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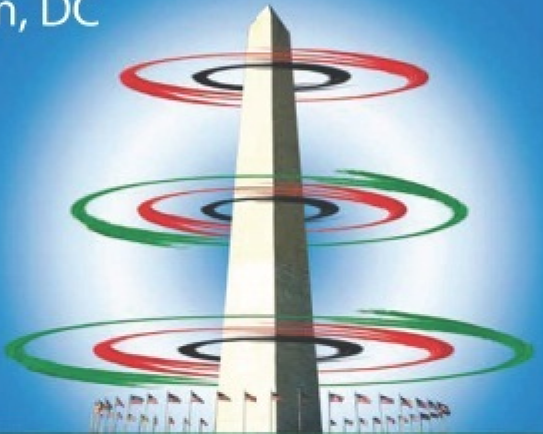
DIGESTS



FQ-08

Mössbauer study of strong magnetoelectric effect $LiNi_{0.99}{}^{57}Fe_{0.01}PO_4$ compound.

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Introduction

In recent years strongly correlated materials that simultaneously show electric and magnetic orderings have attracted much attention due to the applications for promising multifunctional device and because of interesting physics as well [1,2]. In the olivine phosphate materials, $LiMPO_4$ (M = Ni, Co, Fe, Mn), a strong magnetoelectric (ME) effect is observed in the antiferromagnetic phases [3]. The $LiNiPO_4$ compound exhibit the exceptionally large magnetoelectric (ME) coefficients and the hysteresis of the "butterfly" type at low temperature. A general mechanism leading to the ME effect in $LiNiPO_4$ has not yet been established. In this paper, we discuss the magnetic properties of $LiNi_{0.99}{}^{57}Fe_{0.01}PO_4$, focusing on the Mössbauer studies. Here, We have substituted a small amount of ^{57}Fe ions for Ni sites and investigated the hyperfine electromagnetic interaction of Fe ions in crystal symmetry.

Experiment

The pure $LiNi_{0.99}{}^{57}Fe_{0.01}PO_4$ sample was prepared using the following ceramic method. A mixture of the proper proportion of Li_2CO_3, $NH_4H_2PO_4$, NiO, and ^{57}Fe, respectively, was ground, then pressed into a pellet at 5000 N/cm^2 and sealed in an evacuated quartz tube. The temperature was 400 ^{0}C initially and was slowly rasied to 700 ^{0}C over a period of 1 day. Structural characterization of the samples was examined by X-ray diffraction (XRD) measurements using Cu Kα radiation (λ=1.5405 Å) and was analyzed by using a Rietveld refinement. Magnetization measurements were performed in a Quantum Design superconducting quantum interference device (SQUID) magnetometer. Mössbauer spectra were collected using a ^{57}Co (Rh) source in a constant acceleration mode.

Results and discussion

We analyzed the x-ray diffraction patterns of $LiNi_{0.99}{}^{57}Fe_{0.01}PO_4$ using Rietveld's refinement of the Fullprof code. The crystal structure of the sample at room temperature was determined to be orthorhombic with space group *Pnma*. The lattice parameters for the $LiNi_{0.99}{}^{57}Fe_{0.01}PO_4$ were a_0 = 10.041 Å, b_0 = 5.862 Å, c_0 = 4.681 Å, respectively. Temperature dependence of magnetization M showed an anomalous antiferromagnetic system. The magnetic moment decreased with increasing temperature up to 11 K, and then it show a cusp-like maximum value at 22 K, indicating an antiferromagnetic (AF) transition. We determined it as the Néel temperature (T_N). In order to investigate the microscopic interaction around Fe ion occuping Ni ion sites, Mössbauer spectra of $LiNi_{0.99}{}^{57}Fe_{0.01}PO_4$ were measured at various temperatures ranging from 4.2 K to room temperature. We performed an analysis by using the full Hamiltonian for the ^{57}Fe nucleus and by considering both the magnetic dipole and the electric quadrupole interactions. Magnetic hyperfine field and electric quadrupole interactions at 4.2 K have been fitted, yielding the following results: H_{hf} = 293 kOe, ΔE_Q = 2.82 mm/s, θ = 90°, ϕ = 90°, η = 0.75, and R = 1.3, where, θ and ϕ are the polar and the azimuthal angles of the direction of the magnetic hyperfine field at the ^{57}Fe nuclei with respect to the principle axes of the electric field gradient (EFG) tensor, η is the asymmetric parameter and R is the ratio of electric quadrupole interaction to magnetic dipole interaction. The charge state of the Fe ions is ferrous (Fe^{2+}) in character by isomer shift; 1.08 mm/s at RT relative to Fe metal. Not only the magnitudes of the magnetic hyperfine fields but also their slopes change between 9 and 11 K, suggesting that the superexchange interaction also changes simultaneously with the spin-rotation. We find an abrupt change in quadrupole splitting near 11 K and T_N for $LiNi_{0.99}{}^{57}Fe_{0.01}PO_4$. This can be explained from the point of view of magnetic phase transition related to spin ordering. The change of ΔE_Q is caused by the influence of magnetic hyperfine field.

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[2] T. Kimura, Y. Sekio, H. Nakamura, T. Siegrist, and A. P. Ramirez, Nature materials 7, 291 (2008).
[3] I. Kornev and M. Bichurin, J.-P. Rivera, S. Gentil, and H. Schmid, A. G. M. Jansen and P. Wyder, Phys. Rev. B 62, 12247 (2000).

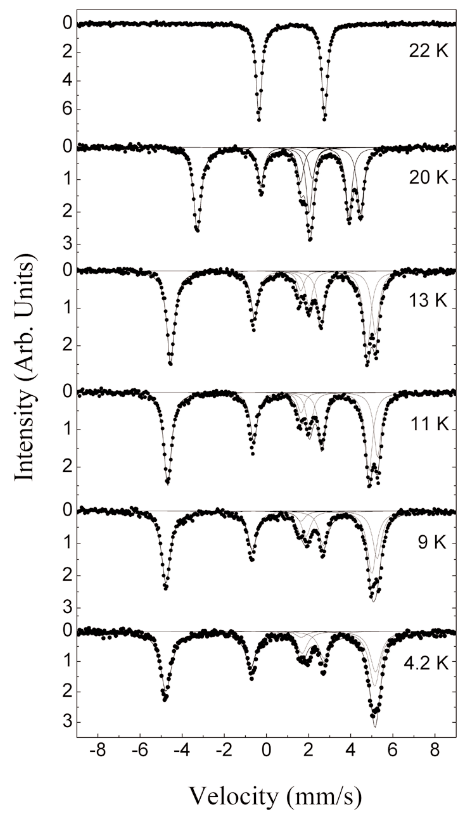

Fig. 1. Mössbauer spectra of $LiNi_{0.99}{}^{57}Fe_{0.01}PO_4$ at various temperature.

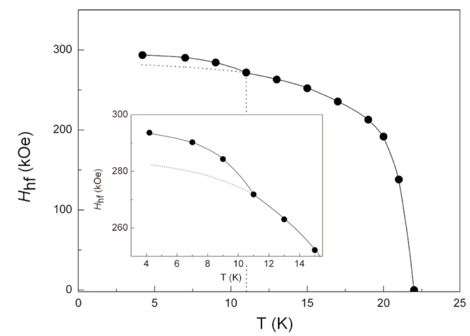

Fig 2. Temperature dependence of the magnetic hyperfine field of $LiNi_{0.99}{}^{57}Fe_{0.01}PO_4$.