# Coexistence of Localized and Itinerant $e_g$ Electrons in a Bilayer Manganite

Perovskite manganites are intriguing materials as a subject of research because of a variety of physical properties, such as metal-insulator transition, colossal magnetoresistance effect, charge ordering, and orbital ordering. In these systems, the Mn-3*d* electronic state, which is split into the $e_g$ and $t_{2g}$ orbitals in the crystal field of $MnO_6$ octahedron, is responsible for the physical properties. Thus far, the ferromagnetic metal (FM) state below $T_c$ has been explained on the basis of the double exchange mechanism in which the spin degree of freedom is taken into account. Recent studies on manganites have opened our eyes anew to the importance of the orbital degree of freedom as well as charge and spin ones. In this study, two-dimensional spin momentum density (2D-SMD) has been reconstructed from directional magnetic Compton profiles (MCPs) measured in a bilayer manganite and has been examined from the viewpoints of the momentum wave functions derived from both molecular orbital and band calculations in order to verify the localized and/or itinerant characteristics of the Mn-3d electronic state [1].

MCP, $J_{mag}(p_z)$, is defined as

$$J_{mag}(p_z) = \iint (\Sigma|\chi_{i\uparrow}(\mathbf{p})|^2 - \Sigma|\chi_{j\downarrow}(\mathbf{p})|^2)\, dp_x dp_y, \quad (1)$$

where $p_z$ is an electron momentum component along the scattering vector of X-rays, and $\chi_{i\uparrow(\downarrow)}(\mathbf{p})$ is a momentum wave function of the *i*-th electron with an up spin (down spin) in the initial state. As shown by eq. (1), $J_{mag}(p_z)$ is represented as the double integral of the difference in momentum density, $|\chi_i(\mathbf{p})|^2$, between the majority and minority spin states with respect to $p_x$ and $p_y$. Thus, the integrated intensity of $J_{mag}(p_z)$ is proportional to the amount of spin magnetic moments in a ferromagnetic sample, and the shape of $J_{mag}(p_z)$ depends on both the orbitals occupied by magnetic electrons and observation directions when we measure MCP in a single crystal. These features enable us to evaluate the occupation number with respect to each orbital on the basis of the fact that the spins in Mn-3*d* orbitals induce the magnetization of manganite system. When we measure a number of MCPs in different directions, SMD can be reconstructed from them. This approach reveals the Mn-3*d* electronic state visually.

The sample used is a single crystal of $La_{2-2x}Sr_{1+2x}Mn_2O_7$ with x = 0.35. The Brillouin zone shows almost a two-dimensional nature because of a structural feature, that is, $MnO_2$ bilayers alternate with $(La,Sr)_2O_2$ blocking layers along the *c* axis. According to the magnetic phase diagram determined by neutron-diffraction measurement, the present sample shows an in-plane ferromagnetic structure below

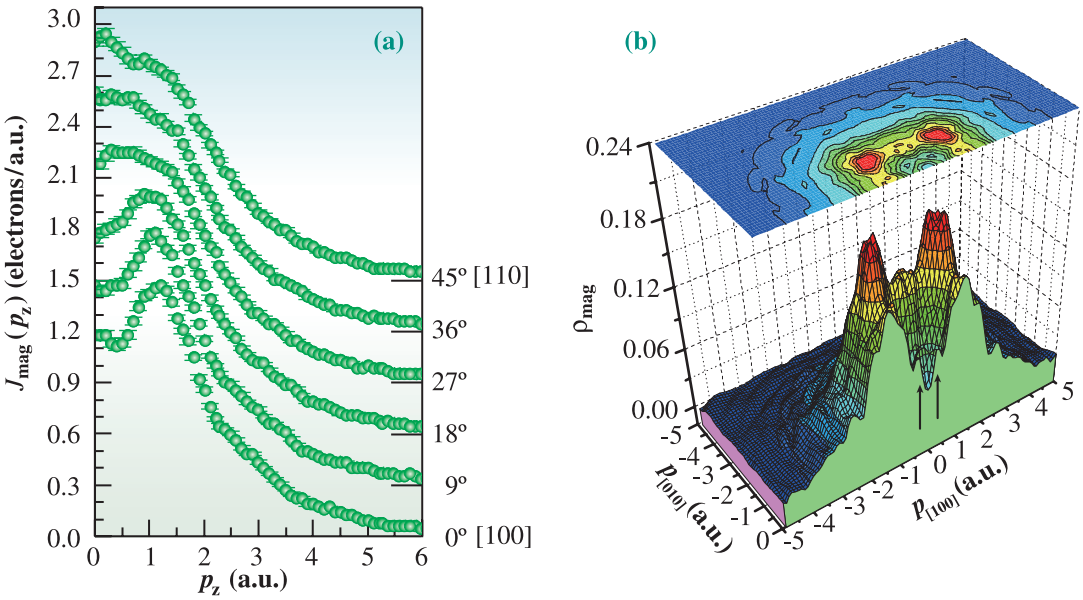


Fig. 1. MCPs and 2D-SMD. **(a)** Directional MCP's measured between the [100] and [110] axes at intervals of 9°. **(b)** 2D-SMD projected on (100) plane, which was obtained by the expansion of the measured area according to structural symmetry.

$T_c$ (≈ 125 K) [2]. MCP measurement was performed at beamline **BL08W**. An external magnetic field of ±2.5 T was alternately applied to the sample at 10 K. The magnetic effect was obtained using the difference between the two Compton profiles measured in the opposite field directions.

Figure 1(a) shows the MCPs measured between the [100] and [110] axes at 9° intervals. The orbital structure, which consists of the $t_{2g}$ and $e_g$ orbitals, is clearly reflected in the change in MCP depending on observation direction. The 2D-SMD, which is the projection of the spin momentum density of Mn-3*d* orbitals on the (001) plane, was reconstructed from the directional MCPs by using the direct Fourier-transform method as shown in Fig. 1(b) [3]. In order to evaluate orbital occupancy, we derived the 2D-SMDs of the $x^2 - y^2$, $3z^2 - r^2$ and $t_{2g}$ orbitals from the result of a molecular orbital calculation for $(MnO_6)^{8-}$ cluster [4], and fitted them to the experimental 2D-SMD. The occupation numbers obtained are 0.39, 0.23 and 3.03 for the $x^2 - y^2$, $3z^2 - r^2$ and $t_{2g}$ orbitals, respectively, indicating the dominance of the $x^2 - y^2$ orbital in the $e_g$ state. We also derived the theoretical 2D-SMD from the result of a local spin density approximation band calculation and compared it with the experimental 2D-SMD. The theoretical 2D-SMD externally appears to reproduce the experimental one. Then, the Lock-Crisp-West (LCW) folding method [5], which transforms the momentum density to an occupation-number density (OND) in the first Brillouin zone, was applied to both the 2D-SMDs. The obtained result is shown in Fig. 2. The upper and lower left panels indicate the 2D-ONDs derived from the experimental and theoretical 2D-SMDs, respectively. In the figure, $t_{2g}$-like bands basically draw a flat structure because of full occupancy, emphasizing on $e_g$-like bands. The electron pocket around the Γ point, the zonal distribution along the $k_{[100]}$ and $k_{[010]}$ axes, and hole pockets around corners are consistent between the two 2D-ONDs, while the dense areas around both ends of the axes cannot be explained only by the present band calculation. The discrepancy would be understood by considering the contribution of a molecular orbital, because the small but sharp peaks denoted by arrows in Fig. 1(b) well reflect a hybridization effect between the $x^2 - y^2$ and O-2*p* orbitals. Then, we tentatively obtained the 2D-OND of the $x^2 - y^2$ orbital, although the LCW method ought to be basically applied to the momentum density described by the Bloch state. The result shown in the lower right panel of Fig. 2 well explains the dense areas in the experimental 2D-OND. This is considered an indicator of the coexistence of localized and band like $e_g$ electrons in the FM state below $T_c$.

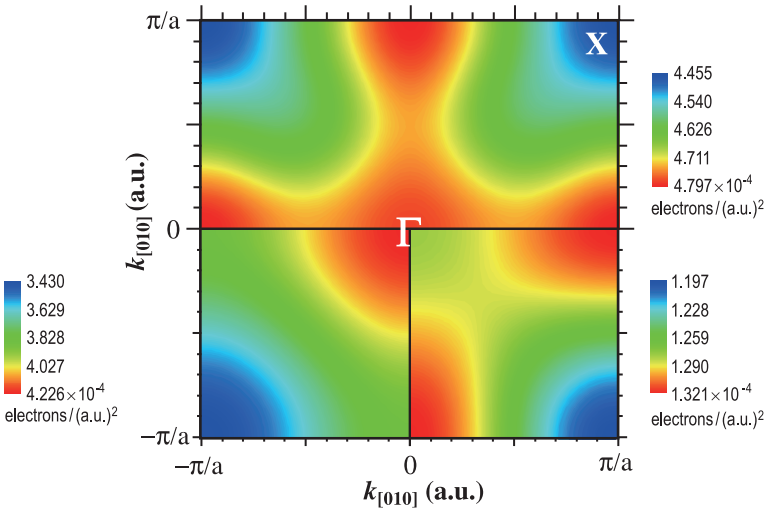


Fig. 2. 2D-OND derived from experimental 2D-SMD (upper), band calculation (lower left) and $x^2 - y^2$ orbital (lower right).

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**References**

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