X-Ray Diffraction Study on High-Temperature Colossal Magnetoresistance in Single-Crystal NdBaMn$_2$O$_6$

The crystal structure of the insulating phase in NdBaMn$_2$O$_6$ was investigated using synchrotron radiation X-ray diffraction. A unit cell of the insulating phase is the same as that of the charge-ordering phase of SmBaMn$_2$O$_6$ between 200 and 380 K. The resistance in the insulating phase steeply decreases in a certain magnetic field. The high-magnetic-field phase is the same as the metallic phase just above the metal-insulator transition temperature in zero magnetic field. A resistance change larger than two orders of magnitude is observed upon the phase transition by the application of a magnetic field lower than 2 T at 297 K.

Double-perovskite manganese oxides REBaMn$_2$O$_6$ (where RE is a trivalent rare earth) have various electrical and magnetic phases depending on the ionic radius of RE [1-3]. When the RE sites are occupied by Sm or smaller ions, charge ordering is stabilized in a wide temperature range. The charge ordering phase transition temperature is above 380 K. In contrast, in NdBaMn$_2$O$_6$ and PrBaMn$_2$O$_6$, the A-type antiferromagnetic phase is stabilized below room temperature. Some previous studies using poly-crystalline samples suggested that the A-type antiferromagnetism should take place upon the metal-insulator phase transition. Our recent study using single crystals, however, showed that the onset of A-type antiferromagnetism is lower than the metal-insulator phase transition temperature [4]. Moreover, a crystal structure analysis by means of synchrotron X-ray diffraction has suggested that the metal-insulator phase transition might be caused by x^2-y^2-type orbital ordering [4]. Though the orbital ordering phase is consistent with A-type antiferromagnetism, the insulating behavior within the *ab*-plane cannot be clearly explained. To clarify the inconsistency, we carried out synchrotron X-ray diffraction measurement in the insulating phase using a larger single crystal [5].

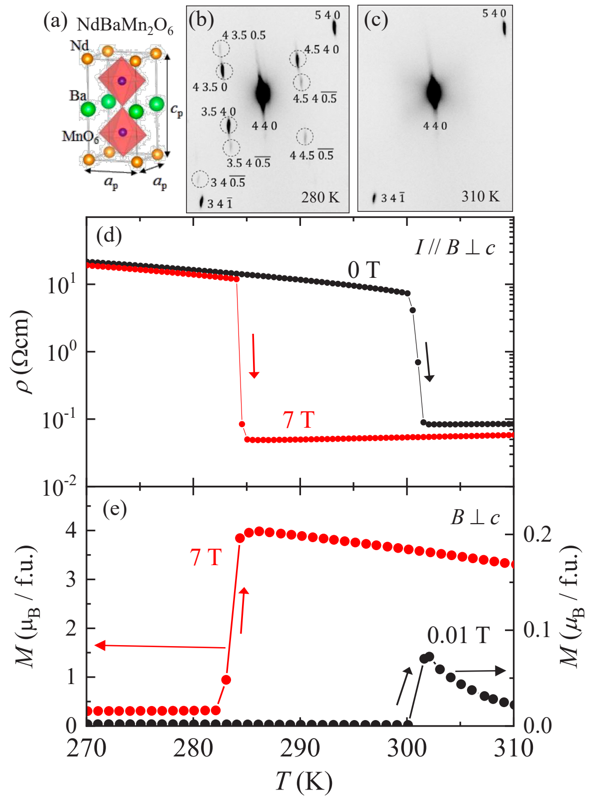

Figure 1: X-ray oscillation photographs of a NdBaMn$_2$O$_6$ single crystal around 4 4 0 at (b) 280 K and (c) 310 K. The indices are given for the $\sqrt{2}a_p \times \sqrt{2}a_p \times 2c_p$ unit cell, where a_p and c_p are lattice constants of the simple tetragonal unit cell shown in (a). Temperature dependence of (d) electrical resistivity at 0 T and at 7 T, and (e) magnetization at 0.01 T and 7 T. The magnetic field is applied perpendicular to the *c*-axis, and parallel to the electric current in (d).

The resistivity within the ab-plane steeply changes at 300 K(T_{MI}) in Fig. 1(d). Figure 1(b) and (c) are single-crystal X-ray oscillation photographs which were measured at BL-8A and 8B. The diffraction pattern above T_{MI} [Fig. 1(c)] is clearly different from that below T_{MI} [Fig. 1(b)]. All the X-ray reflections in this report are indexed on the basis of the $\sqrt{2}a_p \times \sqrt{2}a_p \times 2c_p$ unit cell, where a_p (~0.4 nm) and c_p (~0.8 nm) are the lattice constants of the primitive tetragonal unit cell of double perovskite, as shown in Fig. 1(a). Only the fundamental reflections of the $\sqrt{2}a_p \times \sqrt{2}a_p \times 2c_p$ unit cell are observed above T_{MI}, as shown in Fig. 1(c). In contrast, many superlattice reflections appear below T_{MI} in Fig. 1(b). The crystal contains orthorhombic twins, as mentioned in our previous paper. The superlattice reflections indicate that the unit cell below T_{MI} is . This unit cell is the same as that of the charge ordering phase of SmBaMn$_2$O$_6$ between 200 K and 380 K [6]. Note that the intensities of the superlattice reflections are more than six orders of magnitude weaker than some fundamental reflections. In our previous study, we used a crystal of dimensions and could not detect superlattice reflections. In the present X-ray study we used a much larger crystal with dimensions of 1 mm x 1 mm x 2 mm. The checkerboard-type charge ordering which is the same as that of SmBaMn$_2$O$_6$ is consistent with the insulating behavior within the *ab* plane. The microscopic origin of the coexistence of the A-type antiferromagnetic order and the checkerboard-type charge order remains unexplained.

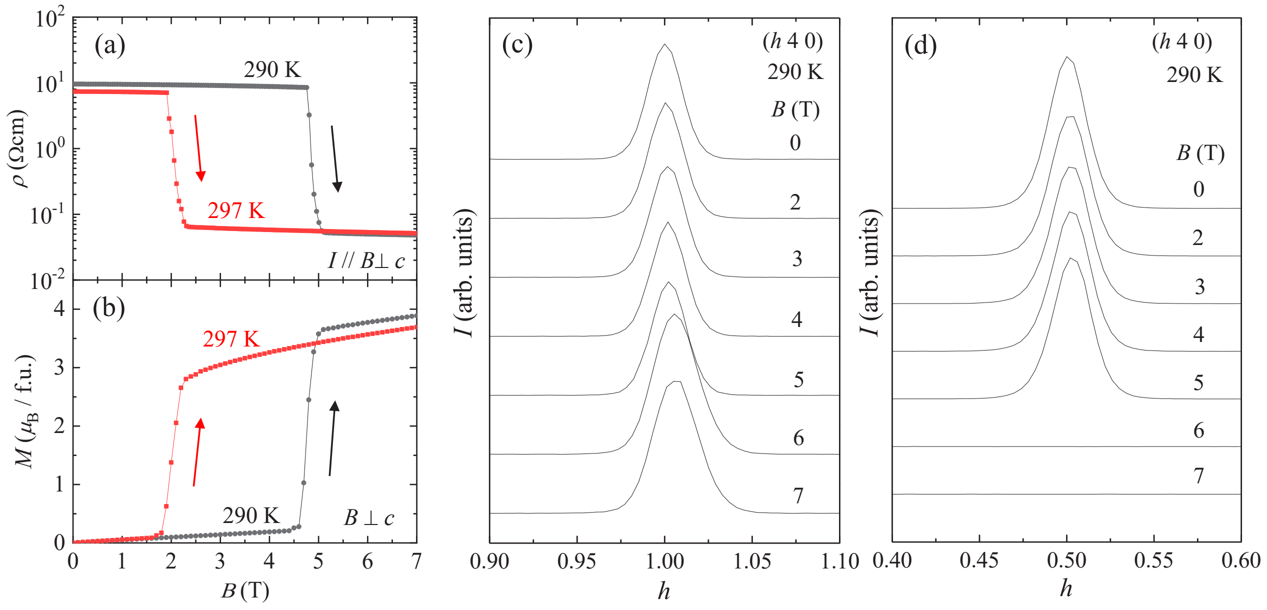

Figure 2: Magnetic field dependence of (a) resistivity and (b) magnetization at 290 K and 297 K. Peak profile along (h 4 0) around (c) h = 1 and (d) h = 0.5 at 290 K in a magnetic field for the warming run. The indices in (c) and (d) are given for the $\sqrt{2}a_p \times \sqrt{2}a_p \times 2c_p$ unit cell.

In an external magnetic field of 7 T, the metal-insulator phase transition temperature decreases to 283 K, as shown in Fig. 1(e). Between 283 and 300 K, hence, the insulating phase is suppressed by the magnetic field. The X-ray reflection profiles at 290 K upon increasing the field, which were measured at BL-3A, are shown in Fig. 2. Though the fundamental reflections of the $\sqrt{2}a_p \times \sqrt{2}a_p \times 2c_p$ unit cell do not disappear up to 7 T, the superlattice reflection of 0.5 4 0 disappears at a magnetic field of 6 T or higher. The critical magnetic field closely corresponds to that of the insulator-metal phase transition [Fig. 2(a)]. This result indicates that the high magnetic field phase is the same as the zero magnetic field phase above T_{MI}. The resistance change is larger than two orders of magnitude at the phase transition induced by a magnetic field lower than 2 T at 297 K.

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BEAMLINES

BL-8A, BL-8B and BL-3A

S. Yamada[1] N. Abe[2, 3], H. Sagayama[4], K. Ogawa[1], T. Yamagami[1] and T. Arima[2] ([1]Yokohama City Univ., [2]The Univ. of Tokyo, [3]Nihon Univ. (Present address) [4]KEK-IMSS-PF)