

Substrate-Dependent Bonding Anisotropy of Epitaxial Multiferroic DyMnO₃ Thin Films

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We investigated the substrate-dependent electronic structure and anisotropic bonding of the Mn $3d$ states in DyMnO₃ thin films on SrTiO₃(001) and LaAlO₃(110) substrates using polarization-dependent x-ray absorption spectroscopy (XAS) at O K -, Mn L - and Mn K -edges for three polarizations, E || a, E || b and E || c. Polarization-dependent x-ray absorption spectra at O K -, Mn $L_{2,3}$ - and Mn K -edges of orthorhombic DyMnO₃/LaAlO₃(110) thin films show a strong polarization dependence, whereas orthorhombic DyMnO₃/SrTiO₃(001) thin films show nearly isotropic spectral structure. The main peak in polarized Mn $L_{2,3}$ -edge XAS spectra of DyMnO₃/LaAlO₃(110) thin films for the E || b polarization lies at a lower energy than for polarizations E || a and E || c. This indicates a great anisotropy in Mn $3d$ -O $2p$ hybridization, reflecting an orbital ordering and a highly anisotropic coplanar Mn-O bonding in DyMnO₃/LaAlO₃(110) thin films. Orbital ordering of e_g -orbital and the highly anisotropic in-plane Mn-O bonding is an indispensable factor to the formation of complicated incommensurate modulated magnetic structures observed in orthorhombic DyMnO₃. The present results provide important implications for the microscopic understanding of the multiferroic DyMnO₃.