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Authors

Liberati, M.
Bettinger, J.
Chopdekar, R.
et al.

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New materials for spintronics: CuCr_2Se_4

Marco Liberati^{1,2}, Janell Neulinger³, Rajesh Chopdekar^{4,5}, Elke Arenholz², Yuri Suzuki⁴, Angelica M. Stacy³, Yves U. Idzerda¹

¹Department of Physics, Montana State University, Bozeman, MT 59717, USA

²Advanced Light Source, Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA

³Department of Chemistry, UC Berkeley, Berkeley, CA 94720, USA

⁴Department of Materials Science and Engineering, UC Berkeley, Berkeley, CA 94720, USA

⁵Department of Applied Physics, Cornell University, Ithaca, NY 14853, USA

One of the leading fields in magnetism nowadays is the research of new spin polarized materials for spintronics applications. CuCr_2Se_4 can be considered one of them being a highly spin-polarized chalcogenide spinel of interest for magneto tunnel junction [1] as its electrical conductivity and magnetic properties can be chemically tuned by substitution of halogen atoms onto the anion site. In this work we present our data on CuCr_2Se_4 single crystals structurally and magnetically characterized as a function of different SeCl_4 doping level.

Both crystal growth and halogen doping have been obtained by means of the chemical vapor transport technique (CVT) and using powdered Cu, Cr, Se and SeCl_4 as starting materials. Electron microprobe measurements of the crystal and powder compositions (Table 1) show that the doping level depends on the SeCl_4 concentration during growth but saturates at relatively low doping.

Single crystal X-ray diffraction measurements show a rhombohedral structure for the chlorine-doped $\text{CuCr}_2\text{Se}_{4-x}\text{Cl}_x$ single crystals where ordered substitution of chloride onto selenide sites reduces the symmetry compared to the cubic CuCr_2Se_4 (Fig. 1). The resulting structure has two octahedral Cr environments (shown in blue) and a pseudo-tetrahedral Cu coordination (yellow). Each Cu tetrahedron consists of one bond to a disordered Se/Cl site and three bonds to sites occupied only by selenium. Similarly, one of the Cr environments comprises six selenium-only sites while the other environment includes a pair of sites, *trans* to one another, which have disordered occupation by selenium and chlorine dopant. These disordered sites are indicated by red spheres in Fig. 1. Bulk magnetic measurements in order to extract the saturation magnetic moment and the Curie temperature (T_C) as a function of the doping level have been performed respectively by SQUID magnetometry at 10K and vibrating sample magnetometry (Table 1). While the saturation magnetic moment increases with the doping level, the T_C falls off to lower temperatures with higher concentrations of the chlorine dopant. Surface and element sensitive magnetic measurements have been performed by X-ray circular magnetic dichroism (XMCD). While the lineshape of the Cr $L_{2,3}$ absorption edges (top of Fig. 2) does not change as a function of the different SeCl_4 doping level, the dichroism (bottom of Fig. 2, normalized here to the max intensity of the absorption at the L_3 edge) shows an increasing magnitude as the doping level increases, in agreement with the SQUID bulk measurements. A smaller dichroism signal (Fig. 3) has also been detected for the Cu at the $L_{2,3}$ edges in agreement with previous results [1]. The small induced moment on the Cu is aligned antiparallel to the Cr moment and also increases in magnitude with doping level. Halogen doped CuCr_2Se_4 single crystals have been also grown with other transport agents (CrCl_3 and SeBr_4). XAS and XMCD measurements show comparable lineshapes to crystals grown by SeCl_4 .

In conclusion we have synthesized a new structural variation of CuCr_2Se_4 by using the SeCl_4 transport agent. Both bulk and surface magnetic measurements are in agreement in measuring an enhancement in the saturation moment as the doping level is increased. On the other hand, a decrease in T_C is observed for increasing Cl doping.

SeCl_4 (mg cm ⁻³)	Doping level (x)	T_C (°C)	Saturation moments (μ _B /f.u.)
0.00	0.00	157 [2]	-
1.30	0.13	147	0.60
2.66	0.29	133	2.90
4.44	0.28	-	2.25

Table 1: Doping level x in $\text{CuCr}_2\text{Se}_{4-x}\text{Cl}_x$, T_C and saturation moments as a function of SeCl_4 concentration during synthesis and crystal growth.

Fig. 1: Crystal structure of $\text{CuCr}_2\text{Se}_{4-x}\text{Cl}_x$.

Fig. 2: XAS and XMCD at the Cr $L_{2,3}$ edges for different chlorine-doped $\text{CuCr}_2\text{Se}_{4-x}\text{Cl}_x$ single crystals.

Fig. 3: XMCD at the Cu $L_{2,3}$ edges for different chlorine-doped $\text{CuCr}_2\text{Se}_{4-x}\text{Cl}_x$ single crystals.

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