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Mössbauer Studies of Abnormal Relaxation Phenomena on Copper Doped Sulphur Spinel

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Samples of $\text{Fe}_{1-x}\text{Cu}_x\text{Cr}_2\text{S}_4$ (x = 0.0, 0.1, 0.3, and 0.5) have been studied by using Mössbauer spectroscopy, X-ray diffraction, magnetization, and magnetoresistance (MR). Neutron diffraction on FeCr₂S₄ above 10 K shows that there is no crystallographic distortion and reveals a ferrimagnetic ordering, with the magnetic moment of Fe²⁺ ($-3.52 \ \mu_B$) aligned antiparallel to Cr³⁺ ($2.72 \ \mu_B$). A cusp-like anomaly is observed in both the field-cooled (FC) and the zero-field-cooled (ZFC) magnetization curves of the sample x = 0.1, near 130 K, under an applied field H = 100 Oe. The MR of the sample x = 0.1 shows a semiconducting behavior in the low-temperature region, and the metal-metal transition occurs near the Néel temperature. The charge state of iron ion for the sample x = 0.1 is ferrous (Fe²⁺), whereas it is ferric (Fe³⁺) for the sample x = 0.3. The Mössbauer spectra of the sample x = 0.1 show asymmetric line broadening, and this is considered to be due to dynamic Jahn-Teller relaxation. The unusual reduction of magnetic hyperfine field below 110 K is interpreted by the cancellation effect between the mutually opposite orbital current field (H_L) and Fermi contact field (H_C).

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