Phase Separation between Electron-rich Ferromagnetic and Electron-poor Antiferromagnetic Regions on $La_{2-2x}Sr_{1+2x}Mn_2O_7$ Studied by Magnetic Compton Profile Measurement

Perovskite Mn oxides have attracted much interest because of the colossal magnetoresistance (CMR) which appears just above its metal-insulator transition temperature T_c. The ferromagnetism and metallic conductivity below T_c have been interpreted in terms of the double-exchange (DE) mechanism, where e_g orbital electrons go around Mn sites through hybridization with O 2*p* orbitals, and align the localized t_{2g} spins by the strong Hund's coupling [1]. However, the CMR phenomenon cannot be explained by the simple DE mechanism. It is currently pointed out that the orbital degree of freedom is important, as are the charge and spin ones. The determination of e_g (x^2-y^2 and $3z^2-r^2$ orbitals) and t_{2g} orbital occupation in Mn 3*d* state will provide a clue for the clear understanding of the CMR phenomenon existing in this system.

These orbital states can be distinguished on a magnetic Compton profile (MCP) by their characteristic line shapes. For instance, directional Compton profiles of x^2-y^2 and $3z^2-r^2$ atomic orbitals are shown in Fig. 1(a) and 1(b), respectively. This feature makes it possible to determine the e_g and t_{2g} orbital occupation separately [2]. Recently, the temperature dependence of MCP has been measured on a single crystal of $La_{2-2x}Sr_{1+2x}Mn_2O_7$

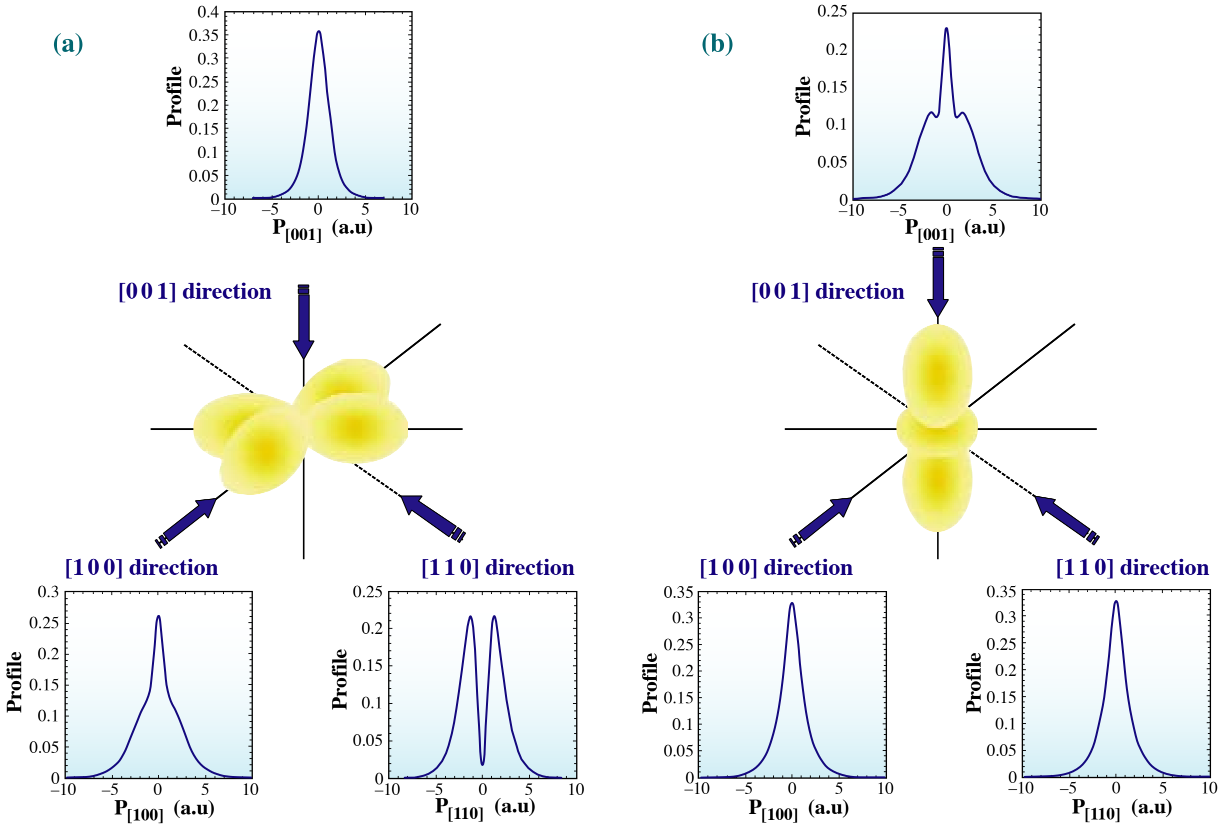

*Fig. 1. Directional Compton profiles of **(a)** x^2-y^2 and **(b)** $3z^2-r^2$ atomic orbitals.*

at x = 0.42 along the c-axis [3]. Experiments were made on beamline **BL08W** using circularly polarized X-rays at 174 keV. MCP's measured at 10 and 150 K are shown in Fig. 2. The area of each MCP is normalized to the magnetic moment measured at each temperature, and it should be noted that T_c lies in between these temperatures. Thus MCP at 150 K reflects a field-induced ferromagnetic state above T_c.

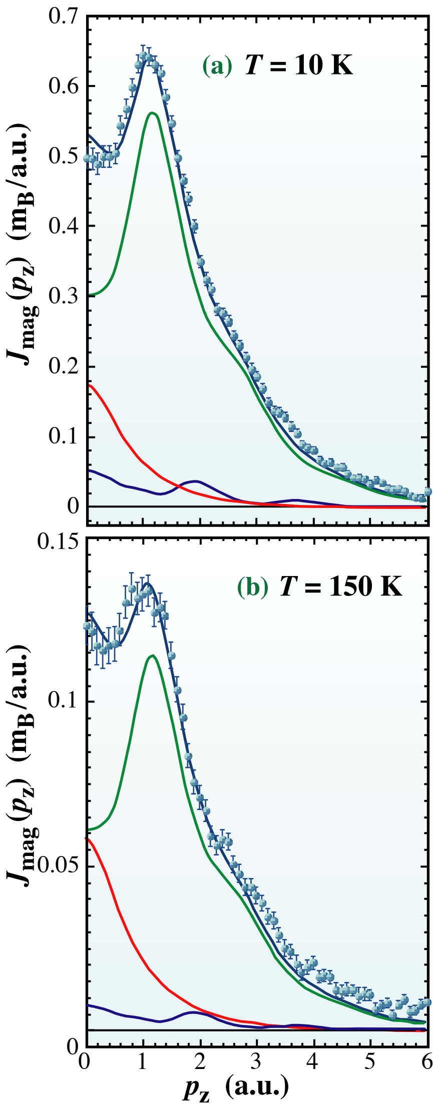

Fig. 2. The magnetic Compton profiles of $La_{2-2x}Sr_{1+2x}Mn_2O_7$ with x = 0.42 measured at ***(a)*** *10 K and* ***(b)*** *150 K. Experimental data and the fitting result are shown with solid circles and solid line, respectively. The green line represents the t_{2g} orbital contribution. The red and blue lines are for the x^2-y^2 and $3z^2-r^2$ contributions in the e_g orbital state, respectively.*

Significant change in shape is observed in a low momentum region; the dip is shallower at 150 K than at 10 K. Since the magnetic moment of the sample almost originates in the spins in Mn 3d orbitals, this behavior means that the ratio of e_g spin to t_{2g} increases above T_c, because the e_g-orbital profile shows a peak at p_z=0, while the t_{2g}-one has a dent. To evaluate the spin magnetic moments in the respective orbitals, a fitting analysis of each MCP was carried out using theoretical profiles of t_{2g} and e_g type orbitals obtained from an *ab initio* molecular orbital calculation for the $(MnO_6)^{8-}$ cluster. In the case of manganites, it is reasonable to assume that the electron number in each orbital is proportional to the number of spins due to the strong Hund's coupling between t_{2g} and e_g spins. If we assume that all Mn ions have the same electron number, that is, t_{2g} and e_g orbital occupations are 3 and 0.58, respectively, the e_g/t_{2g} ratio is expected to be 0.193. However, the ratio at 10 K deduced from the fitting result is 0.234, which is slightly larger than the expected ratio, and the ratio at 150 K shows an even large rvalue of 0.306. This can be interpreted in terms of the phase separation between electron-rich ferromagnetic and electron-poor antiferromagnetic regions [4], because the MCP measurement only observes the ferromagnetic component in the sample. From the ratios, it is found that the e_g electrons are highly segregated in the ferromagnetic region above T_c.

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