Effect of Ni Substitution on the Antiferromagnetic Ordering in Olivine $LiFe_{1-x}Ni_xPO_4$

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Ni-doped LiFe_{1-x}Ni_xPO₄ polycrystalline samples were prepared by using the solid-state reaction method in order to investigate the effect of Ni substitution in antiferromagnetic LiFe_{1-x}Ni_xPO₄ system by using a superconducting quantum interference device (SQUID) magnetometer and Mössbauer spectroscopy. The crystal structure was determined to be orthorhombic with the space group Pnma for all LiFe_{1-x}Ni_xPO₄ samples. The magnetic order for all LiFe_{1-x}Ni_xPO₄ samples was antiferromagnetic with different superexchange interactions. From the analyzed Mössbauer spectra, the value of the hyperfine field for LiFe_{1-x}Ni_xPO₄ decreased with increasing Ni substitution at temperatures ranging from 4.2 K to below the Néel temperature. This result suggests that the Fe-O-Fe superexchange interaction is larger than that of the Fe-O-Ni link. Moreover, we found that Ni ions in LiFe_{1-x}Ni_xPO₄ could induce an asymmetric charge density in the FeO₆ octahedral site due to the charge on distant Ni ions.

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