# Growth Peculiarities and Magnetic Properties of $(LuBi)_3Fe_5O_{12}$ Films by LPE Method

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**Abstract.** *The series of $(LuBi)_3Fe_5O_{12}$ film were grown on (111) oriented GGG substrate with diameters 1, 2 and 3 inch by liquid phase epitaxy using $Bi_2O_3$-base flux. Different types of surface morphology on the grown films were observed. The films' surface was smooth and mirror while the film thickness was less than 13 μm and becomes rough for thickness above this value. The grown films were characterized by measuring magnetization loops and magneto-optic Faraday rotation under magnetization reversal as well as ferromagnetic resonance (FMR). All films with mirror surface demonstrate the in-plane magnetization, high Faraday rotation and FMR linewidth about 0.8 Oe at 9.1 GHz and room temperature.*

**Key words:** iron garnet, liquid phase epitaxy, surface morphology, magnetic properties.

## I. Introduction

Bismuth substituted iron garnets films have been shown to be a prospective material for magneto-optic (MO) device applications such as Faraday isolators, MO modulators, visualizers etc. as well as magneto-static wave devices [1,2]. Under certain conditions microwave frequency magneto-static waves can interact with the optical radiation that propagates in the film [3,4] and allows modulation of optical signal that can be used in communication and signal processing devices [5]. That's why the linewidth of the ferromagnetic resonance (FMR) corresponding to losses at relaxation of magnetic excitations is one of the most important parameters of the films desired for microwave applications. On the other hand, to increase the MO activity it is necessary to incorporate bismuth ions as much as possible into the garnet structure. Because the Bi ions have a large radius, the small ions for the rare earth garnet component are required. From all rare earth ions the lutetium ions have the smallest ion radius.

This work is focused on growth process by liquid phase epitaxy (LPE) method of $(LuBi)_3Fe_5O_{12}$ films, examination surface morphology changes with film thickness increasing and investigation of magnetic properties of grown films.

## II. Growth Process and Experimental Methods

The garnet films with calculated formula $Lu_{2.1}Bi_{0.9}Fe_5O_{12}$ were grown by conventional dipping LPE method on (111)-oriented gadolinium gallium garnet (GGG) substrate with 1, 2 and 3 inch in diameter from supersaturated melt on the base of $Bi_2O_3$ flux. All technological experiments were carried out in air using the five-heating-zone LPE furnace "Garnet-3" (LPAI, France). MgO and CaO has been added to the flux for compensate $Pt^{4+}$ impurities resulting from Pt crucible usage. To calculate a charge composition the molar ratios for the Bi-doped lutetium iron garnet (LuIG) films LPE growth in [6] were used as start values. Then they were modified to obtain an optimal growth conditions for BiLuIG films.

The film-substrate lattice mismatch was determined by X-ray diffractometry (XRD) method using the DRON-3 (Burevestnik, Russia) diffractometer. The surface morphology and magnetic domain structure were observed using the Nomarski interference contrast microscope (Nachet, France) and polarizing microscope ECLIPSE LV100 POL (Nikon, Japan) respectively. Ferromagnetic resonance (FMR) was measurement at perpendicularly applied magnetic field at frequency 9.1 GHz by short-cut stripe resonator method. The magnetization loops were measured at room temperature by vibration sample magnetometer (VSM) method. Faraday rotation (FR) at magnetization perpendicular to the film plane was measured at wavelength 633 nm.

## III. Results and Discussion

### A. Growth Peculiarities and Film's Surface Morphology

The BiLuIG films were grown at temperature 800…810 °C and growth rate was changed from 0.3 to 0.75 μm/min. Saturation temperature for the melt was about 817 °C. After growth process the film surface is covered with flux residuals because of $Bi_2O_3$-based flux has a high viscosity compared with PbO-based flux [7]. Usually, in order to clean a film surface after growth the high speed rotation with 800…1000 rpm is used to spin off flux residuals. But in this case a possibility to destroy grown film increases, especially for films with large diameter. To avoid the complicate high speed rotation procedure the substrate was set before growth into the holder with small inclination about 5° and was kept after growth process above melt for a few minutes till full cleaning of film surface. It was observed that the cleaning time depends from the film diameter and for 1, 2 and 3 inch is equal to 2, 5 and 7 min respectively. It depends also from content of divalent metal oxides in the flux and decreases with increasing MgO or CaO concentration.

The surface morphology was investigated for films grown with different thicknesses and growth rates. It was observed two types of surface morphology. The first type was a smooth and mirror film surface, and the second was classified as a roughness. The surface morphological investigation results are demonstrated by diagram in fig. 1 in the "growth rate – film thickness" coordinates where observed types of surface morphology are marked by different symbols. One can see that the types of surface depend only from film thickness and mirror surface was observed while the film thickness was less than about 13

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μm. This value can be considered as critical thickness. For all other cases the rough surface was observed. Moreover, each type of the surface morphology was independent on the film-substrate mismatch, which changed from $-8\cdot10^{-3}$ to $+4\cdot10^{-3}$ Å.

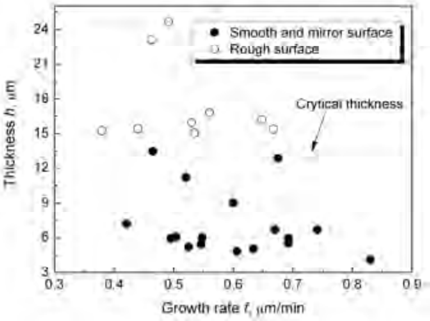


**Fig. 1. The films' surface morphology diagram in the "growth rate – film thickness" coordinates.**

At temperature less than 804 °C the melt surface was covered with spontaneously grown crystals and grown films at this case have turbid surface. This phenomenon considerably limit supercooling region of the melt. Thus, supercooling level of melt acceptable for films growth is about 13 ºC.

### *B. Magnetic Properties of Growth Films*

The FMR linewidth of grown films was calculated on half height level of main resonance line in FMR spectra. Fig. 2 shows the typical FMR spectrum measured at 9.1 GHz of the LuBiIG film. The best results for FMR linewidth was 0.8 Oe. Such a narrow FMR linewidth is desirable for the applications in microwave devices, for example, magneto-static wave filters and delay lines [7,10].

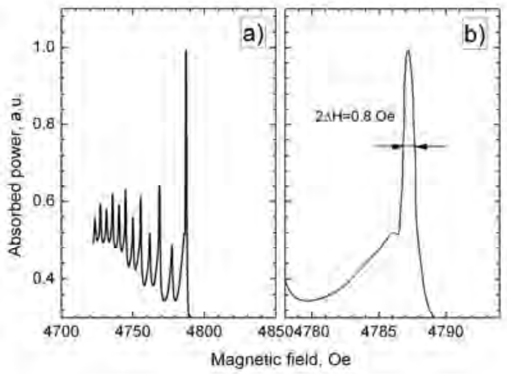


**Fig. 2. The typical FMR spectrum for BiLuIG films at 9.1 GHz (a) and FMR linewidth (b).**

The Faraday rotation angles for grown films were measured under magnetic field up to 3 kOe applied in the perpendicular to the film plane direction. Saturation field was about 1800 Oe and specific Faraday rotation varies in the range 1.2…1.4 deg/μm at wavelength 633 nm. From the FR results and taking into account lattice parameter measurements the Bi content was estimated in the films as 0.86…0.94 atoms per formula. The LuBiIG films demonstrate a stripe-like magnetic domain structure. Domain period increases with film thickness increasing.

The magnetization loops for the LuBiIG films were measured at room temperature along both parallel and perpendicular to the film's plane directions. Typical magnetization loops are shown in fig. 3. The saturation magnetization $4\pi M_s$ of the films is about 1600 G. The magnetization loop in the magnetic field parallel to the plane saturates in much lower magnetic field (~ 200 Oe) in comparison with a case of magnetic field applied perpendicular to the film plane (~ 2500 Oe).

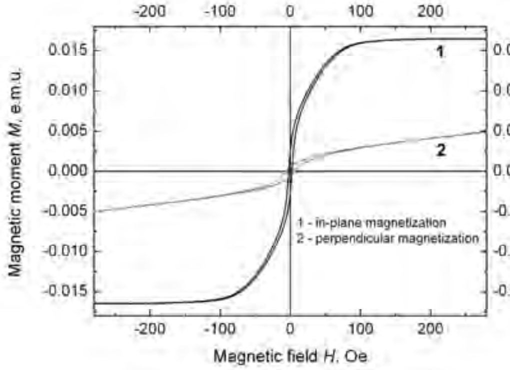


**Fig. 3. The typical magnetization curves of BiLuIG films in the magnetic field parallel and perpendicular to the film plane measured at room temperature.**

The in-plane and perpendicular coercivity of the measured film are about 5 Oe and 12 Oe, respectively. Thus, all BiLuIG films demonstrated in-plane magnetic anisotropy.

## IV. CONCLUSIONS

The conditions of LPE growth from lead-free flux of films of nominal composition $Lu_{2.1}Bi_{0.9}Fe_5O_{12}$ with diameters 1, 2 and 3 inch were determined. The grown films had the smooth and mirror surface while the thickness was less than 13 μm. All such films demonstrated the in-plane magnetization at room temperature and possessed the high Faraday rotation and narrow FMR linewidth as well. The combination of large size, good magneto-optical and microwave properties of these films promise a wide potential application in MO and microwave integrated devices.


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