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In-situ photoemission study of $Pr_{1-x}Ca_xMnO_3$ epitaxial thin films

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Introduction

Hole-doped perovskite manganese oxides $R_{1-x}A_xMnO_3$, where R is a rare-earth (R = La, Nd, Pr) and A is an alkaline-earth atom (A = Sr, Ba, Ca), have attracted much attention because of their remarkable physical properties such as colossal magnetoresistance and the ordering of spin, charge, and orbitals [1]. Half-doped manganites ($x \sim 0.5$) are particularly interesting because most of them exhibit a CE-type charge-ordered (CO) state. The compound $Pr_{1-x}Ca_xMnO_3$ (PCMO), where the bandwidth W is smaller than the other manganites, has a particularly stable CO state in the wide hole concentration range between $x \sim 0.3$ and 0.75 [2]. In this study, in order to observe how the electronic structure of PCMO changes with hole doping, we have performed *in-situ* photoemission spectroscopy and x-ray absorption spectroscopy (XAS) measurements of epitaxial thin films of PCMO (x = 0.2 - 0.6) grown on $LaAlO_3$ (001) substrates (which induces compressive strain) by laser molecular beam epitaxy (laser MBE).

Experimental

The PCMO thin films were fabricated in a laser MBEchamber connected to a synchrotron radiationphotoemission system at BL-2C of the Photon Factory [3]. PCMO thin films were deposited on $LaAlO_3$ (001) substrates at 500 °C at an oxygen pressure of 1×10^{-4} Torr. The fabricated PCMO thin films were transferred into the photoemission chamber under vacuum of 10^{-10} Torr. The photoemission and XAS spectra were taken at room temperature with the total energy resolution of about 200 meV at the photon energy of 600 eV.

Results and Discussion

Figure 1 shows the doping dependence of the valence-band photoemission spectra. One can observe four main structures, labeled as A, B, C, and D. From Mn $2p \rightarrow 3d$ (643 eV) and Pr $3d \rightarrow 4f$ (930 eV) resonant photoemission spectra, structures A, B, C, and D are assigned to Mn $3d\ e_g$, Mn $3d\ t_{2g}$ plus Pr $4f$, non-bonding O $2p$, and Mn $3d$ - O $2p$ bonding states, respectively. Satellite structures were not observed. With hole doping structures A-D moved toward the Fermi level (E_F) as in the rigid-band picture, but there was no density of states at E_F for all values of x, suggesting a non-rigid-band behaviour near E_F with spectral weight transfer across E_F.

From the binding-energy shifts of core levels, it was found that the chemical potential was shifted monotonically without any sign of suppression. The suppression of the chemical potential shift was observed in bulk samples of PCMO [4]. Since the electronic phase separation results in the pinning of the chemical potential, the monotonic chemical-potential shift of PCMO thin films suggests the absence of phase separation on a microscopic scale as in the case of LSMO [5, 6]. The absence of chemical potential pinning is consistent with the suppression of charge ordering under the compressive strain from the $LaAlO_3$ substrates.

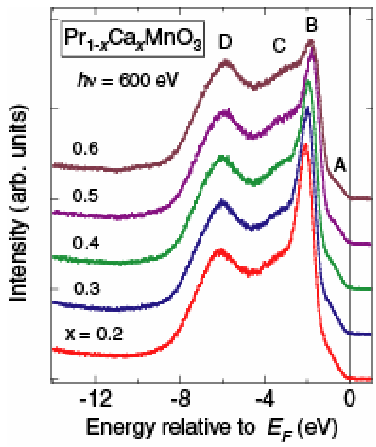

Fig. 1: Valance-band photoemission spectra of $Pr_{1-x}Ca_xMnO_3$ epitaxial thin films.

References

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