

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 $5\mu_B$ because both the Cu sites ($-0.12\mu_B$) and the Se (-0.18 to $-0.20\mu_B$) sites acquire small magnetic polarizations opposite to that of the Cr.

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 $\text{AB}_2\text{X}_3\text{Y}$ 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

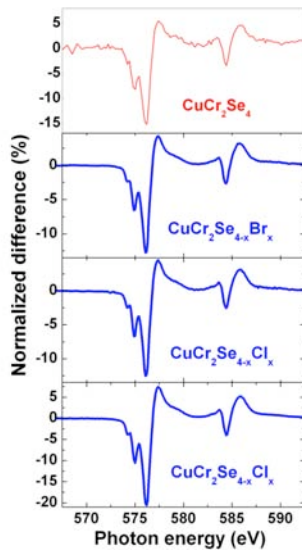


Figure 3: XMCD spectra of the Cr edge of a. thin film of CuCr_2Se_4 (red); single crystal of $\text{CuCr}_2\text{Se}_{4-x}\text{Br}_x$ using transport agent SeBr_4 ; single crystal of $\text{CuCr}_2\text{Se}_{4-x}\text{Cl}_x$ using transport agent CrCl_3 ; single crystal of $\text{CuCr}_2\text{Se}_{4-x}\text{Cl}_x$ using transport agent SeCl_4 .

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 $\text{CuCr}_2\text{Se}_{(4-x)}\text{Y}_x$ ($\text{Y} = \text{Cl}, \text{Br}, \text{I}, 0 < x < 0.3$). 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.

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 as well as published data[1] of Cr in the octahedral environment of CuCr_2Se_4 .

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.

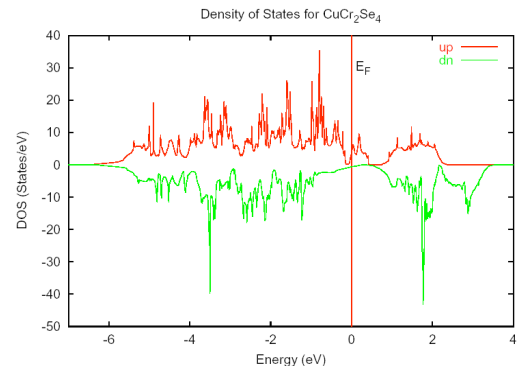


Figure 1. Calculated density of electronic states for CuCr_2Se_4 .

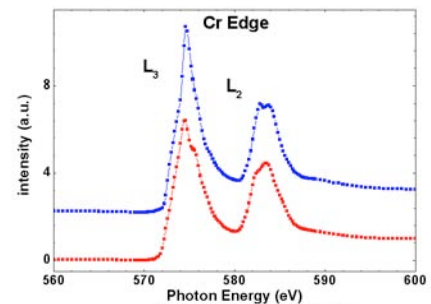


Figure 2: XAS spectra of Cr edge in a Cl-doped single crystal (blue) and a thin film of CuCr_2Se_4 (red)

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