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# Magnetic properties of hexagonal barium ferrite films on Pt/MgO(111) substrates annealed at different temperatures

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ABSTRACT

In this work, hexagonal barium ferrite thin films have been deposited on Pt/MgO(111) substrates by pulsed laser deposition. The anneal temperature dependence of crystal structures, extents of diffusion and magnetic properties have been studied. X-ray diffraction patterns reveal that the crystal structure changes from the hexagonal to the spinel when the anneal temperature increases. The texture with c-axis perpendicular to the film plane and the small *c*-axis dispersion angles ($\Delta\theta_c$) have been obtained in the film annealed at 950 °C for 10 h. Both the X-ray photoelectron spectroscopy profiles and energy dispersive spectrometer show that the diffusions of $Mg^{2+}$ and $Fe^{3+}$ cations are more obvious when the annealing temperature is higher than 950 °C. The film annealed at 950 °C show anisotropic and hard magnetic properties. The magnetic properties of film annealed at 1050 °C are soft.



## 1. Introduction

For decades, hexagonal barium ferrite ($BaFe_{12}O_{19}$, BaM) films have attracted much attention as potential materials for self-biased microwave devices, such as circulators, phase shifters and notch filters due to their high saturation magnetization, large magnetocrystalline anisotropy and proper ferromagnetic resonance linewidth [1,2]. In order to meet the requirements of self-biased devices, the films should have better magnetic properties and a thickness which should be larger than 50 μm. Some of substrates such as $Al_2O_3$, Si/$SiO_2$ and 6H–SiC have been employed to deposit high quality BaM films [3,4]. However, the thermal expansion coefficients of those substrates are less than that of BaM ($10\times10^{-6}$ °$C^{-1}$),such that the films sustain significant biaxial tensile stress upon cooling from the deposition temperature. The large stress is the cause of fracture and delamination of the film from the substrate [5]. Owing to this shortcoming, the maximum attainable thickness of BaM film on those substrates should be less than 20 μm, which is too thin for practical self-biased devices.

Recently, magnesium oxide (MgO) has been thought as one of the candidates substrates for depositing high quality thick BaM film ( > 50 μm) [6–8], thanking to the small lattice mismatch between MgO (111) plane ($a_{MgO}$=0.5957 nm) and (00l) plane ($a_{BaM}$=0.5893 nm) of $BaFe_{12}O_{19}$. Furthermore, the measured thermal expansion coefficient of MgO ($13.6\times10^{-6}$ °$C^{-1}$) is larger than that of BaM [9], which will result in the BaM film experiencing a biaxial compressive stress upon cooling. Attempts have been made to deposit BaM films by various techniques including pulsed laser deposition (PLD) [10,11], RF magnetron sputtering [12] and screen printing [2]. Compared with other deposition techniques, PLD is more suitable for depositing oxides thin films with complicated stoichiometry, for instance BaM thin films, due to its easy adjusting the film properties in non-thermodynamic equilibrium condition [13]. However, BaM films are always in amorphous states after PLD deposition. Accordingly, post-annealing at high temperature for crystallization is an essential step to obtain high quality epitaxial BaM films. However, Mg atoms in MgO substrate are easy to diffuse into the films under high temperature, which would greatly deteriorate the quality of films [14]. Although several buffer layers, such as gold (Au) or platinum (Pt), have been widely used to reduce the diffusion [12], the diffusion to some extent still exists. Therefore, it remains imperative to study the effect of post-annealing on the magnetic properties of BaM films. In this paper, we will report our work on the PLD preparation of BaM films on MgO substrate with Pt underlayer. The effect of annealing temperature on phase transformations, cations diffusion and magnetic properties of the prepared films will be discussed.

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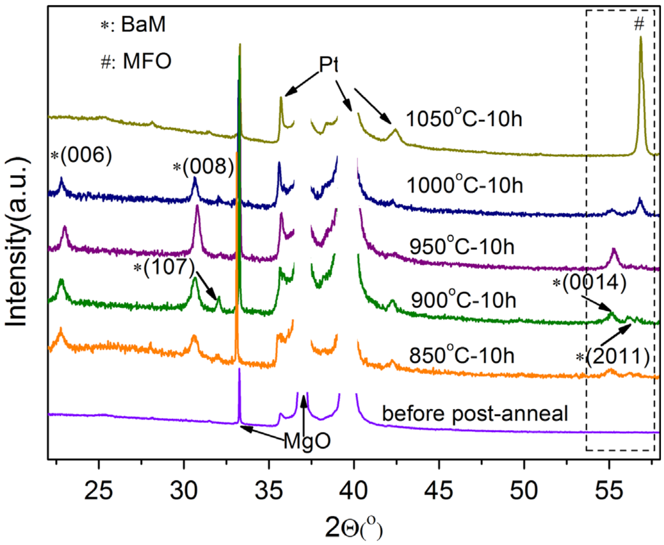


**Fig. 1.** XRD patterns of films deposited on Pt/MgO(111) substrates.

## 2. Experimental details

PLD deposition technique with a KrF excimer laser (248 nm) is used to prepare BaM thin films on MgO (111) substrates. The BaM target used for thin films deposition was synthesized by a traditional sintering route. The distance between the target and the substrate is 45 mm. The MgO substrates are cleaned according to the RCA method. The underlayer (Pt) with a thickness ($t$) of 50 nm was firstly prepared on a substrate with 300 °C and under an argon atmosphere with 1 Pa pressure. Subsequently, the substrate temperature was increased to 800 °C and BaM films with a thickness of 850 nm were deposited under oxygen with 0.3 Pa pressure. When the depositions were completed, the thin films were annealed at different temperatures ($T_a$) for 10 h in air. Besides, in order to study the cations diffusion between the film and the substrate, several BaM films with a thickness of 130 nm had also been deposited under the same condition.

The crystal structures of the films were characterized by X-ray diffraction (XRD, Ultima IV). In addition, rocking curves of the BaM (008) peak were also performed to study the quality of the thin films. A vibrating sample magnetometer (VSM, Lake Shore 7410) was employed to measure the magnetic hysteresis loops of the films. The magnesium content in the films was examined by an energy dispersive spectrometer (EDS). The compositional profiles of the atoms in the film with a thickness of 130 nm were investigated by x-ray photoelectron spectroscopy (XPS, ESCALAB 250Xi).

## 3. Results and discussions

XRD patterns of prepared BaM films annealed at different temperatures for 10 h are shown in Fig. 1. The diffraction peaks of hexagonal phase (BaM) and spinel phase ($MgFe_2O_4$, MFO) peaks are marked as "*" and "#" respectively. The dashed box shows three obvious diffraction peaks at 55.1°, 56.1° and 56.9° which belong to BaM (0014), BaM (2011) and MFO (511) respectively. For the thin film without annealing treatments, no diffraction peaks appear other than the peaks of Pt and MgO substrate, which indicates that thin film are not crystallized even the temperature of substrate is 800 °C. For samples experienced post-annealing, the intensity of diffraction peaks other than the substrate and buffer layer gradually increases with $T_a$ increasing. The hexagonal phase can be identified for the films annealed at low temperature. When $T_a$ increases further, the diffraction peak (511) of spinel phase is gradually obvious, while the intensities of BaM peaks are gradually reduced. At 1000 °C, the hexagonal phase and the spinel phase coexist. When $T_a$ is up to 1050 °C, only MFO (511) diffraction peak can be observed, which indicates that the crystal structure of films transforms from the hexagonal phase into the spinel phase. In addition, the diffraction peaks of BaM (107) and BaM (2011) can be also found except the predominant (00l) peaks in films annealed at 850 °C and 900 °C, which indicates that those films have grains without having their $c$-axis perpendicular to the film plane. However, those unwanted peaks are almost disappeared when the film was annealed at 950 °C, which indicates that the film has a good $c$-axis orientation perpendicular to the film plane. In other word, the film has a feature of texture, and such a texture is often required for films used from self-biased microwave devices.

To quantify the texture degree for the films annealed at different temperatures, a Lotgering factor ($f$), expressed in Eq. (1), is used [15]:

$$f = (P - P_0)/(1 - P_0) \tag{1}$$

where $P$ denotes the ratio of sum intensities of the (*00l*) and (*hkl*) reflections in a textured sample. $P_0$ stands for a non-textured film whose diffraction intensities are obtained from the randomly oriented grains. Note that the values of $f$ can vary between 0 and 1. When $f$ is 0, it denotes the sample has randomly oriented grains.

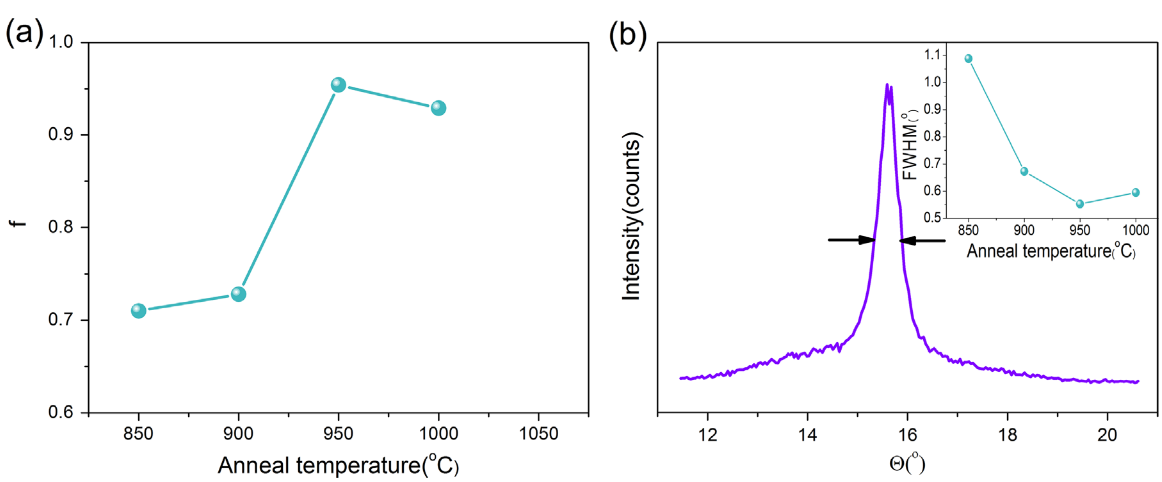


**Fig. 2.** (a) Dependence of Lotgering factor ($f$) upon $T_a$; (b) Rocking curve of (008) peak of BaM film annealed at 950 °C for 10 h. Inset: FWHM values of (008) peak varying with $T_a$.