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Low-temperature Kerr spectroscopy on half-metallic Sr_2FeMoO_6

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The polar Kerr rotation and ellipticity spectra of epitaxially grown (001)-oriented half-metallic Sr_2FeMoO_6 thin films have been determined in the photon energy range from 1.2 to 4.9 eV. The Kerr rotation spectrum shows three maxima at $E=1.6$, 4.0, and 4.65 eV. The maxima at 4.0 and 4.65 eV are consistent with spin-polarized band structure calculations for interband transitions from the O-2p to the minority-spin π^*Mo/Fe and majority-spin Mo-t_{2g} bands, respectively. The overall maximum intrinsic Kerr rotation is $\Theta_K=-0.045°$ at a photon energy of 4.65 eV. The maximum of Θ_K at $E=1.6$ eV coincides with a minimum in the reflectivity due to the plasma edge of Sr_2FeMoO_6 and, therefore, is not related to an interband transition.

Spin-polarized half-metallic ferromagnetic oxides and metals[1–4] have attracted intensive interest recently for devices based on tunneling and intergrain-tunneling magnetoresistance (ITMR). The performance of these devices is directly related to the degree of spin polarization of the ferromagnetic material near the Fermi energy,[5] to the presence of grain-boundary trap states, and to the intergrain tunneling probability. Kobayashi *et al.*[4] and Tomioka *et al.*[6] have shown that the ITMR in the double perovskite Sr_2FeMoO_6 scales with the square of the normalized saturation magnetization $M_S(T)/M(0)$ and is measurable at room temperature (RT), which contrasts with the low-temperature ITMR found with the half-metallic manganites and CrO_2.[7–10] In this letter, we report on the polar Kerr rotation and ellipticity spectra of epitaxially grown (001)-oriented Sr_2FeMoO_6 films and compare them with spin-polarized band structure calculations.

Sr_2FeMoO_6 crystallizes in an ordered double-perovskite structure in which the Fe and Mo order (imperfectly) on alternate octahedral sites.[11] The crystal symmetry may be cubic[12] or tetragonal[13] depending on the degree of ordering of the Fe and Mo atoms. In a perfectly ordered structure, a theoretical spin-only saturation moment of $4\mu_B$ per formula unit can be predicted depending on whether the valence states are Fe^{3+} and Mo^{5+} with antiferromagnetically coupled localized spin moments[11,14] or whether the Fe^{3+}/Fe^{2+} and Mo^{6+}/Mo^{5+} redox energies overlap to give a majority spin $S=5/2$ on the iron atoms and a minority-spin π band associated with both the Fe and Mo atoms.[4] The former configuration with localized spins would give ferrimagnetism with a paramagnetic Curie Weiss constant $\theta<0$; the latter would give ferromagnetism with a $\theta \geqslant T_C$. Experimentally, a $\theta>0$ is found[15] and the compound is metallic with a measurable ITMR at RT,[4,16–18] which indicates that the latter model is applicable. A spin-polarized local-density-approximation (LDA) calculation[4] places the Fermi level in the minority-spin π^* band to make ideally ordered Sr_2FeMoO_6 a half-metallic ferromagnet (see Fig. 1). However, the highest saturation magnetization found experimentally is in the range of $2.7-3.2\mu_B$ per formula unit;[4,6,15,19] the cubic phase has a little smaller saturation magnetization. The saturation magnetization can be reduced to $2.7\mu_B$ per formula unit by Fe, Mo pair disorder on the octahedral sites according to a Monte Carlo simulation study;[20] it can be reduced to a lesser extent by antiphase boundaries separating well-ordered volumes.[15] The Curie temperature of cubic Sr_2FeMoO_6 is reported to be $T_C \approx 390$ K, that of the tetragonal phase to be $T_C \approx 415$ K.[4,6,15]

(001)-oriented 150-nm-thick Sr_2FeMoO_6 films were deposited on (001)$LaAlO_3$ substrates by pulsed laser deposition using a KrF($\lambda=248$ nm) excimer laser. The experimental details of the thin-film deposition and structural characterization have been described elsewhere.[19] Superconducting quantum interference device measurements reveal a saturation moment per formula unit of $3.2\mu_B$ at 10 K and $2.0\mu_B$ at 300 K with a Curie temperature $T_C=400$ K.[19] Figure 2 shows out-of-plane Kerr hysteresis loops at 10 K for photon energies of 1.4 and 4.0 eV. Magnetic saturation was almost achieved at $B=\pm 1.5$ T, the magnetic field applied during the polar Kerr spectroscopy measurements.

The significant reduction of the saturation moment at RT suggested the performance of the Kerr spectroscopy at 10 K. An optical cryostat inset allows measurements from 10 K to

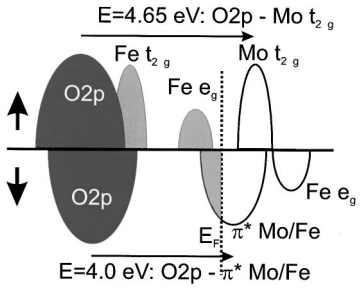

FIG. 1. Schematic representation of the spin-polarized density of states of Sr_2FeMoO_6 according to the LDA calculation. The magnetooptical transitions are indicated by arrows.

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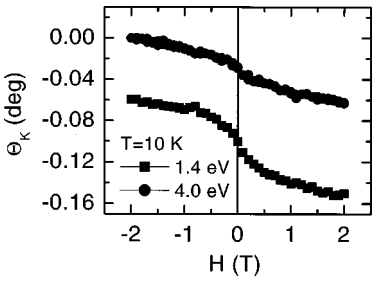

FIG. 2. Out-of-plane (polar) Kerr hysteresis loops of Sr_2FeMoO_6 for photon energies from 1.4 and 4.0 eV at $T=10$ K.

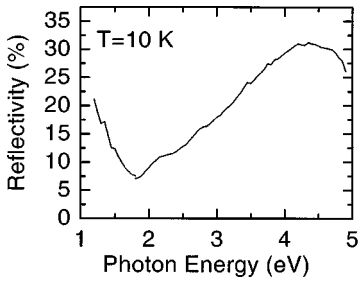

FIG. 4. Reflectivity spectrum of Sr_2FeMoO_6 at $T=10$ K in the photon energy range of 1.2–4.9 eV determined relative to an Al mirror.

RT. A fully automated polar Kerr spectrometer was used to determine the polar Kerr rotation Θ_K, the ellipticity η_K, and the reflectivity spectra in the photon energy range from 1.2 to 4.9 eV. All data were taken relative to an Al mirror. During the Θ_K and η_K measurements, the Sr_2FeMoO_6 films were magnetized to saturation with an out-of-plane field of $B=\pm 1.5$ T in both field directions to eliminate birefringence effects.

In Fig. 3, the polar Kerr rotation spectrum at 10 K shows three maxima of $\Theta_{K1}=-0.07°$, $\Theta_{K2}=-0.04°$, and $\Theta_{K3}=-0.045°$ at photon energies of $E_1=1.6$ eV, near $E_2=4.0$ eV, and at $E_3=4.65$ eV, respectively. The Kramers–Kronig relationship between Θ_K and η_K is obvious, a maximum in Θ_K is connected to a point of inflection in η_K. The first maximum of Θ_K at $E_1=1.6$ eV can be interpreted in terms of an optical enhancement effect due to the presence of a plasma edge in the same energy range.[6,21,22] The reflectivity spectrum in Fig. 4 shows the appearance of a broad minimum around 1.8 eV, which gives rise to the first maximum in Θ_K at 1.6 eV. Generally, for small reflectivities, e.g., near plasma edges, the complex index of refraction $\tilde{n}$ tends towards unity, thus leading to an enhanced Kerr rotation. The complex Kerr rotation can be written as

$$\tilde{\Theta}_K=\Theta_K+\eta_K=\frac{\tilde{\sigma}_{xy}}{\tilde{n}\tilde{\sigma}_{xx}}=i\frac{4\pi}{\omega}\frac{\tilde{\sigma}_{xy}}{\tilde{n}(1-\tilde{n}^2)},$$

where $\tilde{\sigma}_{xy}$, $\tilde{\sigma}_{xx}$, and ω are the complex off-diagonal and diagonal components of the conductivity tensor and the frequency, respectively. For small reflectivities ($\tilde{n}\rightarrow 1$), the complex Kerr rotation $\tilde{\Theta}_K$ tends to exhibit a maximum that is not directly related to a corresponding interband transition energy.

The second maximum at $E_2=4.0$ eV is very broad and can be interpreted as a charge-transfer (CT) transition from the occupied minority-spin O2p states into the strongly hybridized Mo/Fe minority-spin π^* band that crosses the Fermi level. Such a broad peak in the same photon energy range has also been observed in the optical conductivity spectrum determined from reflectivity measurements.[6] At higher photon energies within this broad maximum, a third maximum at $E_3=4.65$ eV appears, which may agree with a CT transition from occupied O-2p majority-spin states into the empty majority-spin Mot_{2g} states. The Fe-e_g minority-spin band is predicted at a higher energy (Fig. 1). In comparison with the transition into the hybridized Mo/Fe minority-spin π^* band, the 4.65 eV peak is significantly sharper as can be expected since the majority-spin Mo-t_{2g} and Fe-t_{2g} bands do not overlap. A diamagnetic Kerr rotation contribution due to Zeeman splitting can be also an alternative explanation for the presence of the sharp peak at 4.65 eV. Nevertheless, this energy region is close to the shortest limit of the MOKE spectrometer. For a more reliable measurement of the polar Kerr rotation and ellipticity the photon energy range has to be extended in this energy region. In the schematic model of the spin-polarized density of states in Fig. 1, the two transitions at 4.0 and 4.65 eV are labeled by an arrow.

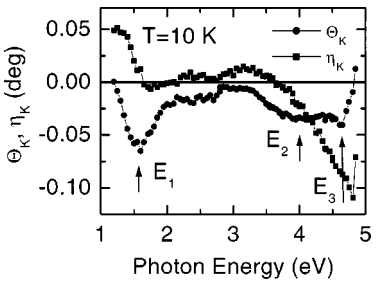

FIG. 3. Polar Kerr rotation Θ_k and elipticity η_k spectra of Sr_2FeMoO_6 at 10 K in the photon energy range from 1.2 to 4.9 eV.

In summary, we have interpreted the polar Kerr rotation spectrum in terms of possible CT transitions that are derived from the spin-polarized density of states. The overall intrinsic polar Kerr rotation is smaller than $|0.045°|$ in the photon energy range investigated. This value is significantly smaller than those observed for other half-metallic ferromagnets.[23,24]

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