Electronic and magnetic structure of Fe ions in $NiCr_2S_4$

Jae Yun Park
Department of Materials Science and Engineering, Incheon University, Incheon 402-749, Korea

Heung Moon Ko, Woon Hwa Lee, Sang Hee Ji, and Chul Sung Kim
Department of Physics, Kookmin University, Seoul 136-702, Korea

The magnetic semiconductor $Ni_xFe_{1-x}Cr_2S_4$ ($x=0.985$, 0.97, 0.96) has been investigated over the temperature range from 12 to 600 K using a Mössbauer technique. The electronic structure of Fe ions in $NiCr_2S_4$ was calculated with the Hamiltonian incorporating free-ion term, axial and rhombic crystal field, spin-orbital couplings, and exchange interactions. The ground orbital state is separated by 9.64 $|\lambda|$ from the first excited state, thereby making the quadrupole splitting somewhat insensitive to temperature. Using x-ray crystallographic data, the contribution of direct lattice sum to the electric-field gradient has been considered. In calculating the temperature dependence of quadrupole splitting, the axial field parameter $\Delta_1=-3.0|\lambda|$, the rhombic field parameter $\Delta_2=-2.8|\lambda|$, and the covalency factor $\alpha^2=0.73$ in $Ni_{0.985}Fe_{0.015}Cr_2S_4$ were determined. Magnetic hyperfine and quadrupole interactions in the antiferromagnetic state of $Ni_{0.96}Fe_{0.04}Cr_2S_4$ at 12 K have been studied, yielding the following results: $H=147.8$ kOe, $\frac{1}{2}e^2qQ(1+\frac{1}{3}\eta^2)^{1/2}=-1.96$ mm/s, $\theta=66°$, $\phi=90°$, and $\eta=1.0$. The line broadening which suggests the electron relaxation was observed with decreasing temperature.