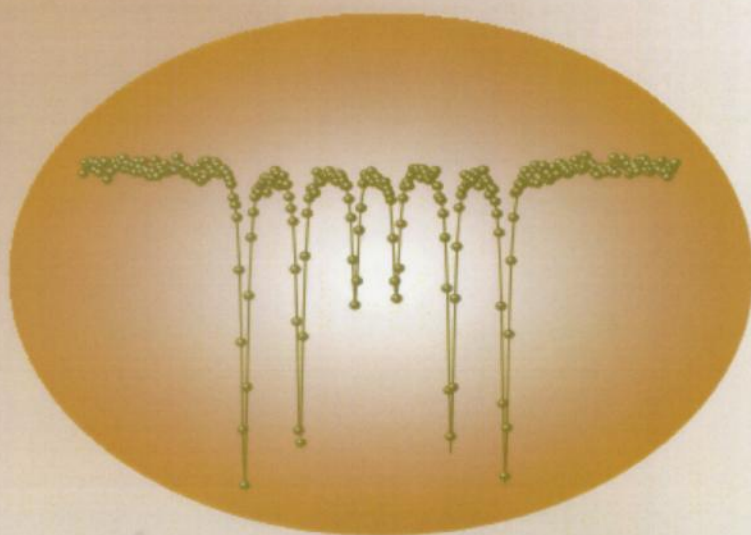


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Mössbauer studies of the cooperative Jahn-Teller effect in spinel $\text{Fe}_{1-x}\text{Cd}_x\text{Cr}_2\text{O}_4$ ($x=0.0, 0.1$)

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The orbital degrees of freedom of $3d$ electrons are very important parameters that significantly affect physical properties of strongly correlated electron system. Various types of electron orbital ordering are caused by strong coupling with ordering of other degrees of freedom, lattice, spin, and charge.[1,2] The investigation of magnetic hyperfine interaction with magnetic and structural phase transitions in spinel $\text{Fe}_{1-x}\text{Cd}_x\text{Cr}_2\text{O}_4$ ($x=0.0, 0.1$) is very few. Polycrystalline $\text{Fe}_{1-x}\text{Cd}_x\text{Cr}_2\text{O}_4$ ($x=0.0, 0.1$) powder was prepared by using a solid state reaction method. The crystallographic and magnetic properties of powder were characterized by X-ray diffraction (XRD), Mössbauer spectroscopy, and vibrating sample magnetometer (VSM). The crystal structure at room temperature was found to be single phase of cubic normal spinel structure with lattice constant $a_0 = 8.3827 \text{ \AA}$ for FeCr_2O_4 and $a_0 = 8.4024 \text{ \AA}$ for $\text{Fe}_{0.9}\text{Cd}_{0.1}\text{Cr}_2\text{O}_4$ by the Rietveld refinement method. Mössbauer spectra of $\text{Fe}_{1-x}\text{Cd}_x\text{Cr}_2\text{O}_4$ ($x=0.0, 0.1$) have been taken at various temperatures ranging from 4.2 to room temperature. A systematic change in the Mössbauer spectrum with decreasing temperature was found and attributed to the Jahn-Teller distortion. Below a T_N of 72 K for FeCr_2O_4 and 65 K for $\text{Fe}_{0.9}\text{Cd}_{0.1}\text{Cr}_2\text{O}_4$, the spectrum displayed an asymmetric eight-line shape indicating a large electric quadrupole contribution with spin ordering. The magnetic hyperfine field and electric quadrupole interaction at 4.2 K have been fitted with Mössbauer hyperfine parameters of $H_{\text{hf}} = 192 \text{ kOe}$, $\theta = 87^\circ$, $\phi = 54^\circ$, $\eta = 0.2$, $\Delta E_Q = 3.24 \text{ mm/s}$, $R = -2.5$ for the FeCr_2O_4 sample and $H_{\text{hf}} = 186 \text{ kOe}$, $\theta = 75^\circ$, $\phi = 87^\circ$, $\eta = 0.02$, $\Delta E_Q = 3.41 \text{ mm/s}$, $R = -2.7$ for the $\text{Fe}_{0.9}\text{Cd}_{0.1}\text{Cr}_2\text{O}_4$ sample, respectively. A sudden change in both the magnitude of magnetic hyperfine field and its slope below 40 K for FeCr_2O_4 and 30 K for $\text{Fe}_{0.9}\text{Cd}_{0.1}\text{Cr}_2\text{O}_4$ suggests that magnetic phase transition related to the spiral spin ordering takes place abruptly. Each line of the Mössbauer spectra becomes broadest at the cubic-to-tetragonal transition temperature of 135 K for the FeCr_2O_4 sample and 105 K for the $\text{Fe}_{0.9}\text{Cd}_{0.1}\text{Cr}_2\text{O}_4$ sample, which is considered to be due to the Jahn-Teller effect of Fe^{2+} ions. Isomer shift at room temperature is 0.76 mm/s for FeCr_2O_4 and 0.81 mm/s for $\text{Fe}_{0.9}\text{Cd}_{0.1}\text{Cr}_2\text{O}_4$, which means that the charge state of the Fe ions was ferrous in character. Macroscopic magnetic properties of samples will be discussed in the results.

[1] K. Tsuda et. al. *Phys. Rev. B* **81**, 180102(R) (2010).

[2] M. Matsuda et. al. *Phys. Rev. Lett.* **104**, 047201 (2010).