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Report:

In the search for new spintronic materials with high spin-polarization at room-temperature, we have synthesized an osmium based double perovskite with a Curie-temperature of 725 K. Our combined experimental results confirm the existence of a sizable induced magnetic moment at the Os site, in agreement with a proposed kinetic energy driven mechanism of ferrimagnetism in these compounds. The intriguing property of Sr_2CrOsO_6 is that it has at the same time the highest Curie-temperature in its family, and is also an almost fully spin-polarized semiconductor.

We have investigated the Os 5d spin and orbital magnetic moments in the ferrimagnetic double perovskite Sr_2CrOsO_6 by X-ray magnetic circular dichroism (XMCD) at the $L_{2,3}$ edges. In fair agreement with recent band-structure calculations, at the Os site a large 5d spin magnetic moment of $-0.17\,\mu_{\rm B}$ and a considerable orbital moment of $+0.15\,\mu_{\rm B}$ at room temperature are detected. One important result of our study is that the Curie temperature of the ferrimagnetic double perovskites scales with the spin magnetic moment of the 'non-magnetic' atom. The most important result however is, that actually Sr_2CrOsO_6 seems to belong to a new class of material, i.e. a half-metallic semiconductor.

The spectra were recorded using the total fluorescence yield detection mode. The XMCD spectra were obtained as direct difference between consecutive XANES scans (X-ray Absorption Near Edge Spectrum) recorded with opposite helicities of the incoming X-ray beam. To ensure that the XMCD spectra are free from experimental artefacts the data was collected for both directions of the applied magnetic field of 6 T (parallel and antiparallel to the X-ray beam). The measurements were performed at about 280 K. Since the samples measured in backscattering geometry were very thick, the spectra were corrected for self-absorption effects. The edge jump ratio L_3/L_2 was normalized to 2.20/1. This takes into account the difference in the radial matrix elements of the 2p1/2 to $5d(L_2)$ and 2p3/2 to $5d(L_3)$ transitions.

One specialty of the measurement using Sr_2CrOsO_6 was that it could not be done at low temperatures. The reason is, that the sample had to be frozen in a magnetic field in $LiNO_3$ in a quartz tube because of the enormously high anisotropy field of about $40\,\mathrm{T}$. After breaking the tube, probably the high sample magnetization at low temperatures lead a loss of sample volume.

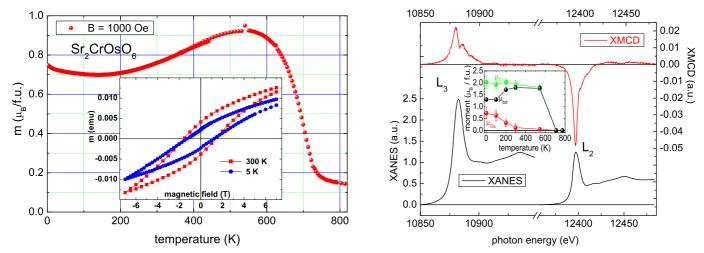


Figure 1: Left: SQUID data shows that Sr₂CrOsO₆ is the ferrimagnetic double perovskite with the highest Curie temperature discovered so far. Right: XANES and XMCD spectra for Sr₂CrOsO₆ at 280 K. The inset shows the magnetic moments as determined from neutron scattering.

We now discuss the XANES spectra of Sr_2CrOsO_6 shown in the right figure. A very clear XMCD signal is observed even at room-temperature, signaling a strong magnetic moment at the Os-site opposite to the net magnetization. The experimentally determined value is in good agreement with neutron scattering results. Neutron scattering gives an overall moment of -0.12 at the Os site, compared to -0.155 from the XMCD. It is important, that qualitatively the induced magnetic moment is opposite to the magnetic moment at the Cr site. It is interesting to note, that for Os in the XANES spectrum no indication of the crystal field splitting is present. The detailed comparison of 5d elements in the 5+ valence state will be a future project.

A key result of accompanying band-structure calculations is that Sr_2CrOsO_6 is, due to the filled Os t_{2g} orbitals, instead of a half-metal a half-semiconductor. This is also in agreement with preliminary conductivity measurements on the powder samples. Therefore, in the system $Sr_2CrRe_{1-x}Os_xO_6$ one can tune the degree of spin-polarized conductivity from half-metallicity to half-semiconductivity without a reduction (even with an increase) in T_C .

In summary, based on our XMCD measurements we (i) confirm a scaling law between the critical temperature of the ferrimagnetic double perovskites and the magnetic moment at the 'non-magnetic' site; (ii) a new type of spintronics material has been discovered, following the same mechanism as the already known ferrimagnetic double perovskites, but in a tunable conductivity range.

Based on the recent publications coming out from the last measurement time ((i) P. Majewski, S. Geprägs, A. Boger, M. Opel, A. Erb, R. Goss, G. Vaitheeswaran, V. Kanchana, A. Delin, F. Wilhelm, A. Rogalev, and L. Alff, Phys. Rev. B **72**, 132402 (2005); and (ii) P. Majewski, S. Geprägs, O. Sanganas, M. Opel, R. Gross, F. Wilhelm, A. Rogalev, and L. Alff, Appl. Phys. Lett. **87**, 202503 (2005).) one more publication has now been published as a Rapid Communication in Physical Review B:

[1] Y. Krockenberger, K. Mogare, M. Reehuis, M. Tovar, M. Jansen, G. Vaitheeswaran, V. Kanchana, F. Bultmark, A. Delin, F. Wilhelm, A. Rogalev, A. Winkler, and L. Alff, Phys. Rev. B **75**, 020404(R) (2007).