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Magnetic property and stress study of barium hexaferrite thin films with different structures

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M-type barium hexaferrite (BaM) thin films with different structures (d_{s100}: single-layer 100 nm thick film; d_{s200}: single-layer 200 nm thick film; d_{b200}: bilayer 200 nm thick film, where the thickness of each layer is 100 nm and the total film thickness is 200 nm) were deposited on (001) sapphire (Al_2O_3) substrate by radio frequency (RF) magnetron sputtering. Effects of different structures on the crystallographic, morphological, magnetic properties and stress were investigated in details. For the structure of d_{s100}, BaM film processes platelet-like grains, which shows excellent magnetocrystalline anisotropy and large squareness ratio $S_\perp$ (0.62). For the structure of d_{s200}, there are a lot of acicular grains. Thus, the c-axis in-plane oriented and/or randomly oriented grains induce poor magnetocrystalline anisotropy. For the structure of d_{b200}, the platelet-like grains dominate the microstructure. Both squareness ratio $S_\perp$ (0.67) and coercivity $H_{c\perp}$ (98.6 kA/m) of the d_{b200} show the highest values among the three kinds of the film structures. In addition, with the increase of film thickness from 100 nm to 200 nm, the stress in film decreases gradually. Compared with the other structures, the bilayer 200 nm thick film (d_{b200}) obtains the smallest stress.

1. Introduction

M-type Ba-hexaferrite ($BaFe_{12}O_{19}$, BaM) thin films, due to excellent electromagnetic performance, high chemical stability and mechanical durability [1,2], have been widely used in magnetic recordings [3,4], absorbing materials [5,6], and microwave devices [7–9]. Especially, M-type Ba-hexaferrite thin films have built-in high anisotropy fields and can provide a self-biasing for millimeter wave applications in the 30–100 GHz range. Therefore, it can be used to make millimeter wave filters [10,11], circulator [12] and phase shifters [13]. In order to prepare BaM films with excellent performance, many methods have been adopted, such as liquid phase epitaxy, pulsed laser deposition, molecular beam epitaxy and radio frequency (RF) magnetron sputtering. Among these deposition technologies, RF magnetron sputtering has been extensively studied for its large area deposition, high adhesion, efficiency and compatibility with semiconductor fabrication technology. Previous studies have reported that the suitable process parameters, such as RF power level [14], oxygen partial pressure [15], substrate temperature [16] and annealing process [17] can improve the microstructure and magnetic properties of the deposited M-type Ba-hexaferrite thin films. However, because of the difference between the lattice parameters and thermal expansion coefficients of the BaM thin films and substrates, the deposited BaM films usually have large stress, resulting in bad structures and poor magnetic properties. Thus, the preparation of BaM thin films with perfect perpendicular c-axis orientation is still not easy, a small amount of c-axis in-plane oriented and/or randomly oriented grains were grown in the film. For obtaining the BaM ferrite thin films with good electromagnetic performance and low stress, the researchers usually adopt two methods: choosing appropriate substrates and buffer layers. On the one hand, various kinds of substrates like Al_2O_3 [18–21], MgO [22,23], SiC [24], Si_3N_4 [25] are adopted to prepare the BaM thin films. On the other hand, many kinds of buffer layers such as Pt [26–28], ZnO [29,30], AlN [31] have been used to

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improve performance of BaM thin films. But, introducing these buffer layers will cause other undesirable results, such as the extra complexity and high cost. Through introducing a 20 nm thick BaM layer, Xu et al. [22] prepared 500 nm thick BaM film onto Al_2O_3 (001) substrate successfully and achieved great crystallographic and magnetic properties. Peng et al. [32] deposited BaM ferrite films onto sapphire substrates by RF magnetron sputtering. A thin amorphous barium ferrite film of about 20 nm was deposited at low temperature as a buffer layer. BaM ferrite films with different thickness of 100 nm, 1 μm and 8 μm were prepared. The results showed that the 100 nm thick film had good c-axis orientation and strong magnetic anisotropy. However, the c-axis orientation and magnetic anisotropy of the 1 μm and 8 μm films deteriorated sharply. Shide et al. [20] deposited BaM thin films on the Al_2O_3 (001) substrate with different thicknesses, and analyzed the changes in magnetic properties and stress. The increasing film thickness can decrease the stress of BaM films, but the magnetic property of the films is greatly deteriorated. However, there are few reports about the stress of M-type barium hexaferrite thin films with different structures. In order to clarify this problem and obtain the BaM film with low stress and excellent magnetic properties, this work prepares three kinds of film structures, i.e., d_{s100}: single-layer 100 nm thick film; d_{s200}: single-layer 200 nm thick film; d_{b200}: bilayer 200 nm thick film. BaM films with both d_{s100} and d_{b200} structures own good c-axis perpendicular orientation and platelet-like grains, which produce better magnetic property than that of d_{s200}. Especially, due to the stress release resulting from the increasing thickness, BaM film with d_{b200} structure shows the smallest stress.

2. Experimental procedures

2.1. Sample preparation

BaM thin films were deposited on the polished 7 × 7 × 0.5 mm (001) single crystal sapphire (Al_2O_3) substrate by RF magnetron sputtering. A sintered $BaFe_{12}O_{19}$ ferrite disk was selected to deposit films. After evacuating the chamber to a base pressure of 3×10^{-4} Pa, a mixture of 1% oxygen gas (O_2, 99.999% in purity) and 99% argon gas (Ar, 99.999% in purity) was introduced into the chamber as working gas. The general working pressure was maintained at 1.4 Pa. Meanwhile, the RF power was run at 140 W. For the three kinds of films, a 20 nm thick BaM layer was first deposited onto the Al_2O_3 (001) substrate at the substrate temperature of 400 °C, which will act as a buffer layer for the next deposition of the BaM film. Single-layer BaM thin films were deposited directly onto the 20 nm thick BaM buffer layer at the substrate temperature of 400 °C for 80 nm and 180 nm. For the bilayer 200 nm thick films, a 80 nm thick BaM layer was first deposited onto the 20 nm thick BaM buffer layer at the substrate temperature of 400 °C. After that, it was annealed at 850 °C in air for 2 h. Then a second 100 nm thick BaM layer was deposited successively onto the 80 nm layer at the substrate temperature of 400 °C. Prior to this experiment, the deposition rates under the conditions mentioned in the study had been measured. Then by controlling the deposition time, three kinds of BaM thin film structures were deposited on the Al_2O_3 (001) substrate. After deposition, post-deposition thermal annealing was performed at 850 °C in air for 2 h. In addition, the single-layer thin films were labeled as d_{s100} for 100 nm and d_{s200} for 200 nm. The bilayer 200 nm thin film was labeled as d_{b200}.

2.2. Sample characterization and measurement

Crystallographic properties of the BaM films were characterized by a four-circle bede D1 X-ray diffractometer (XRD) at room temperature (Cu target, K_α radiation, 45 kV, 30 mA). The surface and cross-section morphologies were observed by FEI Inspect F field emission scanning electron microscopy (FESEM). The hysteresis loops were carried out with applied static magnetic fields up to 1200 kA/m by a vibrating sample magnetometer (VSM, VSM-220) at room temperature. The stress of films was analyzed by Zygo New View 7300 3D surface profiler.

3. Results and discussion

3.1. Crystallographic characteristics

Fig. 1 shows the XRD patterns of BaM films with various structures. Compared with the known JCPDF card (No. 84–0757) for $BaFe_{12}O_{19}$, it is found that the diffraction patterns of three groups of BaM films are similar and the diffraction peaks are only from BaM (00*l*) planes.

For the sake of plenarily characterizing the crystallographic structures of the BaM ferrite films, the full wave at half maximum (FWHM) of the BaM film rocking curves is measured, and then the c-axis dispersion angles are obtained. Because the (008) crystal plane of sample diffraction peaks is the strongest among all diffraction peaks, the (008) crystal plane is selected to measure the rocking curves. The rocking curves of the films are shown in Fig. 2. The FWHM of both d_{s100} and d_{b200} is much smaller than that of d_{s200}. This indicates that d_{s100} and d_{b200} obtain better perpendicular c-axis orientation than d_{s200}. For the structure of d_{s100}, the induced effect of the substrate makes the film get better perpendicular c-axis orientation. When the thickness of the film increases, the induced effect of the substrate is weakened gradually [21]. Then, the perpendicular c-axis orientation of d_{s200} deteriorates. For the structure of d_{b200}, the first 100 nm thick BaM film can be seen as a self buffer-layer. This self buffer-layer has strong induced effects on the second 100 nm thick BaM layer with perpendicular c-axis orientation, maintaining a good epitaxial growth and improving the crystal structures [1,18]. In addition, the first 100 nm thick BaM film of d_{b200} is annealed at 850 °C in air for 2 h, which can reduce the defects in the self buffer-layer as well as alleviate the stress due to the lattice mismatch and thermal mismatch [33].

3.2. Surface and cross-section morphologies

In order to further analyze the crystal structures of the samples and determine the quality of films, the film morphologies are observed by FESEM. Figs. 3 and 4 respectively present the cross-section and surface morphologies of the single-layer and bilayer BaM thin films. It is clearly demonstrated that the grains can be classified into two major categories: acicular shape and platelet shape. For the structure of d_{s100}, all of the grains are platelet-like (see Fig. 4a). However, for the structure of d_{s200} both the acicular grains and platelet-like grains are observed (see Fig. 4b), the acicular grains fuse each other and occupy a much larger area than the platelet-like grains. For the structure of d_{b200}, a lot of platelet-like grains can be found in the film. Previous studies have confirmed that the acicular grains are either c-axis in-plane oriented and/or randomly oriented, while the platelet-like grains are c-axis perpendicularly oriented [27,34]. Thus, the d_{s100} and d_{b200} obtain better perpendicular c-axis orientation than d_{s200}. The above results also agree with the rocking curves of samples (see Fig. 2) and indicate that with the decrease of film thickness, the perpendicular c-axis orientation has great improvement. The layered sputtering can enhance the growth of the perpendicularly c-axis oriented platelet-like grains and suppress the growth of the in-plane and randomly oriented acicular grains [18].