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Polarization dependence of the Mn *K* pre-edge absorption spectra in $LaMnO_3$

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Introduction

$LaMnO_3$ (LMO) is known as a parent compound of colossal magnetroresistance (CMR) perovskite-type manganites and has attracted much attention as a good test ground to study the orbital ordering. Murakami *et al.* reported in their pioneering works to have found a first direct evidence of orbital ordering in LMO by using resonant x-ray scattering at the Mn *K*-edge [1]. There has been a long debate, however, whether the driving force of the orbital ordering is the Jahn-Teller (JT) distortion of the MnO_6 octahedra or the 4*p*-3*d* Coulomb interaction. Recently, we have performed x-ray linear dichroism (XLD) study using the total fluorescence yield method in order to get another insight into the orbital ordering [2] and concluded that XLD at the Mn *K* main absorption edge is due to the JT distortion. The main edge is assigned to 1*s* to 4*p* dipole transition, therefore, we could not rush to a conclusion that our results have some relation to the 3*d* orbital ordering nature.

In this report, we present more precise investigation of XLD spectra focusing our attention on the pre-edge region where two broad peaks exist, which is believed to be derived from 1*s* to 3*d* quadrupole transition. A direct evidence of orbital ordering may be partially obtained.

Experimental

Mn *K*-edge x-ray absorption spectra (XAS) were obtained by the total electron yield (TEY) method mainly on BL-9A. An *ac*-plane single-crystal was mounted on a rotary stage perpendicular to the incident x-ray (the *b*-axis is parallel to the x-ray) and was put in a vacuum of the order of 10^{-2} Torr. The polarization vector (*E*) of the x-ray lies in the horizontal plane. Two-fold symmetry of the spectra was carefully checked in advance. All the experiments were carried out at room temperature.

Results and Discussion

Figure 1 shows the Mn *K*-edge XAS spectra of *E* parallel to the *a*-axis (*E*//*a*: blue dots) and *c*-axis (*E*//*c*: red dots) respectively. In both spectra, white lines together with two broad peaks (A1 at 6539.2 eV and A2 at 6541.5 eV) are observed. The energy shift of the white line is consistent with our previous result that ensures the validity of TEY method. This shift is attributable to the different energy shift of three 4*p* unoccupied bands: the bottom of $4p_x$ band lies several eV below the other $4p_y$ and $4p_z$ bands [3], resulting that the spectra of 1*s* to $4p_z$ dipole transition (namely, *E*//*c* configuration) shifted higher energy.

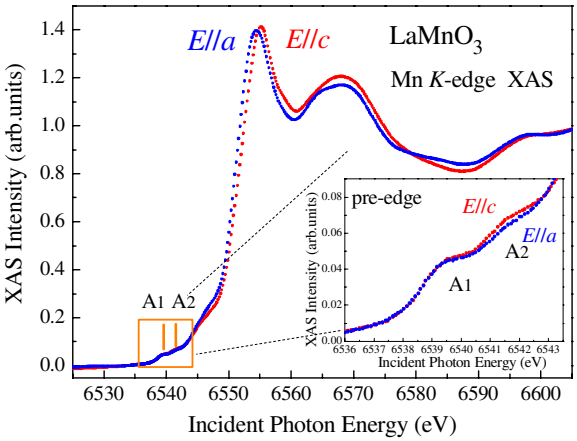

Figure 1. Mn *K*-edge x-ray absorption spectra (XAS) of an *ac*-plane single-crystal $LaMnO_3$. The polarization vector (*E*) of the incident x-ray is parallel to the *a*-axis (blue dots) and *c*-axis (red dots) respectively. The inset shows enlarged spectra in the pre-edge region.

The inset shows the details around A1 and A2 peaks. Weak but clear polarization dependence was observed at A2 peak: the intensity of A2 peak in *E*//*c* configuration is larger than that of *E*//*a* one. Each spectra was obtained by averaging over more than 10 scans, therefore the statistical error is very low, and besides, it should be mentioned that only naïve data analyses were applied to the spectra: constant scaling factor with subtraction of constant backgrounds. These good quality data were obtained by taking full advantage of TEY method: a perfect symmetric configuration without suffering backward scatterings as in the case of TFY method. We tentatively conclude the difference in A2 peak reflect the difference in the occupancy of 3*d* orbitals.

References

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