Novel Complex Magnetic Materials and Devices

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Through the development of novel complex oxide and chalcogenide materials, we have been able to probe spin-based phenomena in heterostructures where the interface quality can be controlled. In this highlight, we focus on a novel spin polarized material, the complex chalcogenide, $CuCr_2Se_4$. The success of this project is possible only through a close collaboration among experimentalists and theorists with expertise in solid state chemistry, thin film synthesis and electronic structure calculations.

The calculated electronic structure of the complex chalcogenide, $CuCr_2Se_4$ is shown in figure 1. The large difference in the density of states for the majority and minority suggests that it is highly spin polarized. We have shown that the Cr DOS is highly spin-polarized with a magnetic moment of approximately $3\mu_B$. The magnetic moment per formula unit, however, is only about 5 μ_B because both the Cu sites (-0.12μ_B) and the Se (-0.18 to -0.20 μ_B) sites acquire small magnetic polarizations opposite to that of the Cr.

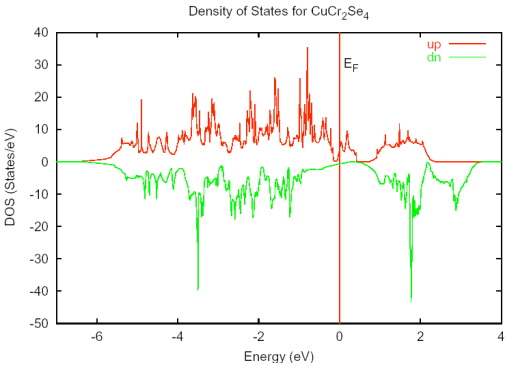

Figure 1. Calculated density of electronic states for $CuCr_2Se_4$.

We have adapted the chemical vapor transport synthesis technique to prepare single crystals of halogen-doped and undoped chalcogenide spinels based on $CuCr_2Se_4$. We report the discovery of a new AB_2X_3Y structure variation when $CuCr_2Se_4$ spinel is doped with chlorine. The new structure differs from the parent compound by ordered substitution of the selenide anion by the chlorine dopant; the overall symmetry of the structure is lowered to rhombohedral and the resultant material is uniaxial. We have extended the synthesis to bromine and iodine doping and found that the rhombohedral distortion is general to $CuCr_2Se_{(4-x)}Y_x$ (Y = Cl, Br, I, 0< x < 0.3). The new phase is ferrimagnetic at room temperature for all compositions studied, and the magnetic moment increases with halogen doping level, although the total moments are lower than the undoped parent phase.

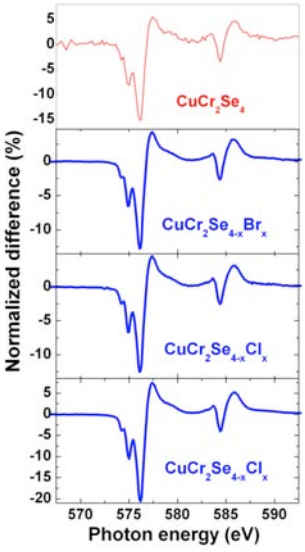

Figure 3: XMCD spectra of the Cr edge of a. thin film of $CuCr_2Se_4$ (red); single crystal of $CuCr_2Se_{4-x}Br_x$ using transport agent $SeBr_4$; single crystal of $CuCr_2Se_{4-x}Cl_x$ using transport agent $CrCl_3$; single crystal of $CuCr_2Se_{4-x}Cl_x$ using transport agent $SeCl_4$.

We have used sintered ceramic pellets of the $CuCr_2Se_4$ in a pulsed laser deposition chamber to synthesize epitaxial thin films. We have probed the structural, chemical, and magnetic data of both the bulk and the surface of the film. Using x-ray diffraction, we have determined that we have grown crystalline thin films of the spinel $CuCr_2Se_4$ with the (111) orientation. Magnetic measurements have shown the film magnetization to be near the ideal value of $5\mu_B$/formula unit at low temperature. Additionally, we have probed the film surface using x-ray absorption spectrometry (XAS) and x-ray magnetic circular dichroism (XMCD). Our XAS (figure 2) lineshapes of the thin film compare well to Cl- doped single crystals, and XMCD (figure 3) lineshapes of the thin film Cr-edge compares well with Br- and Cl-doped single crystals grown using a range of transport agents, as well as published data[1] of Cr in the octahedral environment of $CuCr_2Se_4$.

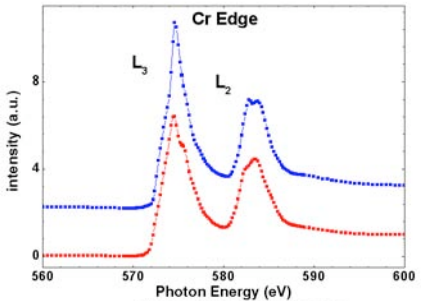

Figure 2: XAS spectra of Cr edge in a Cl-doped single crystal (blue) and a thin film of $CuCr_2Se_4$ (red)

To our knowledge, these results are the first describing successful synthesis of $CuCr_2Se_4$ films. The successful growth of these thin films is extremely promising for the development of an entirely new class of spin polarized materials based on complex chalcogenides.

[1] Kimura *et al*, *Phys. Rev. B* , **63,** 22 (2001)