Pressure Effect on Transport and Magnetic Properties of A_2FeMoO$_6$ (A = Ba, Sr)

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Abstract

For double perovskite oxides Ba_2FeMoO_6 (BFMO) and Sr_2FeMoO_6 (SFMO), resistivity and magnetization have been measured under pressure and magnetic field. By applying pressure the magnetic transition temperature T_C is largely and slightly enhanced for BFMO and SFMO, respectively. The different value of T_C between BFMO and SFMO can be explained in terms of the chemical pressure effect. The saturation magnetization does not exhibit the pressure dependence in both of BFMO and SFMO. The resistivity of BFMO is drastically reduced by applying pressure, although the pressure effect on magnetoresistance is quite small, indicating that the components of conduction which contribute to the colossal magnetoresistance effect hardly depends on pressure.

Key words: double perovskite; CMR; pressure effect;

Recently, double perovskite oxides A_2FeMoO$_6$ (A = Ba, Sr, Ca) have attracted much attention since colossal magnetoresistance (CMR) was observed in these materials in low fields even at room temperature [1]. The origin of the magnetoresistance (MR) was interpreted as due to intergrain tunneling of spin-polarized electrons with the half-metallic electronic structure. The double perovskite structure in A_2FeMoO$_6$ consists of alternating FeO_6 and MoO_6 octahedra. The Fe^{3+} ($3d^5$; $t_{2g}^3 e_g^2$, $S = 5/2$) and Mo^{5+} ($4d^1$; t_{2g}^1, $S = 1/2$) sublattices are believed to be coupled antiferromagnetically. On the other hand, neutron diffraction studies [2,3] reported the absence of the moment at the Mo sites and the value of $\sim 4\ \mu_B$ at the Fe sites. The expected saturation magnetization M_s are 4 μ_B per formula unit. However the observed M_s of A_2FeMoO$_6$ are less than 4 μ_B/f.u. The depression of M_s is ascribed to the disorder between Fe and Mo sites [4]. The notable feature of A_2FeMoO$_6$ is to have the high magnetic transition temperature T_C which is as high as 320-450 K. In the localized-spin model [5], the high T_C is attributed to a large superexchange coupling between Fe and Mo ions. The localized-spin model however contradicts the metallic nature of single crystalline Sr_2FeMoO_6 [6]. The origin of high T_C still remains unsettled.

We present a pressure effect on magnetization, resistivity and magnetoresistance in A_2FeMoO$_6$ (A = Ba, Sr). Pressure is a useful technique for exploring the relation among the crystal structure and the magnetic and transport properties.

The polycrystalline samples of Ba_2FeMoO_6 (BFMO) and Sr_2FeMoO_6 (SFMO) were prepared by solid state reaction. Stoichiometric amounts of $BaCO_3$, $SrCO_3$, Fe_2O_3 and MoO_3 were mixed, ground and calcined at 900 °C for 24 hours in air. The mixture was ground and calcined again in the same conditions. The calcined mixtures were reground, pressed and sintered at 1200 °C for 24 hours in a flow of 5%H_2/Ar. Results of x-ray diffraction indicated that the samples were in a single phase. Magnetization was measured using a SQUID magnetometer in fields up to 5 T. Resistivity and MR were measured by a standard four-prove method in fields up to 7 T. Hydrostatic pressure was generated by using a piston-cylinder cell. The sample space was filled with a pressure transmitting medium of a mixture of Fluorinert FC70 and FC77. Pressure was calibrated from the pressure dependence of superconducting temperature of Pb.

Fig. 1(a) shows the pressure dependence of Curie tem-

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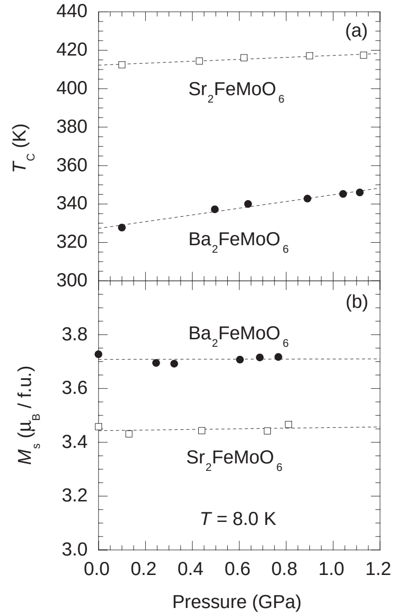

Fig. 1. The pressuee dependence of Curie temperature T_C (a) and saturation magnetization M_s (b) in Ba_2FeMoO_6 (solid circle) and Sr_2FeMoO_6 (open square).

perature T_C of BFMO and SFMO at $T = 8$ K. With increasing pressure, T_C for BFMO increases linearly from 327.7 K to 346.0 K at a rate of $dT_C/dP \sim$ +18 K/GPa. On the other hand, T_C for SFMO slightly increases from 412.4 K to 417.4 K at a rate of $dT_C/dP \sim$ +5 K/GPa. Reduction of the lattice constant yields the enhancement of T_C. The lager value of T_C in SFMO than that in BFMO can be explained in terms of the chemical pressure effect. However, since the value of T_C for Ca_2FeMoO_6 is roughly 350 K, it seems that, with decreasing the ion size of alkaline earth cation, T_C does not increase monotonously but the variation of T_C changes from an increase to a decrease. The slight change of T_C in SFMO suggests that the ion size of Sr is nearly optimum.

For both of BFMO and SFMO the variation of the saturation magnetization M_s is no more than 1% by applying pressure up to 0.8 GPa, as shown in Fig. 1(b). The values of M_s are $\sim$ 3.70 μ_B/f.u. and $\sim$ 3.45 μ_B/f.u. for BFMO and SFMO, respectively. The absence of the pressure effect on M_s is consistent with the report that the deviation from the ideal value of 4 μ_B is due to the disorder between Fe and Mo sites [4].

As shown in Fig. 2, by applying pressure up to 1.19 GPa the resistivity ρ of BFMO is reduced by half over a whole temperature range investigated. Fig. 3 shows the magnetic field dependence of ρ and magnetoresistance (MR) $\{\rho(H)-\rho(0)\}/\rho(0)$ at 5 K in BFMO. The resistivity is drastically reduced by applying pressure at all fields up to 7 T, although the pressure effect on MR is quite small. This result indicates that the components of conduction which does not contribute to the colossal magnetoresistance effect are

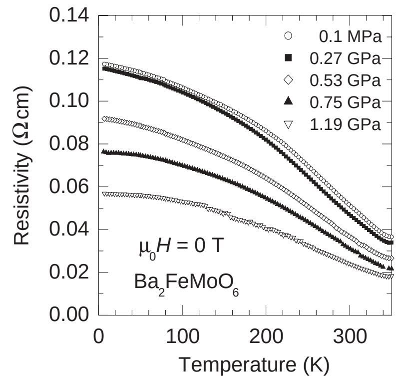

Fig. 2. The temperature dependence of resistivity for Ba_2FeMoO_6 at zero field under several pressures.

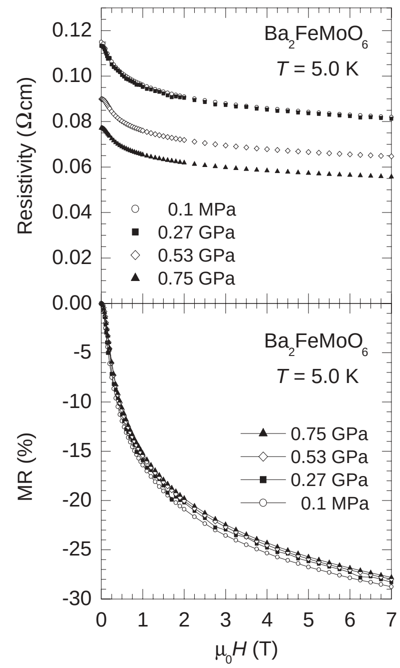

Fig. 3. The magnetic field dependence of resistivity (upper panel) and magnetoresistance (lower panel) for Ba_2FeMoO_6 at 5.0 K under several pressures.

drastically affected by pressure. It seems that the MR have a close connection with M_s because of the slight pressure dependence of both of them.

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