| ESRF | **Experiment title:** Magnetic polarization of W in $A_2CrWO_6$ double perovskites ($A$ = Ca, Sr, and Ba) | **Experiment number:** HE-1658 |
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Report:

We have investigated the magnetic structure of polycrystalline samples of the double perovskites $Sr_2CrWO_6$ and $Ca_2CrWO_6$ by x-ray magnetic circular dichroism (XMCD) at the $L_{2,3}$ edges of W sites. The observed spin magnetic moment on W in $Sr_2CrWO_6$ is about -0.3 $\mu_B$. This result confirms the ferrimagnetic ordering of the compound which has been similarily found in the case of $Sr_2FeMoO_6$. The finite induced moment at the W site invalidates the picture that the ferromagnetic order of the Cr ions is due to the antiferromagnetic coupling to the charge carriers supplied by the W ion. The one $s$ electron of the W site is shared between W an the other sites. In comparison to the spin magnetic moment, the orbital magnetic moments are almost negligible. This shows that spin-orbit coupling which potentially could spoil the predicted half-metallicity of the investigated double-perovskites, does not play an important role.

XANES (x-ray absorption near edge structure) and XMCD measurements were performed at the $L_{2,3}$ edges as these edges have a rich fine structrue sensitive to the valency and the spin state of the probed atom. This information in turn is related to the crystal field and exchange splittings. The spectra shown in Figure 1 are measured on polycrystalline samples of $Sr_2CrWO_6$ taken at 300 K and for $Ca_2CrWO_6$ taken at 5 K. It is clear that there is a dichroic signal on the W site giving evidence for the existence of an induced moment. For the evaluation of the data we have assumed that the number of $d$-holes is $n_h \approx 6.3$. For the Sr compound, -0.3 $\mu_B$ is obtained for the W spin moment which is almost identical to the moment of Mo in $Sr_2FeMoO_6$.

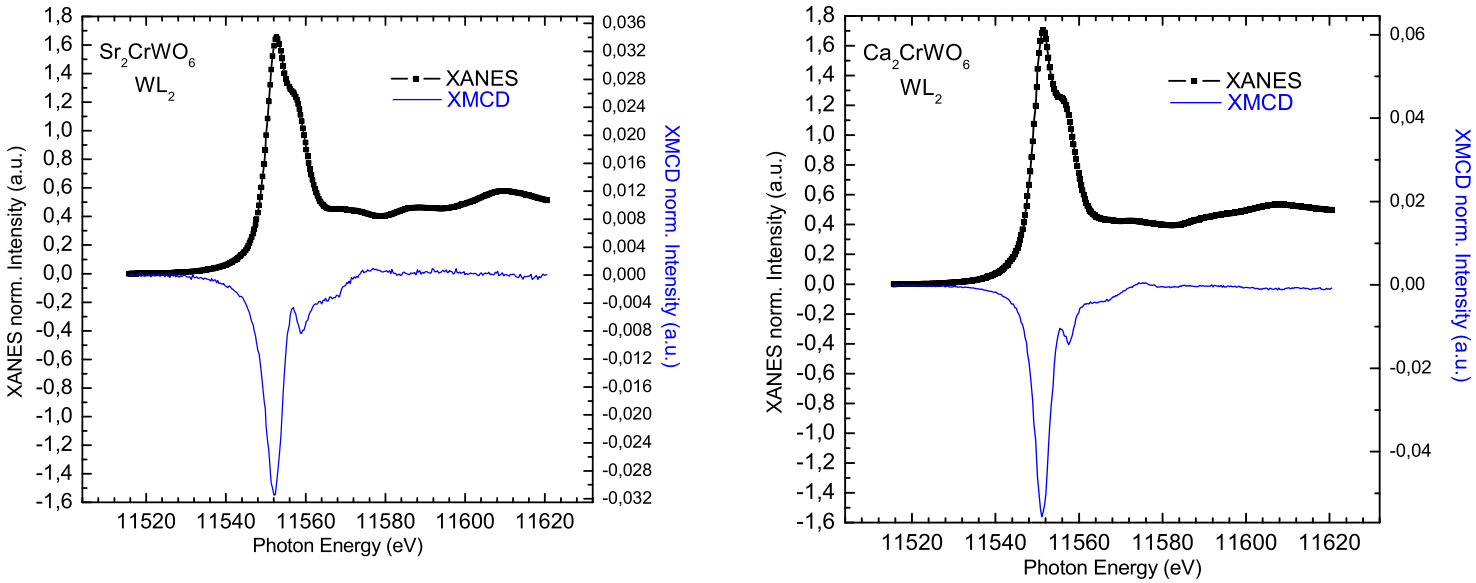


**Figure 1**: XANES and XMCD spectra for $Sr_2CrWO_6$ at 300 K (left) and for $Ca_2CrWO_6$ at 5 K (right).

If Sr is replaced by Ca the structure of the compound changes from cubic to monoclinic. The change of the bonding angles then also implies a change of the magnetic structure. As the electron on the W site is less delocalized, one expects also a reduced moment at the W site in $Ca_2CrWO_6$. This is indeed observed ($\sim -0.2\,\mu_B$). Correspondingly the critical temperature of $Ca_2CrWO_6$ is clearly reduced compared to $Sr_2CrWO_6$. This result is important as it gives first evidence that the magnetic ordering temperature in double perovskites is linked to the induced magnetic moment at the non-magnetic site. It is clear that this relationship has to be established in more experiments including also different elements at the non-magnetic site.

Preliminary band structure calculations by G. Vaitheeswaran *et al.* using the full-potential linear muffin-tin orbital method gives surprisingly good agreement with the experimental data. The calculation also supports the half-metallic nature of the ferromagnetic CrW double perovskites. Note that effects of antisite disorder have not been taken into account.

It was also intended to measure La-doped $Sr_2CrWO_6$ that was found to have reduced $T_C$ (in contrast to La-doped $Sr_2FeMoO_6$). However, it has been found that this behavior is an extrinsic effect due to the formation of $LaCrO_3$. Corresponding thin film had in the first try a too small signal. In principal this would be the way to study the effect of La-doping. Here the correspondence of increased $T_C$ and increased spin magnetic moment at the W-site would be further evidence for the correspondence of both quantities.

A publication with the more complete and detailed analysis of the data is currently under work.