

**Experiment title:**Magnetic circular dichroism in soft X-ray emission of
3d transition metal materials

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Report:

We have studied the influence of excitation by circularly polarized x-rays on the L_{α} and L_{β} x-ray emission spectra of manganese ferrite ($(\text{Mn,Zn})\text{Fe}_2\text{O}_4$) and of manganese arsenide (MnAs). Manganese ferrite is the 3d compound where one expects the largest magnetic effects in fluorescence, because Mn^{2+} ions contains no minority $3d$ electrons. MnAs is metallic and more complicated, probably partly low-spin with a moment of about 3 Bohr magnetons.

Circularly polarized light produces spin-polarized photoelectrons from levels that are split by spin-orbit interaction. Suppose right-handed light gives spin-down electrons from a $2p_{3/2}$ level. If the sample is magnetized, absorption at the L_3 threshold will be larger because these spin-down electrons can find place in the unoccupied $3d$ levels. This is the origin of magnetic circular dichroism in a single particle picture. Above threshold the difference for exciting spin-up or spin-down electrons disappears and the absorption is equal for both polarizations. But the fluorescence rates are different because the core hole is spin-polarized. It is easier to fill up the hole left behind by a spin-up photoelectron than that left by a spin-down electron. So the L_{α} fluorescence is stronger for the polarization that had the smaller absorption at the L_3 threshold.

Fluorescence spectra excited above the $2p$ binding energies are shown in the top traces of the figure. The $L\alpha$ and the $L\beta$ peak show a difference in intensity of the expected sign. The size of the difference is larger than for metallic iron, but smaller than an estimate based on simple considerations. It is difficult to compare the results with a spin-resolved density of states. The Mn $3d$ DOS would be composed of two narrow bands (e_g and t_{2g}) separated by a ligand field splitting of about 1 eV. However, the observed spectra are approximately 5 eV wide.

The other traces show the sharp spectra that are observed when the excitation energy is at two different energies in the L_3 absorption peak. One peak coincides with the excitation energy. It is the recombination peak, where the atom ends up in its electronic $6S$ ground state. But the excited atom can also decay to quartet or doublet final states. The fluorescence energy is then lower than the excitation energy, the difference being the energy for dd -excitation. The lowest excited state of Mn^{2+} is the $4G$ state, 3 eV above the ground state. The resonant x-ray fluorescent spectra are equivalent with electronic Raman spectra of dd - and charge-transfer excitations. A recent study of MnO [Butorin *et al*, Phys. Rev. B 54, 4405 (1996)] showed that the intensities of the dd -excitations can be obtained from atomic calculations of the decay of the intermediate state. We are confident that including circular polarization in these calculation will reproduce the resonant data of manganese ferrite.

In $MnAs$ the magnetic differences in the $L\alpha$ and $L\beta$ spectra excited by continuum excitation were slightly smaller, which would be consistent with minority electrons in the ground state. The $L\alpha/L\beta$ ratio is much larger than in the ferrite. On resonance, the elastic peak is weaker. In this metal, there can be no gap between the ground state and excited states. The shape of the spectra depends less on circular polarization than for manganese ferrite.

