Correlating magnetic anisotropy and electronic structure in complex oxides thin films

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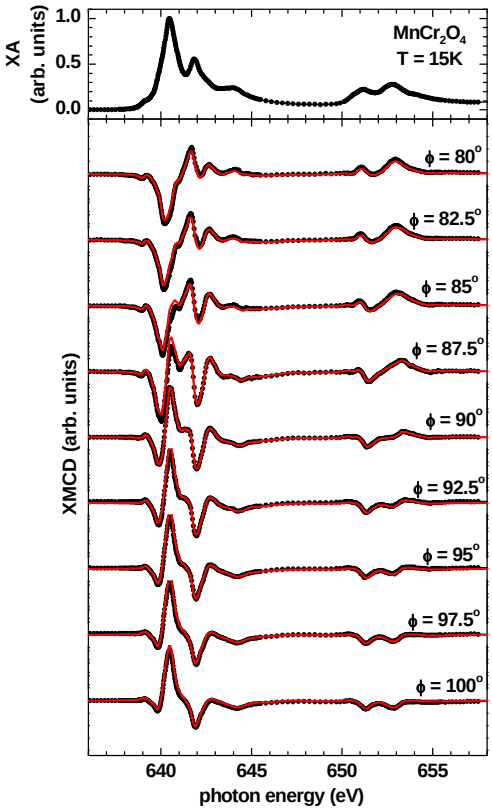
Controlling magnetic anisotropies – and thereby the easy-magnetization directions – in complex oxide thin films is of critical importance for the optimization of novel memory and sensor devices for future use in information storage and processing technology. Since these engineered magnetic nanostructures typically consist of several layers with distinct magnetic characteristics access to element-specific information about the magnetocrystalline anisotropy energy (MAE) is essential. MAEs can be determined quantitatively employing a theoretically derived sum rule for x ray magnetic linear dichroism (XMLD) [1] connecting the integrated intensity of the XMLD signal with the spin-orbit anisotropy and hence the MAE. Moreover, integration and proper normalization of x ray magnetic circular dichroism (XMCD) spectra allow quantifying the anisotropy of the orbital moment, ***L***, which determines the easy-magnetization direction [2]. While the values derived for the MAE using the XMCD and XMLD sum rules agree rather well with each other, the analysis gives energy values larger by a factor of about 10 to 50 compared to macroscopic measurements such as magneto-optical Kerr effect and ferromagnetic resonance [3]. This suggests that the approach of using soft x ray dichroism techniques for the element-specific study of the MAE requires further refinement.

Spin-orbit coupling prefers to align the orbital moment, ***L***, and spin moment, ***S***, collinear with each other to minimize the total energy of a magnetic 3*d* transition metal system. The crystalline electric field, i.e. the electric field created by neighboring charges, acts directly on the electron orbits and prefers ***L*** along a specific crystal symmetry direction. As a consequence, different values of ***L*** are obtained if the spins are oriented along different crystal directions by an external magnetic field. The easy-magnetization direction is given as the direction with the largest component of ***L*** that results in the lowest energy. We aim to evaluate the influence of the crystal electric field and the orbital moment, ***L,*** on the spectral shape and angular dependence of the XMCD signal in order to elucidate the origin of the magnetocrystalline anisotropy.

We have measured the angular dependence of the Mn $L_{3,2}$ XMCD signal from $MnCr_2O_4$ and $MnFe_2O_4$. The XMCD signal was determined for a fixed angle of incidence of 35.3° relative to the sample surface while varying the angle ϕ of the magnetic field relative to the x ray beam. The most pronounced Mn $L_{3,2}$ XMCD feature occurs around 640 eV. Its angular dependence follows as expected a



cosϕ-dependence reflecting the change in projection of the magnetic moments on the photon-spin direction. To clearly show the changes in spectral shape, the Mn $L_{3,2}$ XMCD spectra from $MnCr_2O_4$ in Fig. 1 were normalized to unity at their most pronounced XMCD feature. A very strong variation of the XMCD spectral shape with angle ϕ is evident. Although ϕ was varied in increments of 2.5° the differences between successive spectra near ϕ = 90° are quite pronounced. In transverse geometry, i.e. ϕ = 90°, the XMCD spectral shape resembles the derivative of the XMCD signal observed for ϕ = 0°. Applying the sum rule suggests a vanishing orbital moment, $\boldsymbol{L}$, for ϕ = 90°. The red lines in Fig. 1 show a simulation of the experimental data following the approach outlined in Ref. 4 by using experimental data for the fundamental spectra w_0 and w_2, i.e. XMCD spectra obtained for ϕ = 0° and ϕ = 90°, respectively. The agreement is excellent indicating that for a system without pronounced magnetic anisotropy the model proposed in Ref. 4 is valid.

However, in a comparable data set obtained on $MnFe_2O_4$ exhibiting an in-plane easy axis the angular dependence is distinctly different. For ϕ = 70° the XMCD signal resembles the derivative of w_0, i.e. the XMCD spectrum observed for ϕ = 0°, and application of the sum rule leads to $\boldsymbol{L}$ = 0. By contrast a comparable spectrum was observed at ϕ = 90° for $MnCr_2O_4$. This indicates that a careful study of the angular dependence of the XMCD signal can provide the means to determine the angle of $\boldsymbol{L}$ = 0 and ultimately the anisotropy of the orbital magnetic moment with great precision.

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