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Orbital ordering of layered manganites from resonant soft X-ray scattering

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Abstract

We present measurements of Mn *L*-edge resonant soft X-ray scattering on single-layered manganites with different sizes of cations. Orbital ordering of $Pr_{0.5}Ca_{1.5}MnO_4$ exhibits a stronger three-dimensional character and a dramatically enhanced transition temperature, as compared with those of $La_{0.5}Sr_{1.5}MnO_4$. The *c*-axis correlation length of orbital ordering in $Pr_{0.5}Ca_{1.5}MnO_4$ is about half of the in-plane correlation length. Our results indicate that reduction in one-electron bandwidth and quenched disorder strongly enhances the stabilization of charge–orbital ordering.

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Keywords: Manganite; Charge–orbital ordering; Resonant soft X-ray scattering

1. Introduction

Physical phenomena, such as colossal magnetoresistance and metal–insulator transition, of correlated electron compounds typically arise from the interplay among the spin, charge, orbital, and lattice degrees of freedom [1]. For example, the colossal magnetoresistance of manganites are strongly affected by the lattice distortion resulting from the Jahn–Teller effect or the sizes of the A-site cations; the latter leads to the change of the effective one-electron bandwidth and quenched disorder. The smaller the average radius r_A of the A-site cations is, the narrower the bandwidth is. Such a narrowing of bandwidth tends to stabilize the charge–orbital ordering [2]. The mismatch between the ionic radii of divalent and trivalent cations, i.e., quenched disorder, also affect the stabilization of spin and charge–orbital ordering [3,4].

To observe orbital ordering directly is a difficult task. Experimental results of resonant X-ray scattering (RXS) at the Mn 1s threshold of $La_{0.5}Sr_{1.5}MnO_4$ have been presented to be direct evidence for orbital ordering [5]. However, RXS detects the 3d charge ordering indirectly via the $1s \rightarrow 4p$ transition and the hybridization between 4p and 3d electronic states. Calculations based on a local-density approximation including on-site Coulomb interactions [6,7] and multiple scattering theory [8] indicate that RXS measurements pertain mainly to Jahn–Teller distortion, instead of directly observing 3d orbital ordering. In contrast, resonant soft X-ray scattering around the Mn *L*-edge ($2p \rightarrow 3d$) are dipole-allowed and suitable for probing the Mn 3d charge–orbital orderings directly and with high sensitivity.

Here, we present measurements of Mn *L*-edge resonant soft X-ray scattering on $La_{0.5}Sr_{1.5}MnO_4$ and $Pr_{0.5}Ca_{1.5}MnO_4$

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to measure the transition temperature and the correlation length of the orbital ordering with variations of one-electron bandwidth and quenched disorder.

2. Experimental setup

Single crystals of $La_{0.5}Sr_{1.5}MnO_4$ and $Pr_{0.5}Ca_{1.5}MnO_4$ were grown with the floating zone method, and were characterized with X-ray diffraction at room temperature. $La_{0.5}Sr_{1.5}MnO_4$ has a larger r_A (~1.28 Å) than that of $Pr_{0.5}Ca_{1.5}MnO_4$ (~1.18 Å); $Pr_{0.5}Ca_{1.5}MnO_4$ also has a much smaller variance of ionic radii of the A-site ions [9].

We measured resonant soft X-ray scattering on single crystals of $La_{0.5}Sr_{1.5}MnO_4$ and $Pr_{0.5}Ca_{1.5}MnO_4$ at 90 K with the elliptically polarized-undulator beamline of National Synchrotron Radiation Research Center (NSRRC), Taiwan. Crystals were cut with [1 1 0] surface normal, and aligned in a two-circle soft X-ray diffractometer with the [1 1 0] and [0 0 1] axes defining the scattering plane. The **E** vector of photons were perpendicular to the scattering plane, and the energy resolution was 0.14 eV at 640 eV.

3. Results and discussion

With measurements of soft X-ray scattering, we found that orbital ordering in both $La_{0.5}Sr_{1.5}MnO_4$ and $Pr_{0.5}Ca_{1.5}MnO_4$ exhibits a modulation vector $(\frac{1}{4}\frac{1}{4}0)$ in reciprocal lattice units. Fig. 1 shows the photon-energy dependence of the $(\frac{1}{4}\frac{1}{4}0)$ scattering intensities around the Mn L edges, consistent with previous measurements [10]. The spectra of $(\frac{1}{4}\frac{1}{4}0)$ resonant scattering from these two half-doped manganites have a similar line shape; detailed analysis of the measured energy dependence of soft X-ray scattering from orbital ordering will be discussed elsewhere. The transition temperature T_{CO} of charge–orbital ordering in $La_{0.5}Sr_{1.5}MnO_4$ is 235 K; whereas $Pr_{0.5}Ca_{1.5}MnO_4$ has a dramatically enhanced T_{CO} of 326 K. In addition, the charge–orbital ordering of $Pr_{0.5}Ca_{1.5}MnO_4$ has a thermal hysteresis shown in the inset of Fig. 1, in agreement with measurements of resistivity [9].

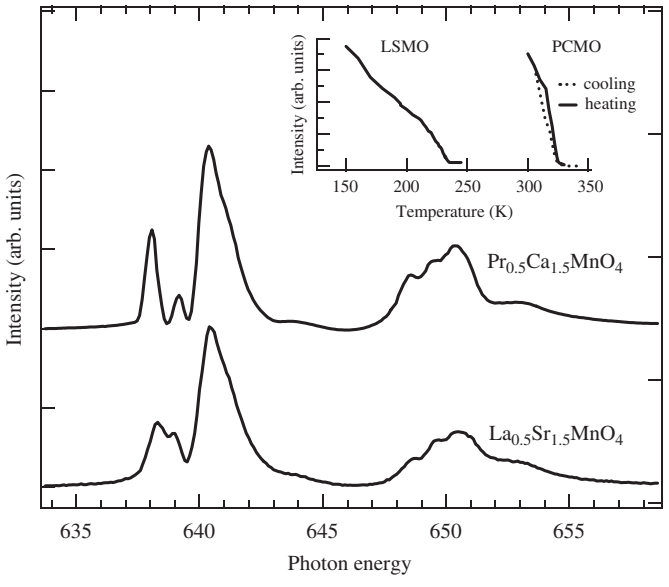

Fig. 1. Photon-energy dependence of the Mn L-edge $(\frac{1}{4}\frac{1}{4}0)$ scattering of $La_{0.5}Sr_{1.5}MnO_4$ and $Pr_{0.5}Ca_{1.5}MnO_4$, denoted as LSMO and PCMO, respectively. The inset displays the temperature dependence of the $(\frac{1}{4}\frac{1}{4}0)$ scattering intensities.

We also measured the correlation lengths along the [1 1 0] and the [0 0 1] directions, denoted as $\xi_{1\,1\,0}$ and ξ_{001}, respectively. Fig. 2 displays momentum-transfer dependence of Mn L-edge resonant $(\frac{1}{4}\frac{1}{4}0)$ scattering of $La_{0.5}Sr_{1.5}MnO_4$ and $Pr_{0.5}Ca_{1.5}MnO_4$ with momentum transfer q along these two directions, i.e., q_{110} and q_{001} scans. The correlation length is defined as the inverse of the half-width at the half-maxima in the q scan. The measured ξ_{110} and ξ_{001} are 340 and 75 Å, respectively, for $La_{0.5}Sr_{1.5}MnO_4$, and 668 and 370 Å for $Pr_{0.5}Ca_{1.5}MnO_4$. The results indicate that the in-plane correlation ξ_{110} of $Pr_{0.5}Ca_{1.5}MnO_4$ about twice of that of $La_{0.5}Sr_{1.5}MnO_4$. In addition, the ratio of ξ_{001}/ξ_{110} in the orbital ordering of $Pr_{0.5}Ca_{1.5}MnO_4$ is 0.55, significantly larger than that of $La_{0.5}Sr_{1.5}MnO_4$, 0.22.

Based on the correlation lengths measured at the same temperature in the two compounds with different ordering temperatures, we show that the reduction in one-electron bandwidth and quenched disorder strongly enhances the stabilization of orbital ordering. $Pr_{0.5}Ca_{1.5}MnO_4$ also exhibits a more three-dimensional-like orbital ordering than that of $La_{0.5}Sr_{1.5}MnO_4$.

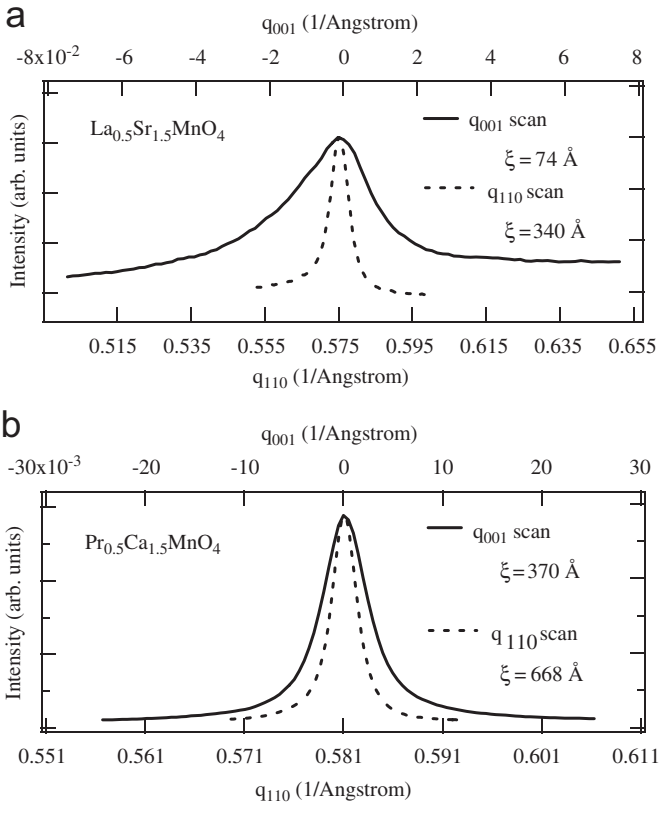

Fig. 2. Momentum-transfer dependence of Mn L-edge resonant $(\frac{1}{4}\frac{1}{4}0)$ scattering of $La_{0.5}Sr_{1.5}MnO_4$ and $Pr_{0.5}Ca_{1.5}MnO_4$ along the [1 1 0] and [0 0 1] directions recorded at 90 K.

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