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Small-Angle Neutron Scattering Study on Giant Magnetoresistance Effect of Sr_2FeMoO_6

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In order to investigate the origin of giant magnetoresistance (GMR) of a Sr_2FeMoO_6 layered perovskite, small-angle neutron scattering (SANS) experiments are carried out under magnetic fields. We measure magnetic scattering within a temperature range where the GMR effect appears conspicuously. Our model calculation reveales that the magnetic scattering is due to mesoscopic ferrimagnetic clusters with the averaged radius of 20 $\sim$ 30 nm. This magnetic scattering drastically decreases with applying the magnetic field of 1 T enough to lead to a large drop of resistivity. This result indicates that a lot of mesoscopic ferrimagnetic clusters fuse under the magnetic field. Therefore, we conclude that the GMR effect of Sr_2FeMoO_6 is dominated by spin-dependent scattering of conduction electrons from the ferrimagnetic clusters.

KEYWORDS: giant magnetoresistance, SANS experiments, mesoscopic ferrimagnetic cluster

§1. Introduction

There has been a growing interest in negative magnetoresistance (MR) from fundamental and applicational points of view. Hole-doped manganese perovskites $R_{1-x}A_xMnO_3$ (R : rare earth and A : divalent alkali) have been intensively studied in recent years due to the discovery of colossal magnetoresistance (CMR).[1–4] The negative MR ratio of these materials expressed as $\Delta\rho = |\rho(H) - \rho(0)|/\rho(0)$ is roughly in the order of 10^{-3} in a narrow temperature range close to the Curie temperature. It has been thought that the metal-insulator transition due to charge ordering or double exchange mechanism with strong on-site Hund's coupling is a key issue of the CMR effect of the materials.[5] However, it has also been reported that the CMR effect is dominated by magnetic or crystallographic structures in mesoscopic scale such as intrinsic two-phase coexistence.[6,7]

Much interest has been devoted to the similar materials with perovskite structures since the discovery of the CMR in hole-doped manganese perovskites. A new GMR compound Sr_2FeMoO_6 is composed of ordered perovskites forming alternating Sr-O layer and transition-metal-O layer where Fe^{+3} and Mo^{+5} ions order alternatively.[8,9] In this compound the Fe spin ($S = 5/2$) aligns antiparallel to the Mo spin ($S = 1/2$). The GMR effect with $\Delta\rho$ of several % is observed for a single crystal in a wide temperature range below the ferrimagnetic transition temperature T_N of 410 $\sim$ 450 K.

In this paper, we report mesoscopic structures of a Sr_2FeMoO_6 single crystal studied by small-angle neutron scattering (SANS) to clarify the origin of the GMR effect of this compound. Magnetic diffuse scattering obtained under magnetic fields can be analyzed by a model of ferrimagnetic clusters with finite boundary in thickness. We will discuss the GMR effect of this compound in comparison with the CMR effect of the manganese system under the present results.

§2. Experimental

A single crystal of Sr_2FeMoO_6 was grown by a floating zone-melting method. The detail of the sample preparation has been described elsewhere.[8] Magnetoresistivity measurements have been performed under magnetic fields up to $H = 7$ T by a conventional four-probe method. SANS experiments were performed on the instrument SANS-J of the research reactor JRR-3M installed at the Japan Atomic Energy Research Institute (JAERI). The wavelength of incident neutrons was 0.65 or 2.00 nm. Azimuthally averaged scattering intensities were transferred to the scattering cross-sections per unit volume $d\Sigma/d\Omega(q)$, where q is a momentum transfer. Magnetic fields up to $H = 1$ T were applied perpendicularly to the neutron beam.

§3. Results and Discussions

Figure 1 and its inset show the SANS cross-sections and resistivity of Sr_2FeMoO_6, respectively. The inset reveals the temperature dependences of the resistivity $\rho(T)$. In no magnetic field, the resistivity curve exhibits a metallic behavior and a residual resistance of 2.7×10^{-4} Ω·cm remains at $T = 4$ K. At $H = 5$ T the GMR effect becomes larger with decreasing the temperature and becomes $\Delta\rho \sim 6$ % at $T = 4$ K. It is noted that this temperature dependence of the GMR effect is different from that of the CMR effect for the manganese system, which appears only in a narrow temperature range close to the Curie temperature. These SANS cross-sections were obtained at $T = 10$ K where the GMR effect becomes more conspicuous. The scattering cross-sections can be de-

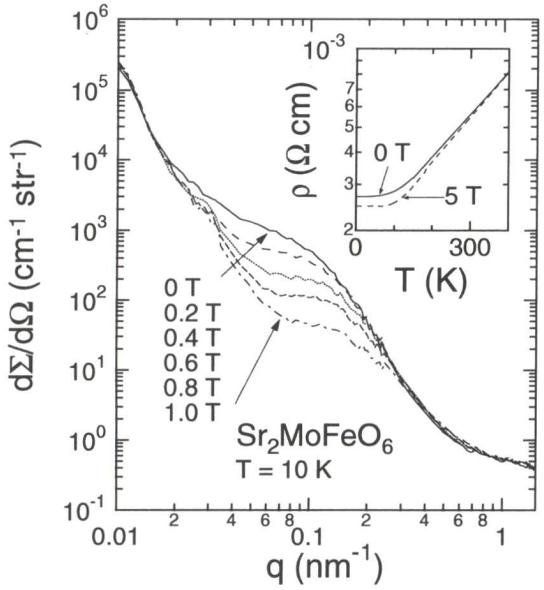

Fig. 1. SANS cross-sections of a Sr_2FeMoO_6 single crystal under various magnetic fields at $T = 10$ K. The inset shows temperature dependences of the resistivity at $H = 0$ and 5 T.

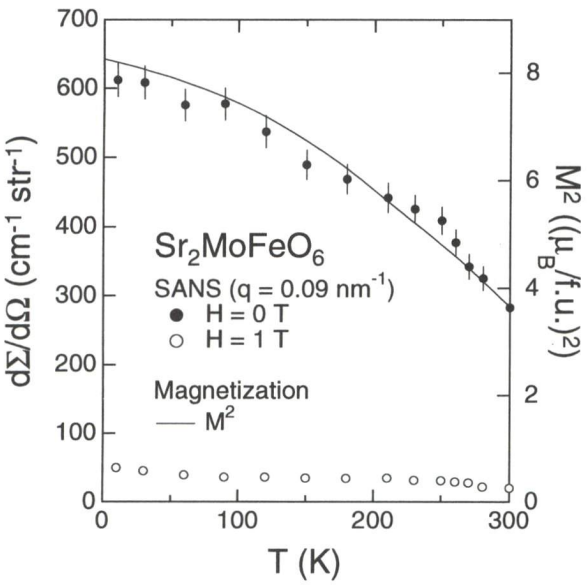

Fig. 2. Temperature dependences of the diffuse scattering cross-sections of Sr_2FeMoO_6 at $q = 0.09$ nm^{-1}. The solid line is the square of magnetization measured by a DC magnetometer.

scribed by the superposition of two scattering components. One is an strong nuclear scattering which shows the q^{-4} dependence (Porod's law) below $q = 0.03$ nm^{-1}. This q dependence indicates the existence of crystallographic microstructures with a sharp boundary such as crystal grains, which sizes are estimated from the inverse of a minmum of q and are then larger than 100 nm. The other is a diffuse scattering which rapidly decreases as the magnetic field increases.

Figure 2 shows the temperature dependences of the diffuse scattering cross-sections at $q = 0.09$ nm^{-1}. In no magnetic field, the temperature dependence of the scattering cross-section is in good agreement with that of the square of magnetization measured by a DC magnetometer. This scattering cross-section is almost suppressed to a background scattering level at $H = 1$ T below T_N. These results indicate that the observed temperature- and field-dependent diffuse scattering is caused by the magnetic scattering from a ferrimagnetic ordering of Fe and Mo spins.

In order to evaluate the magnetic scattering cross-section in the whole q range, we estimate the nuclear scattering cross-section by the extrapolation of the scattering cross-section measured at $T = 300$ K and $H = 1$ T. We then subtracted this extrapolatd nuclear scattering cross-section from each scattering cross-section. The obtained magnetic scattering cross-sections at $T = 10$ K are shown in Fig. 3. The profiles of the scattering cross-sections indicate the existence of mesoscopic magnetic clusters. The asymptotic behavior of the profiles at the higher q region reveals the deviation from the q^{-4} dependence (Porod's law). This deviation implies that the surface boundary of the magnetic clusters has finite thickness. At the boundary the magnetic scattering length-density gradually varies.

We then analyzed these profiles by a model of magnetic clusters as follows:

$$\frac{d\Sigma}{d\Omega}(q) = n\rho_m^2 exp(-\sigma^2 q^2) \times (\frac{4\pi}{q^3})^2 [sin(qR) - qRcos(qR)]^2,$$

where n is a number density of the magnetic clusters, ρ_m is a magnetic scattering length per unit volume, R is an averaged radius of the clusters, and σ denotes the thickness of the surface boundary of the clusters. The solid line in Fig. 3 is the curve fitted to the scattering cross-section at $H = 0$ T. From this fitting we obtained that $R = 28.0$ nm and $\sigma = 2.2$ nm.

Figure 4 shows the field dependence of n and R obtained by the curve fitting at $T = 10$ K. n and R are parameters of the small clusters remaining even under the magnetic field, since in the magnetic field SANS detects a contrast between local moments of the magnetic cluster and the magnetization averaged over the sample. n monotonously decreases from 2.5×10^{15} cm^{-3} to 0.69×10^{15} cm^{-3} with increasing the magnetic field up to $H = 1$ T. R also decreases from 28.0 nm at $H = 0$ T to 10.0 nm at $H = 1$ T. These results can be understood as follows. Most of the magnetic clusters oriented disorderly at $H = 0$ T grow into large domains by the magnetic field of $H = 1$ T. The large domain sizes becomes out of the q-range with increasing the field. However, some parts of clusters still keeps their size in a scale of 10^1 nm even at $H = 1$ T. On the other hand the temperature dependence of R slightly decreases from 28.0 nm at $T = 10$ K to 19.1 nm at $T = 300$ K.

In comparison between the resistivity and SANS measurements, it is noted that the appearance of ferrimagnetic mesoscopic clusters with a radius of several tens nm leads to the large resistivity. A magnetic field contributes to the growth of mesoscopic clusters into large domains, resulting in the suppression of the resistivity. It is a

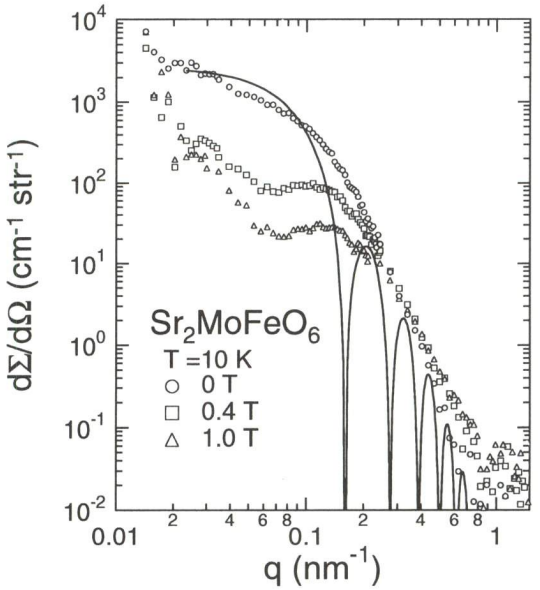

Fig. 3. Magnetic scattering cross-sections at $T = 10$ K. The solid line is a curve fitted by a mesoscopic magnetic cluster model.

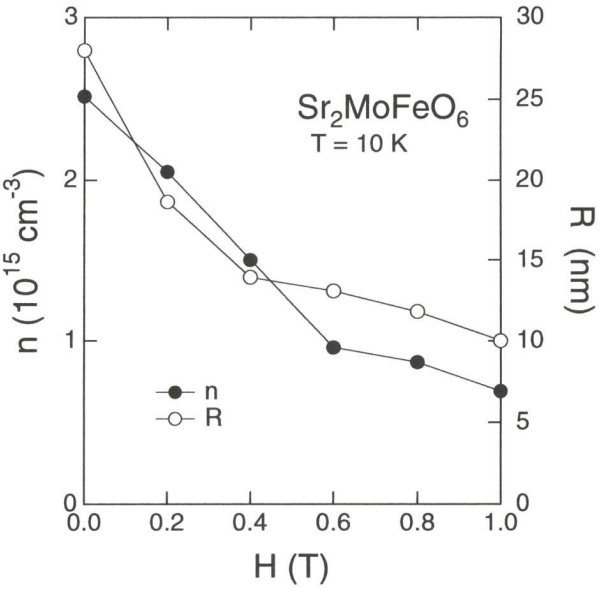

Fig. 4. Number density and averaged radius of magnetic clusters in Sr_2FeMoO_6 at $T = 10$ K.

specious interpretation that these ferrimagnetic clusters play a important part for the GMR effect. In the case of GMR effect explained with a double exchange mechanism, it is expected that the conspicuous GMR effect appears close to $T_N = 420$ K. From our resistivity measurements, the GMR effects become more conspicuous at lower temperatures where thermal fluctuation of the Fe spins is suppressed. This result leads to the conclusion that the origin of the GMR effect is dominated by spin-dependent scattering of conduction electrons from ferrimagnetic clusters with a finite boundary.

De Teresa *et al.* suggested that the CMR effect of $(La_{1-x}Tb_x)_{2/3}Ca_{1/3}MnO_3$ ($x = 0.25$ and 0.33) correlates with the existence of ferromagnetic fluctuation which appears close to the Curie temperature.[3] They analyzed the magnetic scattering by a Lorentzian-type q dependence. The obtained magnetic correlation length was several nm within a temperature range where the CMR effect appears. We notice that the negative MR effect correlates with the existence of magnetic structures in mesoscopic scale in both systems of $(La_{1-x}Tb_x)_{2/3}Ca_{1/3}MnO_3$ and Sr_2FeMoO_6. However, the magnetic correlation length of Sr_2FeMoO_6 is ten times longer than that of the hole-doped manganese perovskite system $(La_{1-x}Tb_x)_{2/3}Ca_{1/3}MnO_3$ within the temperature range where the CMR effect appeares.

§4. Summary

In summary, we performed the SANS experiments on a single crystal of Sr_2FeMoO_6. We found ferrimagnetic mesoscopic clusters with the radius of several tens nm below T_N. These mesoscopic clusters change into large domains with applying magnetic fields enough to lead to a large drop of resistivity. Our result reveals that these ferrimagnetic clusters play a important part for the GMR effect of Sr_2FeMoO_6. The GMR effect of this compound is dominated by spin-dependent scattering of conduction electrons from ferrimagnetic clusters in mesoscopic scale.

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