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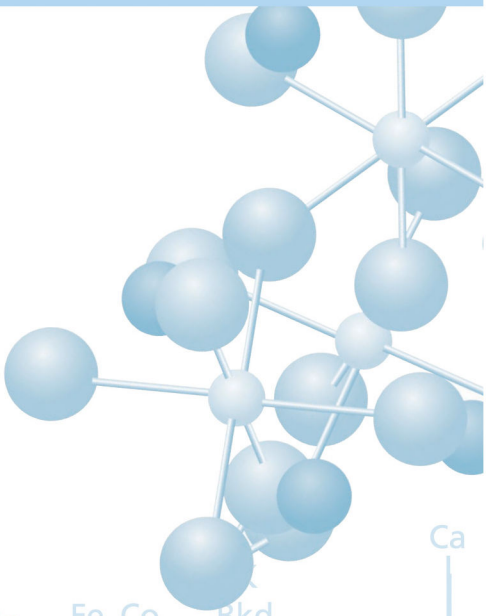

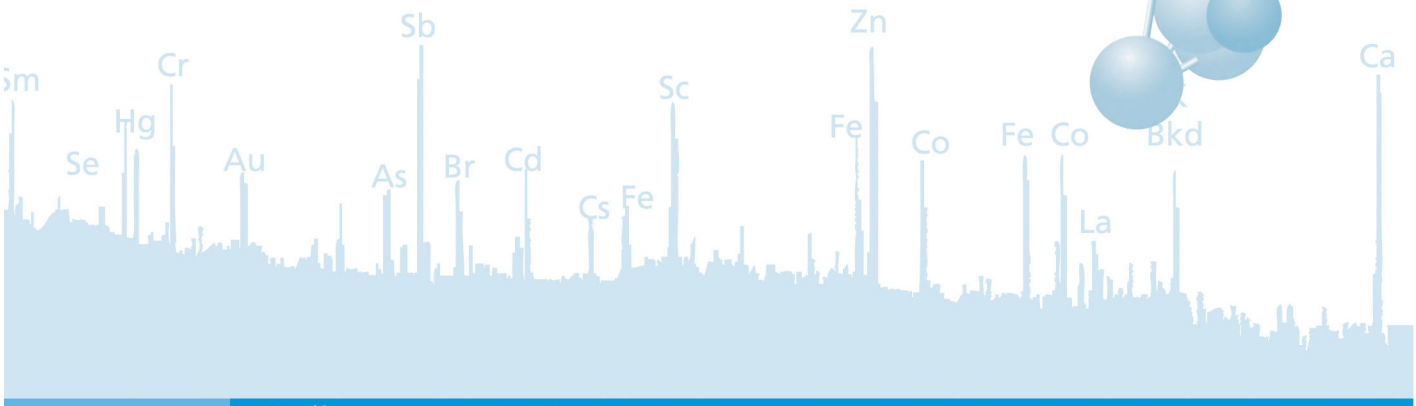

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Neutron diffraction and Mossbauer Studies on Cation Distribution of $GaFeO_3$

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1. Introduction

Gallium iron oxide has been of much interest due to the correlation phenomenon of ferrimagnetism and the piezoelectricity [1]. The crystal structure and local symmetry of oxygen ions around cation have been reported by many authors [2, 3], but the magnetic properties by the various sublattice models have been argued [4]. The knowledge of the magnetic structure and the correct cation distribution of Fe in each magnetic sublattice are inevitable to understand the origin of magnetoelectric effect. In this work, we have investigated the magnetic hyperfine structure of $GaFeO_3$ by Mössbauer spectroscopy and cation distribution of Ga, Fe on each crystallographic site by neutron diffraction.

Also, we studied the change of cation distribution and the magnetic moment of iron occupied on octahedral and tetrahedral sites by different heat treatment method.

2. Experiments

The samples were prepared by standard solid-state reaction. Powders of high purity (99.999 % or better) of Ga_2O_3 and Fe_2O_3 were mixed, ground, pressed into cylindrical pellets, and sintered at 1200 ~ 1400 °C for 12 ~ 30 h in air. The slow-cooled specimen was finally fired at 1400 °C, and then cooled to room temperature at a rate of 1 °C/min. The quenched sample was obtained by rapidly quenching at 1400 °C into ice water.

Neutron diffraction patterns were obtained at 4, 300 K using Korea atomic energy research institute reactor HNARO HRPD. A Mössbauer spectrometer of the electromechanical type was used in the constant-acceleration mode. A ^{57}Co source in a rhodium matrix was used at room temperature.

3. Results and Discussion

FIG. 1 shows neutron diffraction patterns for the quenched sample. The diffraction patterns for the slowly cooled sample were similar to patterns of the quenched sample. The analysis of the neutron diffraction patterns by Rietveld refinement method have shown that the crystal structures of two samples have an orthorhombic structure corresponding to space group $Pc2_1n$ (No. 33). Four different cation sites are labeled Fe1, Fe2 (predominantly occupied by iron), Ga1 and Ga2 (predominantly occupied by Gallium) [2]. Local symmetry of anions around cations is described by an almost regular tetrahedron for G1 site and the distorted octahedrons for the other sites.

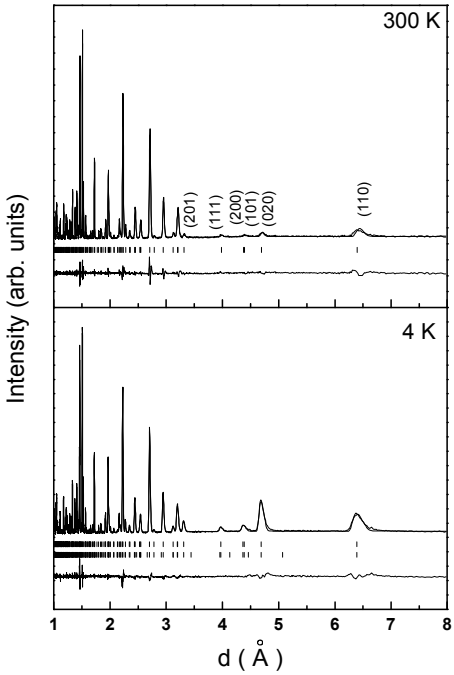

Fig. 1 Neutron diffraction patterns at 300, 4 K for the quenched $GaFeO_3$ and plane indices related to magnetic reflections at 4 K.

In the case of the slowly cooled sample, lattice parameters are a= 8.7457 b= 9.3921 c= 5.0816 Å and for the quenched sample, they are a= 8.7462 b= 9.3896 c= 5.0809 Å at 300 K. The unit cell of samples is not a great difference between two different syntheses

Table 1 Fe ion occupancies and magnetic moment of iron on each site by neutron diffraction analysis at 300, 4 K, respectively.

Site	Slowly Cooled		Quenched	
	Occ.(Fe)	M(μ_B)	Occ(Fe)	M(μ_B)
Ga1	0.18	-	0.19	-
Ga2	0.35	2.378	0.36	2.327
Fe1	0.77	-4.048	0.76	-3.748
Fe2	0.70	3.566	0.71	3.518

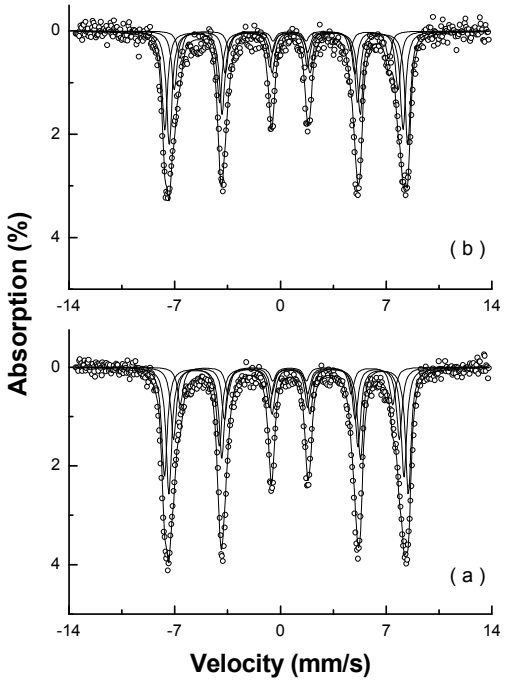

Fig. 2 Mössbauer spectrum at 12 K for (a) the slowly cooled sample and (b) the quenched sample.

methods but distribution of Fe ions on sites of the cation and local symmetry of oxygen ions around each cations show the slight difference. The analyzed occupancies of iron on each site are listed in Table 1.

In neutron diffraction patterns at 4 K below the magnetic transition temperature, we observed the magnetic reflections at (020), (110)… planes. The largely enhanced (020) reflection suggests the arrangement of spin moments in ac plane. The refined magnetic moments of iron on each sublattice are recorded in Table 1. It was noteworthy that the decrease of magnetic moment on Fe1 site was 0.3 μ_B. In addition the bonding angles related to Fe1-O-Fe2 with strong antiferromagnetic superexchange interaction strength decreased from 168.2° to 167.8° for the quenched sample.

To determine the correct distribution of iron ions and magnetic transition temperature for two samples, we have examined the cation distribution of each site using Mössbauer spectroscopy at temperatures from 12 K to room temperature. FIG. 2 shows the Mössbauer spectra taken at 12 K. From the analyzed results of Mössbauer spectra at 12 K, Area absorption ratios of Fe ions on Ga1, Ga2, Fe1, Fe2 are 9.2, 19.5, 38.3, 33.0 % in the case of slowly cooled sample, respectively and 10.9, 20.8, 36.5, 31.8 % in quenched sample, respectively.

A decrease of occupancy and magnetic moment of iron on Fe1 sites and bonding angle in the case of quenched method reduce the exchange interaction strength. Such reduction results in decrease of Néel temperature (T_N).

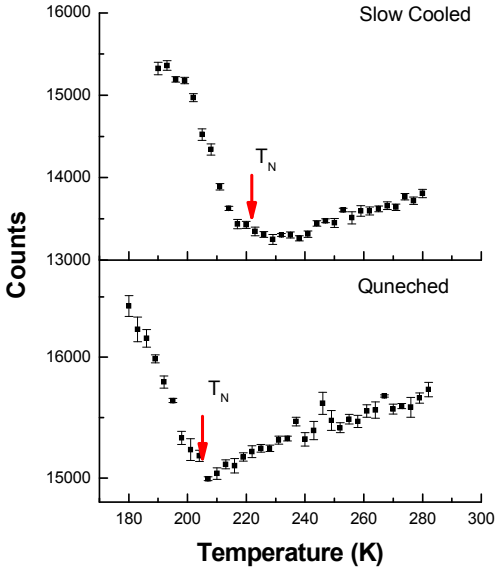

Fig. 3 Temperature dependence of the counts of 14.4 keV γ-ray at zero transducer velocity for $GaFeO_3$

The Néel temperature was measured using the thermal scan method. The relative counts during 10 s as a function of temperature at zero transducer velocity was measured and they are minimized at the magnetic transition temperature. The determined Néel temperature was around 220 K for the slowly cooled sample, decrease to 210 K for the quenched sample.

The isomer shifts at room temperature are found to be 0.38 ~ 0.39 mm/s relative to iron metal, which indicates that iron ions are ferric [5].

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