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Temperature Dependent Magnetic Properties of Bismuth Substituted Terbium Iron Garnets.

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Introduction

Recently $(TbBi)_3Fe_5O_{12}$ have been studied for an optical isolator device in the wavelength range of 1.3-1.6 μm in communication systems[1]. Bismuth substitution in rare-earth garnet enhances the magneto-optical effect, which originates from covalent interactions between Bi^{3+} and Fe^{3+} ions[2]. In terbium iron garnet, the Tb^{3+} ions are in the 24*c* (dodecahedral) while the Fe^{3+} ions occupy the 16*a* (octahedral) and the 24*d* (tetrahedral) sites. The terbium iron garnet has ferrimagnetic property at room temperature, which has been described by the strong *a-d* superexchange interaction between iron sublattices. But the spin structure of terbium iron garnet at compensation temperature (260 K) has been described by the spin reorientation[3] and the double-umbrella magnetic structure below 77 K[4].

In this work, we report on the structural and magnetic properties of $Tb_{3-x}Bi_xFe_5O_{12}$ (x=0.5 and 1.0) powders. We observed the negative magnetization value at low temperature for $Tb_{3-x}Bi_xFe_5O_{12}$ powders, which is explained by the analysis of the local structure of iron sublattices using Mössbauer spectroscopy.

Experiments

$Tb_{3-x}Bi_xFe_5O_{12}$(x=0.5, 1.0) compounds were prepared by a sol-gel method. $Tb(NO_3)_35H_2O$, $Bi(NO_3)_35H_2O$ and $Fe(NO_3)_39H_2O$ were dissolved in 2-methoxyethanol(2-MOE) and acetic acid. The solution was refluxed at 80 °C for 24 h and dried at 120 °C for 48 h in oven. The obtained powder was annealed at 900 °C for 3 h in air. The crystal structure of the samples were examined using an x-ray diffractometer with Cu-Kα radiation and analyzed by Rietveld refinement. Magnetic properties were measured by vibrating samples magnetometer (VSM) with a field up to 5 kG from 60 to 660 K and the Mössbauer spectra were recorded using a constant acceleration Mössbauer spectrometer with a ^{57}Co source in Rh matrix.

Results and discussion

The x-ray diffraction patterns of the compounds show a single phase crystal structure of space group (*Ia3d*). The lattice constants increase linearly with increasing bismuth concentration, since ionic radius of Bi^{3+} (1.17 Å) is larger than that of Tb^{3+} (1.06 Å). These results accord with Vegard's law. Figure 1 shows the temperature dependence of magnetization for the sample (x=1.0) with increasing the external fields. The derivative of magnetization changes from the negative value under low external field to the positive value below the point which is related to the compensation phenomena with increasing the field. Inset of Fig.1 exhibits the magnetization curves of x=0.5 and 1.0 under low external field (100 G). With increase of bismuth substitution, the Néel temperature increases but the crossover temperature from the positive value to the negative value decreases. The above magnetic properties originate from the local magnetic anisotropy which is related to the strength of spin-orbit coupling and covalent interactions between Bi and Fe, etc. In order to study the change of the detailed local structure, we have obtained Mössbauer spectra at various temperatures below Néel temperature. The values of quadrupole splittings analyzed from Mössbauer spectra have been shown in Fig.2. The octahedral site below the crossover temperature divides into two sites, (16a) and (16a'), respectively. The variation of electric quadrupole at (16a') is related to spin-orbit coupling. The isomer shifts at room temperature of (16a) and (24d) site are 0.26 mm/s and 0.04 mm/s, for x=1.0, respectively, and they are 0.27 mm/s and 0.04 mm/s, for x=0.5, respectively. This means that the iron at (24d) site have a strong covalent interaction with bismuth.

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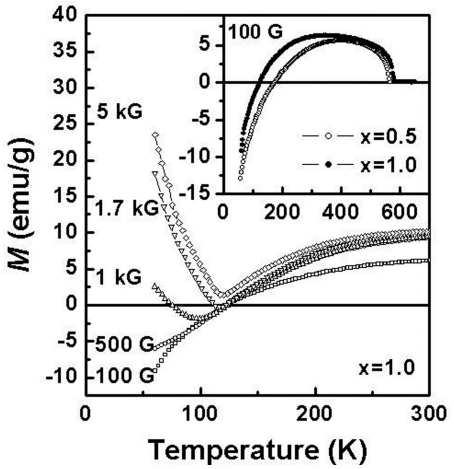

Fig.1. Temperature dependence of field cooled magnetization for $Tb_2Bi_1Fe_5O_{12}$ at various external magnetic fields. Inset: Temperature dependence of magnetization for x=0.5 and x=1.0 (H=100G).

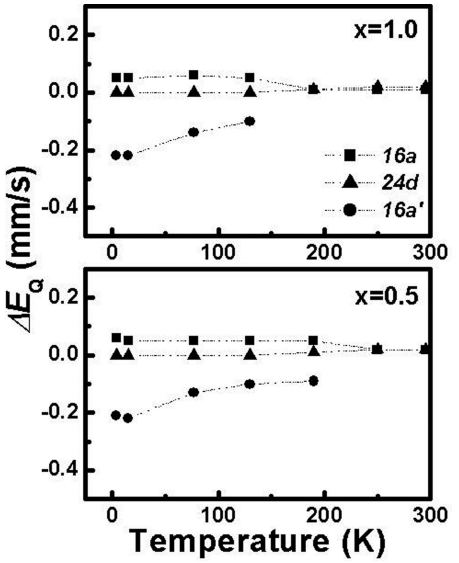

Fig.2. Temperature dependence of electric quadrupole splittings for x=0.5 and x=1.0.