Crystallographic and magnetic properties of the spinel phase for $Ni_xFe_{1-x}Cr_2S_4$

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Ferrimagnetic $Ni_xFe_{1-x}Cr_2S_4$ is found to crystallize with a pure spinel structure in the composition range $0 \leqslant x \leqslant 0.4$. ^{57}Fe Mössbauer spectra of $Ni_xFe_{1-x}Cr_2S_4$ have been taken at various temperatures ranging from 13 K to room temperature. Analysis of Mössbauer spectra and x-ray crystallographic data indicate that Ni ions occupy the tetrahedral site, and the lattice constant is in good agreement with Vegard's law. The isomer shifts indicate that the valence states of the Fe ions have a ferrous character. The Néel temperature increases linearly with Ni concentration, suggesting that the superexchange interaction for Ni-S-Cr link is stronger than that for Fe-S-Cr link. The anomalous reduction of the magnetic hyperfine field below 70 K could be explained in terms of the cancellation effect between mutually opposite orbital current field H_L and the Fermi contact field H_D. It is notable that, as the temperature decreases below the Néel temperature, both quadrupole shift and asymmetrical line broadening appear and increase with decreasing temperature, suggesting the presence of an electric-field gradient and accompanying relaxation effects.