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Effects of gallium distribution on terbium bismuth gallium iron garnet.

I. Park, C. Kim

Physics, Kookmin University, Seoul, South Korea

Introduction

Heavy rare earth (RE) iron garnet has canted magnetic structure which is described as a "double umbrella structure" at low temperature. The heavy RE ion spins form a double cone around the [111] axis, and these spin affect to the iron set of 16a site [1]. These materials related to the negative magnetization have been reported that the net magnetization has negative value at low temperature under field cooled condition for $Tb_2Bi_1Fe_5O_{12}$ [2], and $Ho(Fe_{0.6}Mn_{0.4})_{12}$ systems [3]. Terbium bismuth gallium iron garnet (TbBiGaIG) is the candidate material for Faraday rotator for wide band and temperature-stabilized optical isolators [4]. It is well known that heavy RE iron garnet shows compensation phenomenon. Compensation phenomenon attracted attention for new concepts of integrated optical isolators based on nonreciprocal optical mode interference [5]. In RE iron garnet system, both octahedral (16a) and tetrahedral (24d) sites are occupied by Fe^{3+} ions. When the gallium ion is substituted in garnet system, the cation distribution of gallium and iron ions between 16a and 24d sites depends on the temperature. The heat treatment is able to change the sign of the faraday effect by moving some of the Ga ions from 24d to 16a site [5-6].

In this work, we report on the structural and magnetic properties of $Tb_2Bi_1Ga_1Fe_4O_{12}$ powders, which prepared by sol-gel and vacuum annealing process. The distribution of gallium and iron in $Tb_2Bi_1Ga_1Fe_4O_{12}$ is explained by the analysis of the local structure of iron sublattices using Mössbauer spectroscopy.

Experiments

$Tb_2Bi_1Ga_1Fe_4O_{12}$ compounds were prepared by a sol-gel method. Terbium, bismuth, gallium, and iron nitrate 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 sealed in evacuated quartz tubes. The annealing temperature was room temperature initially and was slowly raised to 1000 °C over a period of seven days.

The crystal structure of the sample was examined using an x-ray diffractometer with Cu-$K\alpha$ radiation and analyzed by Rietveld refinement. 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 pattern of the compounds shows a single phase crystal structure of space group ($Ia3d$) {Tb(24c); Bi(24c); Fe(16a); Ga(16a); Fe(24d); O(96h) (u, v, w)} with its lattice constant $a_0 = 12.465$ Å. The refined X-ray diffraction pattern of sample is shown in Fig. 1. In order to study the change of the detailed local structure, we have obtained Mössbauer spectrum. Mössbauer spectrum of $Tb_2Bi_1Ga_1Fe_4O_{12}$ at room temperature is shown in Fig. 2. The Mössbauer spectrum for the sample was composed of two six-line hyperfine patterns 24d (inner sextet) and 16a (outer sextet). The spectrum of $Tb_2Bi_1Ga_1Fe_4O_{12}$ at room temperature consists of 2 sets of 6 Lorentzians, which is the pattern of single-phase garnet. From the analyzed results of Mössbauer spectrum at room temperature, the absorption area ratios of Fe ions on 24d and 16a sites are 74.7 % and 25.3 % (approximately 3:1), respectively. In case of our previous results for $Tb_2Bi_1Fe_5O_{12}$, the absorption area ratios of Fe ions on 24d and 16a sites are 60.8 % and 39.2 % (approximately 3:2), respectively. This proportion is the conventional absorption area ratios of Fe ions of garnet. It can be analogized the Ga ion distribution by this result. It is noticeable that all of the nonmagnetic Ga atoms occupy the 16a site by vacuum annealing process. This is also in accord with the x-ray dif-

fraction refinement results. We suggest that the control of site preference of Ga cation from 24d site to 16a site was accomplished by high temperature vacuum annealing process.

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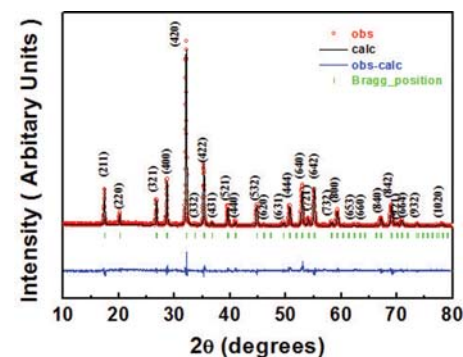

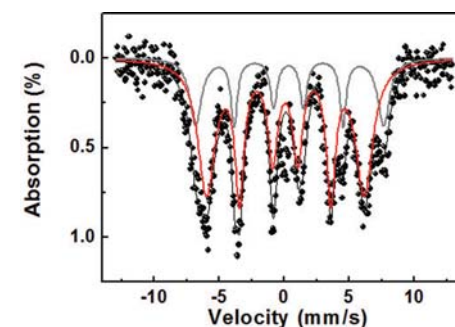Fig. 1. Rietveld refinement of X-ray diffraction patterns for $Tb_2Bi_1Ga_1Fe_4O_{12}$.

Fig. 2. Mossbauer spectrum of $Tb_2Bi_1Ga_1Fe_4O_{12}$ at room temperature.