

## A magnetic Compton scattering study of double perovskite $\text{Sr}_2\text{FeMoO}_6$

This article has been downloaded from IOPscience. Please scroll down to see the full text article.

2004 J. Phys.: Condens. Matter 16 S5717

(<http://iopscience.iop.org/0953-8984/16/48/034>)

View [the table of contents for this issue](#), or go to the [journal homepage](#) for more

Download details:

IP Address: 129.252.86.83

The article was downloaded on 27/05/2010 at 19:20

Please note that [terms and conditions apply](#).

# A magnetic Compton scattering study of double perovskite $\text{Sr}_2\text{FeMoO}_6$

Y Sakurai<sup>1</sup>, A Deb<sup>2</sup>, M Itou<sup>1</sup>, N Hiraoka<sup>3</sup>, A Koizumi<sup>4</sup>, Y Tomioka<sup>5</sup> and Y Tokura<sup>5,6</sup>

<sup>1</sup> Japan Synchrotron Radiation Research Institute (JASRI), SPring-8, 1-1-1 Kouto, Mikazuki, Sayo, Hyogo 679-5198, Japan

<sup>2</sup> Lawrence Berkeley National Laboratory, One Cyclotron Road, MS 70-108B, Berkeley, CA 94720, USA

<sup>3</sup> European Synchrotron Radiation Facility, BP 220, F-38043, Grenoble Cedex, France

<sup>4</sup> University of Hyogo, 3-2-1 Kouto, Kamigori, Ako, Hyogo 678-1297, Japan

<sup>5</sup> Correlated Electron Research Center (CERC), National Institute of Advanced Industrial Science and Technology (AIST), Tsukuba Central 4, Tsukuba, Ibaraki 305-8529, Japan

<sup>6</sup> The University of Tokyo, 7-3-1 Hongo, Bunkyo, Tokyo 113-8656, Japan

Received 19 April 2004, in final form 23 August 2004

Published 19 November 2004

Online at [stacks.iop.org/JPhysCM/16/S5717](http://stacks.iop.org/JPhysCM/16/S5717)

doi:10.1088/0953-8984/16/48/034

## Abstract

The spin-polarized electron momentum distribution (magnetic Compton profile: MCP) of ferromagnetic  $\text{Sr}_2\text{FeMoO}_6$  has been measured using the magnetic Compton scattering technique at room temperature. The experimental MCP is in good agreement with that obtained by a first-principles band-structure calculation. The experiment and calculation reveal a hollow around  $p_z = 0$ , which support the conclusion that there exists a negatively spin-polarized Fe 3d–Mo 4d mixed state.

## 1. Introduction

$\text{Sr}_2\text{FeMoO}_6$  is an ordered double perovskite which possesses an alternating  $\text{FeO}_6$  and  $\text{MoO}_6$  octahedron structure. It has been known as a conducting ferrimagnet with a Curie temperature of 450 K in which the electronic spins on  $\text{Fe}^{3+}$  and  $\text{Mo}^{5+}$  ions couple antiferromagnetically [1–4]. Recently, a large tunnelling magnetoresistance (TMR) effect has been observed at room temperature, which has aroused much interest in  $\text{Sr}_2\text{FeMoO}_6$  and its related compounds as a possible material for electromagnetic devices [5]. First-principles band-structure calculations have predicted a half-metallic electronic state: the up-spin band forms a gap across the Fermi level ( $E_F$ ), while an Fe 3d–Mo 4d mixed state in the down-spin band contributes to the density of states at  $E_F$  [5–7].

Magnetic Compton scattering (MCS) probes uniquely the spin component of magnetization in a sample [8, 9]. When the incident x-rays are circularly polarized, the Compton scattering cross section contains a spin dependent term. The experimental extraction

of the spin dependence results in a magnetic Compton profile (MCP),  $J_{\text{mag}}(p_z)$ , that is defined as the one-dimensional projection of the spin-polarized electron momentum distribution:

$$J_{\text{mag}}(p_z) = \int \int [n_{\text{up}}(\mathbf{p}) - n_{\text{down}}(\mathbf{p})] dp_x dp_y, \quad (1)$$

where  $n_{\text{up}}(\mathbf{p})$  and  $n_{\text{down}}(\mathbf{p})$  are the momentum densities of the up-spin and down-spin bands, respectively. The electron momentum  $p_z$  in atomic units (au) is determined by the following equation:

$$p_z = mc \frac{\omega_2 - \omega_1 + \omega_1 \omega_2 (1 - \cos \theta) / mc^2}{\sqrt{\omega_1^2 + \omega_2^2 - 2\omega_1 \omega_2 \cos \theta}}, \quad (2)$$

where  $\omega_1$  is the incident x-ray energy,  $\omega_2$  is the Compton scattered x-ray energy,  $\theta$  is the scattering angle,  $m$  is the rest mass of the electron, and  $c$  is the velocity of light. The area under the MCP is equal to the total spin moment ( $\mu_{\text{spin}}$ ) per formula unit:

$$\int_{-\infty}^{+\infty} J_{\text{mag}}(p_z) dp_z = \mu_{\text{spin}}. \quad (3)$$

Equations (1)–(3) are valid under an impulse approximation [10], and the errors due to the approximation are within the statistical errors under the present experimental conditions.

In this paper, we report an MCS study on poly-crystalline  $\text{Sr}_2\text{FeMoO}_6$ . Results on a single-crystalline sample and complementary discussion will be presented in a separate paper [11].

## 2. Experiment and calculation

A powder sample was obtained by carefully grinding a single crystal of  $\text{Sr}_2\text{FeMoO}_6$ . The MCS experiments were carried out on the high energy inelastic scattering beamline (BL08W) at SPring-8 [12]. The incident x-ray energy was 175 keV, and the scattering angle was  $178.5^\circ$ . An MCP was measured at room temperature with a momentum resolution of 0.5 atomic units (au). The applied magnetic field was 2.5 T.

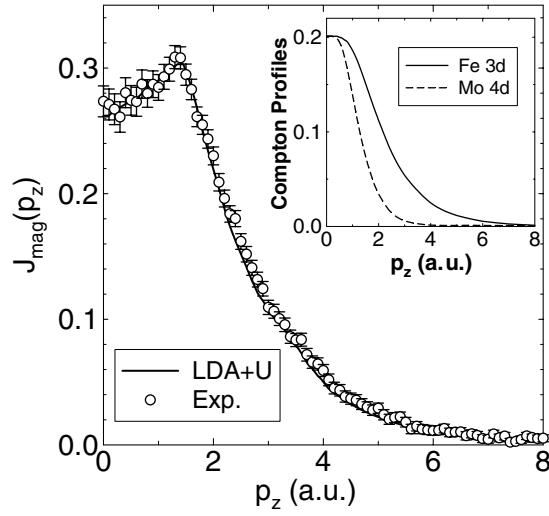
A band-structure calculation was performed with the FLAPW method within the LDA +  $U$  scheme. The computational codes was originally developed by Kodama *et al*<sup>7</sup>. For effective Coulomb repulsions  $U_{\text{eff}}$ , 2.0 eV for Fe and 1.0 eV for Mo were adopted. The exchange–correlation potential was given by Vosko *et al* [13]. The cubic lattice parameters determined experimentally [14] were used for the present computations. An isotropic MCP and bandwise Compton profiles were calculated from obtained wavefunctions, where a mesh of 128  $k$  points in the irreducible Brillouin zone was used for the  $k$ -space integration.

## 3. Results and discussion

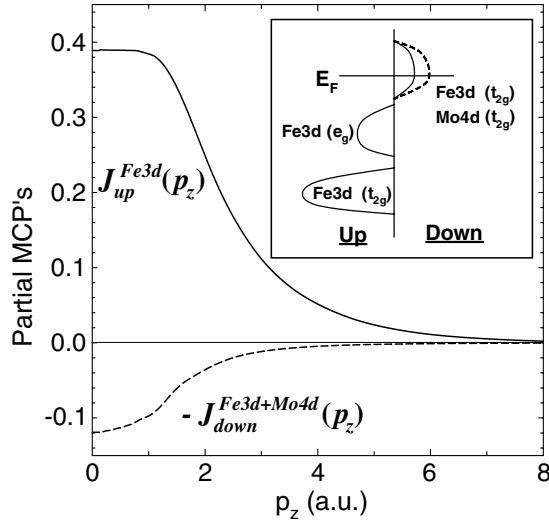
Figure 1 shows the experimental MCP (open circles), together with that obtained by the LDA +  $U$  calculation (solid curve). The areas under both MCPs are normalized to  $1.75 \mu_{\text{B}}$ , which is the value determined by a magnetization measurement. The experiment and calculation show good agreement on the overall shape of the profiles, and reveal a hollow around  $p_z = 0$ . Until now, hollow structures have been observed in the MCPs of 3d metals and alloys, and are well accounted for by the fact that the spin-polarization of itinerant s, p-like electrons is antiparallel to that of the 3d band electrons [9]. In order to examine if a similar scenario will work in  $\text{Sr}_2\text{FeMoO}_6$ , the calculated MCP is divided into two parts (see figure 2):

$$J_{\text{mag}}(p_z) = J_{\text{up}}^{\text{Fe } 3d}(p_z) - J_{\text{down}}^{\text{Fe } 3d + \text{Mo } 4d}(p_z), \quad (4)$$

<sup>7</sup> The computing code BANDS01 was supplied from Fuji Research Institute Co., Ltd.



**Figure 1.** The magnetic Compton profile of polycrystalline Sr<sub>2</sub>FeMoO<sub>6</sub> (open circles) compared with that of the LDA + *U* calculation (solid curve). The inset shows atomic Fe 3d and Mo 4d Compton profiles [15].



**Figure 2.** The partial MCPs for the up-spin Fe 3d states and the down-spin Fe 3d–Mo 4d mixed state. The inset shows a schematic electronic structure in Sr<sub>2</sub>FeMoO<sub>6</sub>.

and

$$J_{\text{up}}^{\text{Fe 3d}}(p_z) = J_{\text{up}}^{1\text{st}-39\text{th}}(p_z) - J_{\text{down}}^{1\text{st}-33\text{rd}}, \quad (5)$$

$$J_{\text{down}}^{\text{Fe 3d+Mo 4d}}(p_z) = J_{\text{down}}^{34\text{th}-36\text{th}}(p_z), \quad (6)$$

where  $J_{\text{up}}^{1\text{st}-39\text{th}}(p_z)$  is the bandwise profile for all (1st–39th) up-spin bands, and  $J_{\text{down}}^{1\text{st}-33\text{rd}}(p_z)$  and  $J_{\text{down}}^{34\text{th}-36\text{th}}(p_z)$  are those for perfectly filled (1st–33rd) and partially filled (34th–36th) down-spin bands. The present calculation shows that all of the up-spin bands (1st–39th) are perfectly filled and are dominated by Fe 3d  $e_g$  and  $t_{2g}$  states and O 2p states, while the perfectly filled

down-spin bands (1st–33rd) are dominated by O 2p states. Therefore, the first term,  $J_{\text{up}}^{\text{Fe}3\text{d}}(p_z)$ , reflects the up-spin Fe 3d states since the contribution from the down-spin O 2p states almost cancels out that from the up-spin O 2p states. This is confirmed by the fact that the profile shape is close to that of an atomic 3d profile [15] especially at high momenta (see the inset of figure 1). The first term dominates the MCP, but does not have any hollow-like feature. The second term,  $J_{\text{down}}^{\text{Fe}3\text{d}+\text{Mo}4\text{d}}(p_z)$ , is the bandwise profile for the partially filled (34th–36th) down-spin bands. The bands are dominated by Fe 3d and Mo 4d  $t_{2g}$  states. The full width at half maximum of the profile (1.56 au) is between those of the atomic Fe 3d profile (2.17 au) and Mo 4d profile (1.28 au) [15], which is consistent with the feature of the Fe 3d–Mo 4d mixed state. The mixed state is a minor part, but its spin polarization is antiparallel to that of the up-spin Fe 3d bands. The sum of two partial profiles forms the hollow structure in the MCP. In short, the presence of the Fe 3d–Mo 4d mixed state in the down-spin band is essential to account for the hollow structure.

In summary, the experiment and calculation reveal a hollow-like structure around  $p_z = 0$ . The bandwise analyses of the calculated MCP support a model that the Fe 3d–Mo 4d mixed state is antiparallel to the up-spin Fe 3d states. The good agreement on the overall shape of the MCP between the experiment and calculations leads to the conclusion that there exists a negatively spin-polarized Fe 3d–Mo 4d mixed state in  $\text{Sr}_2\text{FeMoO}_6$ .

### Acknowledgments

This work was supported in part by a Grant-in-Aid for Scientific Research from the Ministry of Education, Culture, Sports, Science and Technology of Japan, and was performed with the approval of JASRI (proposal No 2004A5008-LD3-np).

### References

- [1] McQuarrie M 1955 *J. Am. Ceram. Soc.* **38** 444
- [2] Glasso F S, Douglas F C and Kasper J 1966 *J. Chem. Phys.* **44** 1672
- [3] Nakayama S, Nakagawa T and Nomura S 1968 *J. Phys. Soc. Japan* **24** 219
- [4] Itoh M, Ohta I and Inaguma Y 1996 *Mater. Sci. Eng. B* **41** 55
- [5] Kobayashi K I, Kimura T, Sawada H, Terakura K and Tokuta Y 1998 *Nature* **395** 667
- [6] Saitoh T, Nakatake M, Kakizaki A, Nakajima H, Moritomo O, Xu Sh, Moritomo Y, Hamada N and Aiura Y 2002 *Phys. Rev. B* **66** 035112
- [7] Kang J-S, Kim J H, Sekiyama A, Kasai S, Suga S, Han S W, Kim K H, Muro T, Saitoh Y, Hwang C, Olson C G, Park B J, Lee B W, Shim J H, Park J H and Min B I 2002 *Phys. Rev. B* **66** 113105
- [8] Cooper M J 1985 *Rep. Prog. Phys.* **218** 415
- [9] Sakai N 1996 *J. Appl. Crystallogr.* **29** 81
- [10] Platzman P M and Tzoar N 1965 *Phys. Rev. A* **139** 410
- [11] Deb A, Hiraoka N, Itou M, Sakurai Y, Koizumi A, Tomioka Y and Tokura Y 2000 *Phys. Rev. B* at press
- [12] Sakurai Y 1998 *J. Synchrotron Radiat.* **5** 208
- [13] Vosko S H, Wilk L and Nusair M 1980 *Can. J. Phys.* **58** 1200
- [14] Chamaisssem O, Kruk R, Dabrowski B, Brown D E, Xiong X, Kolesnik S, Jorgensen J D and Kimball C W 2000 *Phys. Rev. B* **62** 14197
- [15] Biggs F, Mendelsohn L B and Mann J B 1975 *At. Data Nucl. Data Tables* **16** 201