OPTICS AND MAGNETO-OPTICS OF SrRuO₃

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Abstract—*Ab initio* magneto-optical (MO) calculations have been carried out for the cubic perovskite SrRuO₃. The results obtained show a good agreement with the presented experimental optical and MO spectra. Features of the MO spectra are explained in terms of the electronic structure and conductivity tensor.

KEYWORD: MAGNETO-OPTICAL SPECTRA, FIRST-PRINCIPLES BAND STRUCTURE CALCULATIONS

INTRODUCTION

Recently there has been a renewed interest in 3d - and 4d - electron transition metal perovskite-based oxides. It has been driven by the discovery of exotic, colossal magnetoresistance and other unusual magnetic properties. Additional interest has been stimulated by the observation of strong magneto-optical Faraday and Kerr effects in SrRuO₃ epitaxial films [1]. These findings have generated widespread research activity, specifically, in understanding the microscopic origin of the high MO effects in SrRuO₃. Semirelativistic band structure calculations of this compound [2-4] have revealed a good agreement between the calculated and measured ground state properties. However several important questions, such as the connection between the electronic structure and the MO spectra of SrRuO₃ remain largely unexplored.

SrRuO₃ has a perovskite-based crystal structure, metallic-like conductivity and ferromagnetic ordering with the Curie temperature $T_C = 160$ K and with the magnetic moment of $1.55 \mu_B$ at 4.2 K in pulsed fields [5]. Actual structure of strontium ruthenate is an orthorhombic GdFeO₃ type having four formula per unit cell. In this article we report the experimental and *ab initio* calculated results of the optical and MO spectra in SrRuO₃. Calculations were performed for an idealized cubic perovskite structure at a lattice parameter of $3.96 \text{ \AA}$.

500 Å thick films of SrRuO₃ were prepared by laser ablation on MgO films with a 1000 Å SrTiO₃ buffer layer. The optical constants have been measured on thicker ($>1000 \text{ \AA}$) films deposited directly on SrTiO₃, using the Sopra spectroscopic ellipsometer, over the range 1.4–4.8 eV. Faraday rotation spectrometer consists of the monochromatized Xe arc lamp, an input polarizer before the sample, the polarizing beam splitter after the sample, set at 45 degrees to the input polarizer, and the balanced photoreceiver. Measurement range was 1.5–3.2 eV.

Present band structure calculations have been carried out by the LMTO method [6] using scalar relativistic MT orbitals and including the spin-orbit coupling (SOC) in the variational procedure. We used a mesh of 238 **k**-points in the magnetic 3/48 cubic Brillouin zone, whereas the basis functions were s, p, d, f LMTO's for Sr and Ru sites, and s, p, d LMTO's for oxygens and empty spheres.

Our calculations of the optical and MO spectra are based on the RPA method for the interband absorptive part of the conductivity tensor (CT) (see, for example, [7,8]). To obtain the dissipative part of the tensor, Kramers-Kronig transformations were used. In addition, we smoothed spectra of the CT with the inverse lifetime $\Gamma=0.2$ eV, and added the intraband Drude contribution to the diagonal components of the CT.

The polar Kerr rotation angle and ellipticity can be obtained from the standard formula [9]:

EXPERIMENTAL DETAILS AND CALCULATIONAL PROCEDURE

$$\theta_K(\omega) = \frac{4\pi}{\omega} \cdot \frac{A_K(\omega)\sigma_{2,xy}(\omega) + B_K(\omega)\sigma_{1,xy}(\omega)}{A_K^2 + B_K^2}, \quad (1)$$

where $A_K = \frac{4\pi}{\omega} \cdot (\sigma_{2,xx}\mathbf{n} - \sigma_{1,xx}\mathbf{k})$, $B_K = \frac{4\pi}{\omega} \cdot (\sigma_{1,xx}\mathbf{n} - \sigma_{2,xx}\mathbf{k})$, $(\mathbf{n}, \mathbf{k})$ are the optical constants.

RESULTS AND DISCUSSION

Fig. 1 shows the LSDA relativistic total and local, l -partial densities of states (DOS).

Fig. 1. Total and local partial DOS for cubic perovskite SrRuO₃. The dashed vertical line denotes E_F .

Note that the Ru4d and O2p bands have close energies causing strong band mixing through a valence band. There is a gap of about 0.3 eV at -2 eV inside the valence band in both spin channels. Above the Fermi level (E_F) we also find a 0.3 eV gap in spin-minority subband, and a quasigap in spin majority channel. Similar result for the region above E_F was obtained in [2] for the actual orthorhombic SrRuO₃. However in [2] there is no gap inside the valence band neither in cubic nor in orthorhombic structures. To check our computations we switched off the SOC and found out that our semirelativistic DOS turned out to be very close to the full-potential LAPW results of Ref. 2 for cubic perovskite. Moreover our semirelativistic paramagnetic DOS also is in good agreement with that of [2] (large peak of DOS slightly, 0.19 meV, above E_F). Large DOS at E_F (142 Ry⁻¹) leads to an itinerant Stoner instability of the paramagnetic state. Interestingly, our relativistic paramagnetic DOS is rather different from that of [2]. Namely, a large peak of DOS is shifted downward in energy to be *below* E_F . The calculated spin magnetic moment of 1.625 μ_B /cell agrees well with experimental one. We find 1.127 μ_B per Ru site,

the other atoms have smaller spin moments, about 0.027 and 0.139 μ_B per Sr and O atom, respectively. Oxygen contribution to the total spin magnetic moment is surprisingly large and seems to be due to the strong Ru-O hybridization near E_F . Such mixing leads to the ferromagnetic direct exchange interaction between Ru and O and to the magnetic ground state.

Our results for the orbital magnetic moments m_O show that m_O is almost quenched ($\approx -0.003 \mu_B$) in SrRuO₃ that is usual thing for nd-based compounds. This fact follows from the strong mutual cancellation of the both spin channels contributions on the Ru site.

Fig. 2. Absorptive part of the diagonal conductivity for SrRuO₃ found from our calculations (solid line - FM state, dashed line - PM state) and measured (circles) for SrRuO₃ films.

Results for the optical spectra agree well with the presented experimental optical data. Calculations reproduce the shape and main features of the measured optical constants, reflectivity, optical conductivity (Fig. 2) and electron energy loss spectrum (EELS).

Note the substantial (≈ 0.8 eV) shift of main absorptive peak upon the para-ferromagnetic transition. Value of this shift is comparable with the exchange band splitting $\Delta E_{\text{exch}} = 0.7$ eV (see Fig. 1). Off-diagonal absorptive parts of the CT (Fig. 3) are somewhat (≈ 0.2 eV) less accurate than the on-diagonal ones.

Fig. 3. Off-diagonal parts of the CT for SrRuO₃. Circles and squares are the experimental data, solid line represents absorptive part, dashed line is dispersive one.

In view of the good agreement between theory and experiment, there is a good chance to study the origin of the main extrema of the polar Kerr and Faraday rotations (Figs. 4 and 5). We found the calculated Kerr rotation of -2.8 deg at 1.7 eV, corresponding to the peak energy of the EELS function and second extrema at 3.7 eV. Calculated Faraday rotation has also two resonances at 2.7 eV and 3.6 eV, being the former in good agreement with experiment.

Fig. 4. Calculated dependence of the polar Kerr effect in SrRuO₃

Fig. 5. Energy dependence of the Faraday rotation in SrRuO₃. Solid and dashed lines are the calculated Faraday rotation and ellipticity, respectively. Circles and squares are the experimental data.

In order to estimate the role of the diagonal and off-diagonal components of the CT, we show in Fig. 6 the spectral dependences of A_K and B_K present in Eq. (1).

Fig. 6. Factors $A_K(\omega)$ and $B_K(\omega)$ appearing in Eq. (1).

Obviously, that these factors are complicated and non-trivial functions of the indices of refraction $\mathbf{n}$ and absorption $\mathbf{k}$. It is seen that in the vicinity of the first MO Kerr resonance energy the product $A_K\sigma_{2,xy}$ vanishes, whereas $B_K\sigma_{1,xy}$ in this energy region has an extrema. The MO resonance at 1.7 eV, therefore, is formed mainly by the maximum of B_K , that is by the diagonal components effect. This enhancement resembles the Feil-Haas plasma edge resonance.

Indeed, our calculations show that $\epsilon_{1,xx} \approx 1$ at 1.7 eV. However $\epsilon_{2,xx} \approx 2$, and the Feil-Haas condition is not realized. At the same time, both the diagonal A_K , B_K and off-diagonal $\sigma_{2,xy}$, $\sigma_{1,xy}$ are effective in formation of the second extrema of θ_K at 3.7 eV. It is interestingly to note that supposed resonance of θ_K at 3.0 eV due to high peak of $\sigma_{2,xy}$ (Fig. 3), is suppressed via vanishing the both products $A_K\sigma_{2,xy}$ and $B_K\sigma_{1,xy}$ at this energy. Two extrema of θ_F and ϵ_F are due to the corresponding extrema of $\sigma_{1,xy}$ and $\sigma_{2,xy}$ (see Fig. 3).

Our numerical experiments show that the Ru and O atoms, as was expected, are dominant in both the effective SOC and matrix elements of current operator. Further information was extracted from the spin decomposition of $\sigma_{2,xy}$ and $\sigma_{1,xx}$. The excitations in spin minority channel play a leading role in formation of abovementioned absorptive parts of the CT. These facts are closely connected with the DOS in SrRuO₃. Strong hybridization of the Ru4d $\downarrow$ and O2p $\downarrow$ states results in the intensive spin minority electronic transitions inside Ru4d $\downarrow$ -O2p $\downarrow$ band. Interband transitions in spin majority channel are weak due to small phase volume of the unoccupied states in Ru4d $\uparrow$ -O2p $\uparrow$ band.

Finally, we conclude that the LSDA theory is quite able to describe the statical response (the ground state properties) as well as the dynamical response (the optical and MO properties) in strontium ruthenate.

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