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Photoemission, soft x-ray absorption, and magnetic circular dichroism spectroscopy study of $Fe_{1-x}Cu_xCr_2S_4$ ($0.1 \leqslant x \leqslant 0.5$) spinel sulfides

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Abstract

The electronic and magnetic structures of $Fe_{1-x}Cu_xCr_2S_4$ ($0.1 \leqslant x \leqslant 0.5$) spinel sulfides have been investigated systematically by performing photoemission spectroscopy (PES), soft x-ray absorption spectroscopy (XAS), and soft x-ray magnetic circular dichroism (XMCD) measurements using synchrotron radiation. Cr and Cu ions are found to be nearly trivalent (Cr^{3+}) and monovalent (Cu^{+}), respectively, and their valence states do not change with x. The Fe 2p XAS spectra of $Fe_{1-x}Cu_xCr_2S_4$ are very similar to that of Fe metal, indicating that the Fe 3d electrons are strongly hybridized to other valence electrons. The Fe and Cr 2p XMCD spectra show that the magnetic moments of Cr ions and Fe ions are aligned antiparallel to each other and that both the Cr and Fe magnetic moments increase with increasing x. The valence-band PES study reveals that the Cr^{3+} ($t_{2g}^3 \downarrow$) 3d states are located at $\sim$1.5 eV below E_F. The occupied Fe 3d states consist of the broad $t_{2g}^3 \uparrow$ states, the $e_g^2 \uparrow$ states at $\sim$4 eV below E_F, and the $e_g \downarrow$ states very close to E_F. The filled Cu $3d^{10}$ states lie at $\sim$2.5 eV below E_F. This study suggests that the hybridized Fe $e_g \downarrow$ and S 3p states near E_F play an important role in determining the transport properties of $Fe_{1-x}Cu_xCr_2S_4$ for $x \leqslant 0.5$.

(Some figures in this article are in colour only in the electronic version)