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Magneto-optical properties of Bi-YIG particle-composite films prepared by different coprecipitation procedures

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Abstract- Magneto-optical Bi-YIG particles are prepared by different coprecipitation procedures. The crystallite size of the particle increases with the heat treatment temperature. In spite of a similar crystallite size, the saturation magnetization is considerably different by the coprecipitation procedure. The Faraday rotation of the composite film with the mixture of epoxy and the Bi-YIG particles linearly increases with the saturation magnetization for the film with crystallites smaller than 30 nm, while that for the film with larger crystallites deviates from the linearity and exhibits a decrease in the Faraday rotation. These results can be interpreted based on the scattering of the probing light due to the morphology of the composite film. The presence of a magneto-optically inactive surface region of the crystallite is also discussed.

Key words: Faraday rotation, magneto-optical properties, garnet, Bi-YIG, particle-composite films

1. Introduction

Embedding of magneto-optical (MO) particles in a transparent epoxy film is an alternative approach to produce novel MO materials for electrical and optical devices. There are some ingredients that should be improved for the application of the particle-composite films. One of the ingredients is a decrease in the Faraday rotation due to the small volume of MO materials along the light path, compared with single crystals. And also, an irregular atomic arrangement at the surface of the particle causes the scattering of the light and brings about the degradation of the MO performance.

In this work, we report on the MO properties of Bi-YIG particle-composite films prepared by different coprecipitation procedures in order to improve the performance. The enhancement in the Faraday rotation is also demonstrated for the films with the particles below 30 nm in size.

2. Experimental

Bi-YIG particles were prepared by coprecipitation method using nitric acid solution of bismuth(III) nitrate pentahydrate, yttrium(III) nitrate hexahydrate, and iron(III) nitrate enneahydrate. One of the essential factors for the preparation of sized particle is to maintain the acidity over the coprecipitation process. For this purpose, we employ the following different coprecipitation procedures. (1) The solution of the cations was dropped into a large amount of NH₄OH solution so as to suppress a drift of acidity during the coprecipitation. In another way, the acidity of the NH₄OH solution was controlled to be pH=10.5 by automatic titration of (2a) NH₄OH or (2b) NaOH solution, and pH=12.5 by titration of (2c) NaOH solution, when the cation solution was dropped into NH_4OH . The prepared coprecipitates were heated at temperatures from $T_h=525^{\circ}C$ to $675^{\circ}C$ for crystallization. The obtained Bi-YIG particles were embedded in epoxy film on a glass substrate by the coating process described elsewhere [1].

The crystal structure of the particle was confirmed by xray diffraction (XRD), and the average size of the particle was estimated by Scherrer's formula. The saturation magnetization (Ms) was measured by a SQUID magnetometer. Magneto-optical measurements were performed using a probing laser of 520 nm in wavelength.

3. Results and discussion

In the coprecipitation by the procedure (1), the acidity changes from $pH\sim12.5$ to 10 by an addition of 50 ml of cation solution. The change in the acidity should cause some



Fig. 1 The crystallite size and the saturation magnetization of the particles as a function of heat treatment temperature.



Fig. 2 The Faraday rotation of the various films as a function of the saturation magnetization of the particles. Inset shows the θ_r/Ms versus crystallite size.

inhomogeneity in the sample, e.g., crystallite size and/or its agglomerative morphology. Such an ambiguous factor is eliminated in the procedures (2a) - (2c) by the controlled pH.

Figures 1 show the crystallite size (d) and the saturation magnetization (Ms) of the particles as a function of heat treatment temperature (T_h). The crystallite size lies on the shadowed region, suggestive of the growth of the crystallite with increasing T_h except for some scattered points. It is also noteworthy that the coprecipitates prepared by the procedure (2c) crystallize into the garnet structure at lower temperatures. The saturation magnetization of the samples prepared by the procedures (1) and (2a) increases with T_h on a similar curve. The Ms of the particles prepared by the procedure (2b) and (2c), however, deviates from the curve and the deviation is more pronounced in the (2c). The deviation is possibly due to the promotion of the crystallization reaction, which is intermediated by Na ions at low temperatures.

We discuss the relation between the crystallite size and the saturation magnetization. In spite of the similar crystallite size, the saturation magnetization is quite different depending on the coprecipitation procedure. This can be explained by the following model of the morphology and crystallinity of the particles. As stated before, an addition of Na ions promotes the crystallization of the coprecipitates prepared by the procedures (2b) and (2c) at low temperatures. The low temperature heat treatment possibly forms an assembly of Bi-YIG tiny crystallites. In contrast, the particles prepared by the procedure (1) are partially crystallized into garnet phase surrounded by a large amount of other crystallographically irregular phases due to a drift of the acidity and the lack of promotion agent of Na ion. The crystallite size estimated in this study is based only on the several XRD peaks of garnet. Thus, the estimated size is attributed to the crystallized region and could be similar to each other between the two differently prepared samples even the distinctive morphology. Indeed, it is likely that the crystallographically irregular phases merely contribute to the saturation magnetization of the particles prepared by the procedures (1) and (2a). Therefore, the saturation magnetization could be different even in the particles showing similar crystallite sizes in this aspect.

The Faraday rotation $\theta_{\rm F}$ of the film is shown in Fig. 2 as a function of saturation magnetization Ms. The θ_{c} linearly increases with increasing Ms, although it is not the case in the films prepared by the procedure (1). The systematics are clearly visible in the θ_F /Ms versus crystallite size as shown in the inset. The linear dependence of the $\theta_{\rm F}$ corresponds to a constant value of the $\theta_{\rm F}/Ms$. From the inset of Fig. 2, the $\theta_{\rm r}/{\rm Ms}$ suddenly decreases above the crystallite size of 30 nm. The films prepared by the procedures (2a) - (2c) possess only tiny crystallites, and the light passes through the crystallites which contribute to the Faraday rotation. However, the crystallites larger than 30 nm rather scatter the probing light and the light passing through a small amount of tiny crystallites is only responsible for the Faraday rotation for the film prepared by the procedure (1). In other words, the probing light passes through a larger volume of the MO Bi-YIG crystallites in the film prepared by the procedures (2a) - (2c) than in the case of the procedure (1). Thus, this should be a predominant cause for the decrease in the Faraday rotation of the film prepared by the procedure (1) in spite of large Ms values. In this view, the crystallites smaller than 30 nm should be utilized for the MO application of the particle-composite films.

The Faraday rotation proportional to the saturation magnetization is the evidence that the $\theta_{\rm F}$ is attributed only to the saturation magnetization of the particles. This argument is consistent with the previously reported results for typical MO materials²). However, it is interesting to note that the $\theta_{\rm F}$ /Ms somewhat decreases with decreasing the crystallite size below 30 nm. The decrease in the $\theta_{\rm F}$ /Ms indicates the presence of magneto-optically inactive region in the small crystallites. If the surface region of the crystallites has different electronic structure which scarcely contributes to the Faraday rotation, the $\theta_{\rm F}$ /Ms could decrease. In addition, it should be also mentioned that changes in the Bi content with crystallite size may cause some modifications of the MO properties.

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