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Structure characterization and magnetic properties of barium hexaferrite films deposited on 6H-SiC with random in-plane orientation

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ABSTRACT

Oriented barium hexaferrite films grown on wide band-gap semiconductor substrates (such as SiC) are promising candidates for high-power microwave integrated devices. In this work, $BaFe_{12}O_{19}$ (BaM) films with *c*-axis randomly distributed in the film plane were prepared by direct current magnetron sputtering on *a*-plane 6H-SiC substrates. An insight into the orientation relationship and the epitaxial-like growth mechanism demonstrates that $(11\bar{2}0)$ planes of BaM have been grown on $(11\bar{2}0)$ 6H-SiC substrates. The random alignment of [0001] axis within the film plane was revealed by X-ray diffractions, X-ray pole figures, polarized Raman spectra and magnetic hysteresis loops. The BaM films on 6H-SiC substrates exhibited a large remanence ratio, M_r/M_s, of about 0.64 along in-plane axis and a relatively small one of ~0.2 for the out-of-plane hard axis, showing the potential of application in self-biased microwave devises.

1. Introduction

Hexaferrites have drawn increasing interest from both academic and industrial communities since the discovery in the 1950s due to their anisotropy magnetocrystalline properties, relatively high permeability and low eddy current losses [1]. During the past decade, hexaferrites with such unique properties have been found promising for many microwave applications [2]. For example, Z-type hexaferrite $Ba_3Co_2Fe_{24}O_{41}$ (Co_2Z) is of particular value for phase shifter and antenna substrate owing to its *a-b* plane aligned magnetic easy axis [1–3]. Great efforts have also been made to raise the cut-off frequency as well as the permeability of Co_2Z by preparing samples with the perpendicular *c*-axis crystallographic texture [4,5]. However, the application frequency of Co_2Z is still limited to the ultra high frequency (UHF, 0.3–1.12 GHz) region. At even higher frequencies as Q-band (30–50 GHz), M-type barium hexaferrite BaM with substantially higher magnetic anisotropy are required. In addition, just like the out-of-plane *c*-axis aligned Co_2Z, BaM films would also exhibit the distinct magnetic anisotropy with the easy axis perpendicular to the film normal if the *c*-axis (the magnetic easy axis) is randomly distributed in the film plane. In this case, the ferromagnetic resonant frequency of BaM is expected to be in the range from 36 to 54 GHz according to Kittel equation [6,7]. However, most of the previous studies focused on the out-of-plane *c*-axis oriented BaM films with lower application frequency, while the in-plane ones have seldom been reported due to the difficulty in preparation [8–10]. In fact, by virtue of its minimal self-demagnetization effects in the film plane, the latter is expected as a promising candidate for self-biased devices in microwave filters, phase shifters and delay lines [11–14]. Furthermore, a near-zero shape-demagnetizing factor that is critical for miniature microwave ferrite components is expected to be fulfilled only in this type of films rather than the soft magnetic Co_2Z hexaferrite or the out-of-plane *c*-axis oriented BaM films [14].

Another long-sought goal of the ferrite community has been the integration of magnetic microwave devices with semiconductor electronics [2]. Compared with the often used Si substrates, wide-gap semiconductor SiC holds significant advantages in the realization of higher performance, higher power handling capabilities, higher speed and smaller size devices when the frequency shifts into the microwave range [15]. In addition, SiC shares the same hexagonal crystal symmetry (*P*63/*mmc*) and comparable lattice parameters with BaM, and possesses higher temperature stability for the growth of high-quality hexaferrite materials than the majority of semiconductor substrates like Si and GaAs [15,16]. Up to now, however, BaM films have only been prepared on SiC with *c*-axis normal to the film plane [15–18].

In this work, we fabricated BaM films on 6H-SiC substrates with *c*-axis being randomly aligned within the film plane using direct current (DC) magnetron sputtering method [19,20], and investigated the microstructure and orientation relationship between BaM and 6H-

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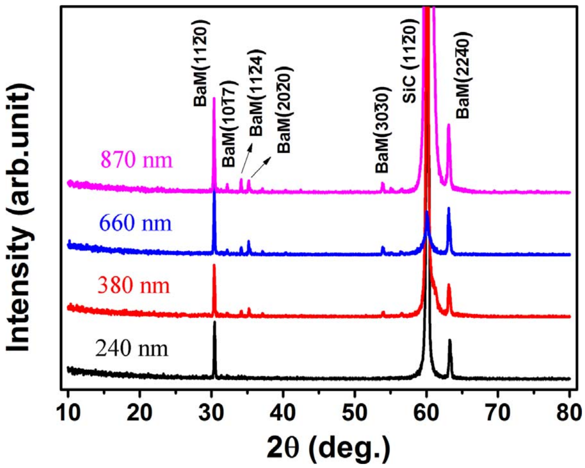

Fig. 1. X-ray diffraction patterns of the BaM films with different thickness deposited on *a*-plane 6H-SiC substrates.

SiC based on X-ray methods and polarized Raman scattering analysis, which has seldom been reported for this system. Through the magnetic hysteresis loop measurements, obvious manetocrystalline anisotropy and relatively large remanence ratio (~0.64) of the BaM films along in-plane axis were detected, implying potential application in the self-biased microwave devices.

2. Experimental

Film deposition was carried out using a DC magnetron sputtering technique as reported previously [19,20]. All BaM films were grown on (11$\bar{2}$0) planes of 6H-SiC at the sputtering power of 100 W, with the optimum sputtering gases of Ar-7% O_2 and substrates temperature set at room temperature. Amorphous films with different thickness about 240, 380, 660, and 870 nm were obtained when deposited for 30, 60, 90, and 120 min, respectively, and subsequently annealed at 920 °C for 30 min in air. Phase and crystal structures of the BaM films were studied by X-ray diffraction (XRD, Rigaku Smartlab, Cu K_α radiation). Surface morphology and film thickness was analyzed using scanning electron microscope (SEM; MERLIN VP Compact, Carl Zeiss, Germany). Room-temperature magnetic properties were obtained using a superconducting quantum interference device (SQUID-VSM, Quantum Design, US). The hysteresis loop measurements were conducted along three mutually perpendicular directions using vibrating sample magnetometry, with one direction being along the film normal (out-of-plane) while the other two being in the film plane (in-plane). The external magnetic field H is applied parallel to substrate surface for the measurement of two perpendicular in-plane directions, as well as perpendicular to substrate surface for the out-of-plane hard direction. The diamagnetic background of the 6H-SiC substrates has been subtracted for all samples to reveal the magnetic properties of the films only. Polarized Raman spectra were recorded using Raman spectrometer (RS; LabRAM HR Evolution, HORIBA Scientific, Japan) in a backscattering configuration, with a 633 nm laser used as the excitation source.

3. Results and discussion

In-plane c-axis oriented BaM films were obtained on (11$\bar{2}$0) 6H-SiC substrates as revealed by the strong diffraction peaks of (11$\bar{2}$0) and (22$\bar{4}$0) from the XRD patterns shown in Fig. 1. This result shows that a-cut plane BaM film was connected to a-cut (11$\bar{2}$0) SiC substrate, which is different from the case for sapphire substrate, where ($h0\bar{h}0$) peaks (m-plane) of BaM would appear on a-cut plane sapphire and ($hh\overline{2h}0$) peaks (a-plane) of BaM on m-cut sapphire. However, we cannot conclude that the c-axis of BaM was randomly distributed in the film plane only from the presence of the strong ($hh\overline{2h}0$) peaks. Directionally aligned c-axis in the film plane will exhibit the same θ–2θ X-ray scan results [19–21].

Actual orientation of the BaM c-axis was further verified by X-ray pole figures as shown in Fig. 2, where ψ is the angle between the film normal and the vector bisecting the incident and diffracted X-ray beams, and Φ is the azimuthal angle in the film plane. Fig. 2(a) represents the BaM (22$\bar{4}$0) reflections, which exhibit one spot on the apex, two pairs of spots at ψ≈60°, and another two pairs of spots at ψ=30°. Based on the above results, we tentatively conclude that both the spot on the apex and the two pairs of spots at ψ≈60° come from BaM (22$\bar{4}$0) crystal plane, while the spots at ψ=30° are from the 6H-SiC substrate. It may be incorrectly thought that the c-axis of BaM is aligned along two directions in the film plane, and the angle between the two in-plane directions is ~35°. However, we found that the spots at ψ≈60° and 30° are actually from the 6H-SiC substrate rather than the BaM film sample, as illustrated in Fig. 2(b). In addition, the BaM (22$\bar{4}$0) reflections at ψ=60° are expected to spread out across the whole ψ=60° ring that, however, was overlapped by the strong signal from the 6H-SiC substrate. Other reflections like BaM (10$\bar{1}$7), the strongest diffraction peak of BaM, were also measured, but the expected ring-like reflection signal was also overlapped by that from the 6H-SiC substrate. The actual orientation of the BaM films would be further verified by SEM, polarized Raman spectra and the magnetic hysteresis curves.

Surface microstructure images of the BaM films are shown in Fig. 3. Different from BaM on sapphire with parallel aligned acicular grains

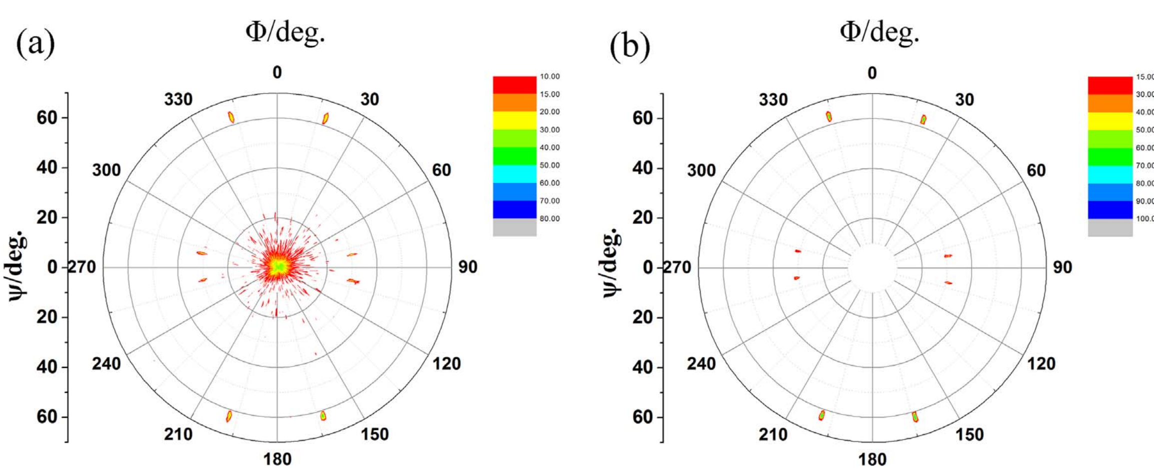

Fig. 2. X-ray pole figures of the 240-nm-thick BaM film and the 6H-SiC substrate without a film on it. (a) BaM (22$\bar{4}$0), (b) 6H-SiC at the same 2θ (63.29°) configuration as (a). Φ is the azimuthal angle in the film plane, and ψ is the angle between the film normal and the vector bisecting the incident and diffracted X-ray beams.