

The influence of oxygen pressure on the crystallographic structure and magnetic properties of BaM films on Al₂O₃(0001) substrate

Enmin Zhou^{1, a}, Hui Zheng^{1, b*}, Peng Zheng^{2, c}, Liang Zheng^{2, d}, Jiangxia Deng^{2, e}, Huibin Qin^{2, f}

¹Institute of Electron Device & Application, Hangzhou Dianzi University, Hangzhou 310018, People's Republic of China.

^azhenghui0551@126.com, ^bzhenghui0551@yahoo.com, ^czhengpeng@hdu.edu.cn, ^dzhengliang@hdu.edu.cn, ^edengjiangxia@hdu.edu.cn, ^f332744493@qq.com

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Abstract. In this work, hexagonal barium ferrite thin films have been deposited on Al₂O₃(0001) substrates by pulsed laser deposition. The influence of oxygen pressure on the crystallographic structure and magnetic properties of BaM films has been studied. X-ray diffraction theta-2theta reveals the films have a good c-axis orientation perpendicular to the film plane under low oxygen pressure. It is also observed from scanning electron microscope that grain shape in the film surface is changed from platelets into elongated acicular with the increase of oxygen pressure. Magnetic hysteresis loops show the saturation magnetization and remanence of BaM films are greatly depended on the oxygen pressure. They are initially increased to a maximum value, then drastic decreased with increase of oxygen pressure. Consequently, films deposited at 0.08Pa have the highest saturation magnetization of 3900 Gs and remanence ratio of 74.5% which are comparable with the theoretical value of BaM bulk.

Introduction

Recently, hexagonal barium ferrite (BaFe₁₂O₁₉, BaM) have attracted great attention as potential materials for self-biased microwave devices, such as phase shifters and circulators due to their high saturation magnetization, large magnetocrystalline anisotropy and proper ferromagnetic resonance linewidth [1-5]. In addition, with the development in miniaturization of complex geometrical devices, traditional BaM bulk was gradually replaced by the films. Therefore, it is necessary to understand how to deposit crystal BaM films with excellent quality in detail [6-9].

The development of BaM-based microwave devices demands BaM films that have a thickness in the micron or submicron thick [10]. It is known that pulsed laser deposition (PLD) will replace the liquid phase epitaxy and to be the best techniques of choice for the deposition of micron and submicron-thick films [11]. Among deposition conditions, the oxygen pressure seems to greatly influence the properties of the films [12]. S.M. Masoudpanah has used PLD to deposite SrM film on Si substrate and studied the effect of oxygen pressure on the microstructure and magnetic properties of SrM films [13]. However, few studies have been systematically reported on the influence of oxygen pressure on the BaM films [14, 15].

Here, we have prepared BaM films on Al₂O₃ substrate by pulsed laser deposition. The effect of oxygen pressure on crystal structure and magnetic properties of BaM film have been investigated in detail.

Experimental Section

PLD deposition technique with a KrF excimer laser (248 nm) is used to prepare BaM thin films on Al_2O_3 (0001) 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 Al_2O_3



substrates are cleaned according to the RCA method. The substrate temperature was increased to 800 $^{\circ}$ C and BaM films with a thickness of 1 μ m were deposited under different oxygen pressure (0.008~8Pa). When the depositions were completed, the films were annealed at 1000 $^{\circ}$ C for 3 hours in air.

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. The surface morphologies of BaM films were measured by field emission scanning electron microscope (FESEM). A vibrating sample magnetometer (VSM, Lake Shore 7410) was employed to measure the magnetic hysteresis loops of the films.

Results and Explanations

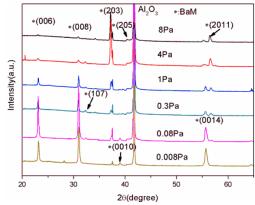


Fig. 1 The crystallographic structure of BaM films at different oxygen pressure.

XRD patterns of prepared BaM films under different oxygen pressure are shown in Fig.1. The diffraction peaks of hexagonal phase (BaM) are marked as "*". It can be seen that the diffractions peaks of BaM was greatly influenced by the oxygen pressure. When the films were deposited in low oxygen pressure, all detected diffraction peaks belong to (00*l*) hexagonal barium ferrite peaks which indicated the films have a good c-axis orientation perpendicular to the film plane. As the oxygen pressure increased, BaM (107) and BaM (2011) peaks were gradually appeared besides the predominant (00*l*) peaks in films. It is indicated that those have grains with c-axis non-perpendicular to the film plane. Once the oxygen pressure up to 4Pa, BaM (00*l*) peaks were almost disappeared.

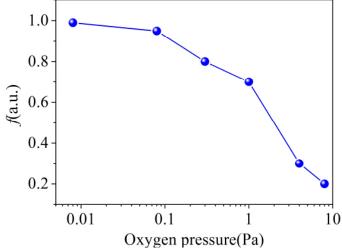


Fig. 2 Dependence of f upon the oxygen pressure.

To quantify the degree of texturing for the films annealed in different temperature, the Lotgering factor (*f*), given by Eq. (1), is calculated [17].



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

where, P denotes the ratio of the sum intensities of the (00l) and (hkl) reflections for oriented sample, while the P_0 for the nonoriented one whose diffraction intensities were obtained from the randomly oriented powder. Note that the values of f can range from 0 to 1, where a value of 0 corresponds to a randomly oriented powder sample and a 1 corresponds to a perfectly textured sample. The dependence of f and oxygen pressure was showed in Fig. 2. Clearly, the perfect texture having strong c-axis orientation perpendicular to the film plane was obtained when the oxygen pressure was 0.008Pa.

To understand the reason of the different c-axis orientations, microstructure of the BaM thin films were studied by FESEM. Fig. 3 displays the SEM morphology of film surface of BaM films with different oxygen pressure. It is clear that the microstructure of the film surface is varying with the oxygen pressure. The grains can be classified into two major categories: those with elongated acicular shape and platelets shape [18]. When the film deposited under low oxygen pressure, platelets shaped grains are formed. However, the elongated acicular grains were formed at high oxygen pressure, and it was increased with increase of the oxygen pressure. Considering the XRD result, the elongated acicular grains has c-axis non-perpendicular to the film plane.

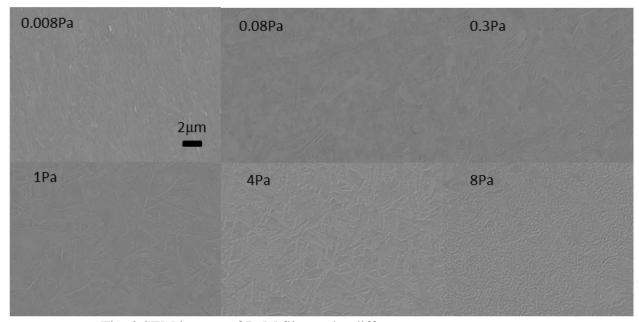


Fig. 3 SEM images of BaM film under different oxygen pressure.

Magnetic hysteresis loops for films deposited at different oxygen pressure were measured in both the out-of-plane and the in-plane geometry by VSM at room temperature with a maximum available applied magnetic field of 15 kOe. Fig. 4 showed the typical magnetic hysteresis loops for the films deposited at 0.008Pa and 1Pa. Clearly, it seen in Fig. 4 that the shape of the loops was changed with the increase of oxygen pressure. From the loops of 0.008Pa, smaller remanence (M_r) was seen in in-plane direction than that of out-of-plane direction, which implied the films exhibit perpendicular magnetic anisotropy. When the film was deposited under high oxygen pressure, M_r in the in-plane direction was almost the same with that of out-of-plane direction which implied the films exhibit low magnetic anisotropy.



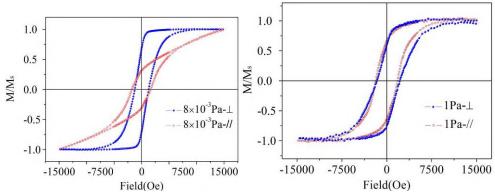


Fig. 4 Magnetic hysteresis loops for BaM films under 0.008Pa (left) and 1Pa (right).

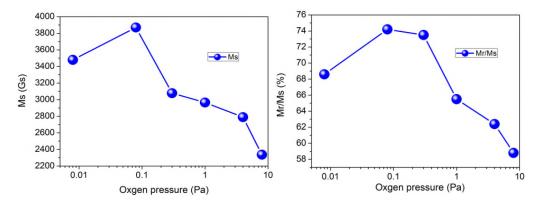


Fig. 5 Oxygen pressure dependence of saturation magnetization (left) and remanence ratio(right).

Fig. 5 showed the M_s and M_r as a function of oxygen pressure. It is found that Ms and Mr/Ms initially increases with the increase of oxygen pressure from 0.008Pa to 0.08Pa, then drastic decreased with the increase of oxygen pressure from 0.08Pa to 8Pa. The initial increasing can be related to the reduced oxygen vacancy. While with the oxygen pressure further increasing, the degree of orientation was gradually reduced which have also been confirmed from XRD and SEM. Thus, 0.08Pa is the most suitable oxygen pressure for BaM film, which has the highest saturation magnetization of 3900 Gs and remanence ratio of 74.5%.

Summary and Conclusions

In this work, BaM films have been deposited on $Al_2O_3(0001)$ substrates by pulsed laser deposition. The oxygen pressure dependence of crystallographic structure and magnetic properties of BaM films has been studied. XRD θ -2 θ reveals the films have a good c-axis orientation perpendicular to the film plane under low oxygen pressure. It is also observed from scanning electron microscope that grain shape in the film surface was changed from platelets into elongated acicular with the increase of oxygen pressure. Magnetic hysteresis loops show the saturation magnetization and remanence of BaM films are greatly depended on the oxygen pressure. They are initially increased to a maximum value with the increase of oxygen pressure for their reduced oxygen vacany. Then drastic decreased with further increase of oxygen pressure for their reduced degree of orientation. Thus, 0.08Pa is the most suitable oxygen pressure for BaM film, which have the highest saturation magnetization of 3900 Gs and remanence ratio of 74.5%.

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