Strong orbital hybridization and single-domain perpendicular magnetism in an ultra-high-quality $SrRuO_3$ epitaxial film

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The itinerant 4*d* ferromagnetic perovskite $SrRuO_3$ [Curie temperature (T_C) = 160 K] has been studied extensively for oxide electronics and spintronics [1]. Especially, the perpendicular magnetic anisotropy induced by the compressive-strain in epitaxial $SrRuO_3$ is beneficial for scalability and the reduction of power consumption in spintronic devices [2]. Recently, an unexpected large magnetic moment of oxygen [3] together with a metal-insulator transition caused by a weakened Ru 4*d* t_{2g}-O 2*p* hybridization [4] have been reported. These results highlight the importance of the oxygen 2*p* states for understanding the perpendicular magnetic anisotropy and electronic structures in $SrRuO_3$.

In this presentation, we report electronic structure and magnetic properties of an ultra-high-quality $SrRuO_3$ film on $SrTiO_3$ grown by machine-learning-assisted molecular beam epitaxy [5,6]. The high itineracy and long quantum lifetimes of the quasiparticles in the Ru 4*d* t_{2g}-O 2*p* hybridized valence band are confirmed by observing the prominent well-screened peak in the Ru 3*d* core-level photoemission spectrum, the coherent peak near the Fermi energy in the valence band spectrum, and quantum oscillations in the resistivity. The element-specific magnetic properties and the hybridization strength between the Ru 4*d* and O 2*p* orbitals were characterized by Ru $M_{2,3}$-edge and O *K*-edge soft X-ray absorption spectroscopy (XAS) and X-ray magnetic circular dichroism (XMCD) measurements. Figure 1 shows the O *K* edge XAS and XMCD spectra for the ultra-high-quality $SrRuO_3$ film. A sizeable negative XMCD signal, which is proportional to the orbital magnetic moment of the O 2*p* states, is observed only in the energy range corresponding to transitions into the Ru 4*d* t_{2g} states. This indicates that a large orbital magnetic moment of oxygen ions is induced by the strong orbital hybridization with the spin-polarized Ru 4*d* t_{2g} states. The $SrRuO_3$ film also shows the single-domain perpendicular magnetization with an almost ideal remnant magnetization ratio of 0.97. These results provide detailed insights into the interplay between orbital hybridization and the perpendicular magnetic anisotropy in $SrRuO_3$/$SrTiO_3$ systems.

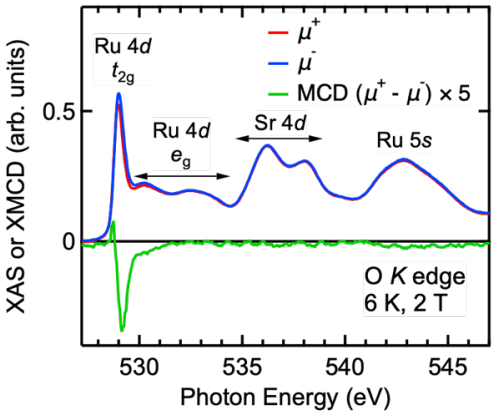

Figure 1 O *K* edge XAS and XMCD spectra for the $SrRuO_3$ film at 6 K with a magnetic field $\mu_0 H$ = 2 T. Here, μ^+ and μ^- denote the absorption coefficients for the photon helicities parallel and antiparallel to the Ru 4*d* majority spin direction, respectively.

References [1] G. Koster *et al.,* Rev. Mod. Phys. **84**, 253 (2012). [2] K. S. Takahashi *et al*., Phys. Rev. B **67**, 094413 (2003). [3] S. Kunkemöller *et al*., Phys. Rev. B **100**, 054413 (2019). [4] H. Jeong *et al*., Appl. Phys. Lett. **115**, 092906 (2019). [5] Y. K. Wakabayashi, *et al*., APL Mater. **7**, 101114 (2019). [6] K. Takiguchi, Y. K. Wakabayashi, *et al*., Nat. Commun. **11**, 4969 (2020).