordering of the spin chirality χ on the behavior of ρ_H, the possibility of which has been pointed out by Ohgushi *et al.* [4].

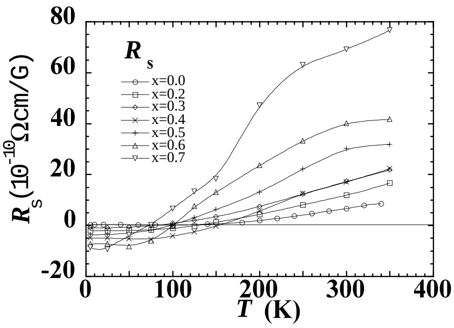

Fig. 1. The anomalous Hall coefficients R_s, are shown. They are obtained by the fittings H-dependence of ρ_H by using the equation $\rho_H = R_0H + 4\pi R_s M$

In the present work, we have adopted the spinel type ferromagnet $Cu_{1-x}Zn_xCr_2Se_4$ as a candidate, in which the spin structure seems to be non-trivial and the magnetic behavior is probably simpler than that of $Nd_2Mo_2O_7$ in the sense that it has only one magnetic elements. For this system, we have studied the transport and magnetic properties. We have also stud-

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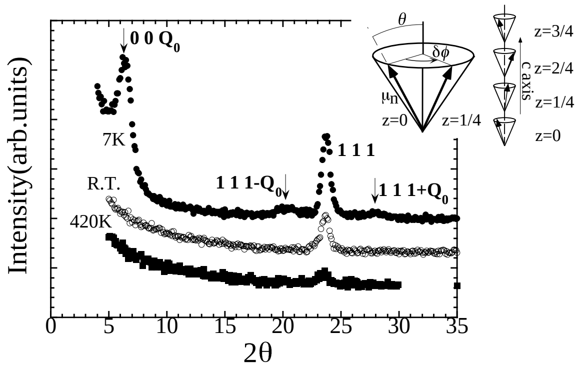

Fig. 2. Neutron diffraction patterns of $Cu_{0.5}Zn_{0.5}Cr_2Se_4$ taken at 7 K, room temperature and 420 K. The arrows show the incommensurate magnetic superlattice reflections which indicate the conical magnetic structure.

ied the magnetic structures by neutron diffraction. At $x = 0$, it is a collinear ferromagnet with Curie temperature $T_C = 450$ K and at $x = 1.0$, it has the helical structure with the Neel temperature $T_N = 20$ K, and from measurements of the M-H curves, the conical structure is expected for the intermediate x region below the characteristic temperature $T_m(\sim 150$ K), at which the slope -dM/dT begins to increase gradually with decreasing T. For $x = 0.5$, we have reported the anomalous sign change of ρ_H at around $\sim T_m$.

Polycrystalline samples which are confirmed to be the single phase by X-ray diffraction are used. The Hall resistivity ρ_H was measured by the four probe method. Neutron diffraction studies have been carried out at JRR-3M of JAERI for three samples with $x =$ 0.3, 0.5 and 0.7. Other details of the sample characterizations and physical measurements are described elsewhere [5,6].

In the analyses of the T- and H-dependences of ρ_H, we have used the equation $\rho_H = R_0 H + 4\pi R_s M$ and the R_s values obtained here are shown in Fig. 1. R_s of the sample with $x = 0$, which has collinear magnetic structure approaches zero as in the case of ordinary ferromagnets, while for the other samples, the sign change takes place at around $T_m(\sim 150$ K), suggesting the possible effects of the non-trivial magnetic structure to the behavior of ρ_H.

In Fig. 2, the neutron diffraction patterns taken at 420 K($T > T_C \sim 390$ K), at room temperature and at 7 K are shown for $x = 0.5$, for example. (The wavelength $\lambda = 2.353$ Å.) At 7 K, additional reflections are observed at the angles which correspond to the reciprocal points, (0,0,Q_0), (1,1,1-Q_0) and (1,1,1+Q_0), with the incommensurate vector $Q_0 \sim 0.467$ in the unit of reciprocal lattice a^*. This indicates that the magnetic structure is non-trivial. The peak intensity of the superlattice reflection at (0,0,Q_0) gradually increases with decreasing T below 150 K. This gradual growth of the superlattice peak or the non-trivial magnetic structure seems to be related to the gradual increase of -dM/dT and the anomalous sig change at around T_m with decreasing T. To reproduce the neutron diffraction patterns observed at low temperatures, we consider that the magnetic structure of $Cu_{1-x}Zn_xCr_2Se_4$ changes from the collinear to the helical one with the modulation vector along the [001] direction, as x changes from 0 to 1 [7], and adopt a model that the structures of the present samples are conical with the modulation vector along [001] and with the magnetic moments within a z-plane directing the same direction (See inset of Fig. 2 for the details, where θ and $\delta\phi$ are the tilting angle of the moments from the z axis and the difference of azimuthal angles between the neighboring z-planes, respectively.). Then, by fitting the calculated intensities to the experimentally obtained integrated intensities of the magnetic reflections, we have obtained the values of the fitting parameters of θ, $\delta\phi$ and the absolute value of the ordered moment $\boldsymbol{\mu}_n$ of Cr atom. The results are as follows. $\theta = 14.5°$, $\delta\phi = 50.3°$, $\mu_n = 2.6\mu_B$, for $x = 0.3$, $\theta = 22°$, $\delta\phi = 42.1°$, $\mu_n = 2.49\mu_B$, for $x =$ 0.5, and $\theta = 32°$, $\delta\phi = 39.6°$, $\mu_n = 2.57\mu_B$, for $x = 0.7$.

One of the interesting thing is the sign change of ρ_H takes place along with the appearance of the conical structure confirmed here by the neutron diffraction. Because the present samples are in the polycrystalline form, we cannot directly argue the effects of the non-trivial structure. However, it is interesting to mention that for the larger value of θ, the larger absolute value of R_s at $T \to 0$ was observed, which may indicate that the spin chirality locally defined by $\boldsymbol{S}_1 \cdot \boldsymbol{S}_2 \times \boldsymbol{S}_3$ for three spins $\boldsymbol{S}_1$, $\boldsymbol{S}_2$ and $\boldsymbol{S}_3$, may contribute in the present polycrystalline samples. To clarify the relationship between ρ_H and the non-trivial spin structure, we may have to use single crystals and study the detailed behavior of ρ_H in various the magnetic field direction.

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