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Structure and magnetism of Ba-Hexaferrite films grown on single crystal 6-H SiC with graduated interfacial MgO buffer layers

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Structure and Magnetism of Ba-Hexaferrite Films Grown on Single Crystal 6-H SiC With Graduated Interfacial MgO Buffer Layers

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M-type barium hexaferrite films were processed by pulsed laser deposition on single-crystal 6-H silicon carbide substrates. MgO buffer and barrier layers were introduced to improve the film quality. Samples were characterized by X-ray photoelectron spectroscopy, atomic force microscopy, scanning electron microscopy, X-ray diffraction, vibrating sample magnetometry, and ferromagnetic resonance (FMR). X-ray $\theta - 2\theta$ diffraction measurements indicated a strong (0, 0, 2n) crystallographic alignment. The magnetization of the BaM film is comparable to bulk values ($4\pi M_s \sim 4320$ G). A derivative power FMR linewidth of 500 Oe was measured at 55 GHz for the as-deposited films. This paper explores a potential next generation of microwave and millimeter-wave monolithic integrated circuit technology based upon a wide band-gap semiconducting material.

***Index Terms*—Ba-hexaferrite, films, silicon carbide.**

I. Introduction

IT HAS been a long-standing goal of the ferrite materials and microwave device communities to integrate nonreciprocal microwave devices (i.e., circulators, isolators, phase shifters, etc.) with semiconductor devices platforms [1], [2]. An integral step in this development is the growth of ferrite materials on semiconductors. Previous attempts to grow spinel ferrites on GaAs lead to the disassociation of the semiconductor due to the high-temperature processing required for the growth of most ferrite materials. The growth of high-quality large crystals of wide band-gap semiconductors, such as GaN and SiC, now make it possible to revisit this problem. Here, we present the structural and magnetic properties of barium hexaferrite (M-type) films grown on single crystal 6-H silicon carbide and the effect of modifying the film-substrate interface.

II. Experiment

SiC is regarded as a promising wide band-gap semiconductor material for next generation electronics. The n-type 6H-SiC substrates used in this paper were acquired from Cree, Inc. The lattice mismatch between Ba (M-type) hexaferrite and the SiC basal plane is around 4.38% [3], [4]. Based on previous work [5], the optimal substrate surface preparation included boiling in $H_2SO_4 + H_2O_2$ and rinses in dilute HF and $HCl + H_2O_2$ solutions. The deposition was carried out by pulsed laser deposition (PLD) with a KrF Excimer laser operating at a wavelength of 248 nm with an energy per pulse of 200 mJ. The background gas and substrate temperature were 300 mtorr oxygen and 925 °C, respectively.

In this paper, we focus on processing schemes intended to mediate the strain and diffusion across the film-substrate interface. To this end, we have adopted the practice of using a thin graduated MgO multilayer in the early stages of film growth. The multilayer consists of MgO layers interwoven with BaM layers, for example, $[\text{MgO}(A)/\text{BaM}(B)]_n$ where A is the thickness of the MgO layer, B is the thickness of the BaM layer, and n is the number of repeats. The layer thickness is controlled by the number of laser pulses or shots incident upon each target. The first period is the thickest at $A = 80$ shots and $B = 150$ shots. Subsequent repeats have the MgO layer reduced by ten shots. After nine periods, the MgO growth is stopped and a thick BaM layer ($\sim$500 nm) is then grown. A single laser shot provides $\sim$0.1 nm of MgO and BaM, respectively. In this growth scheme the interfacial region of the film exists as a layered, and possibly alloyed, structure. The intention is to mediate the lattice mismatch between BaM and SiC. Attempts to achieve this by a pure MgO buffer lead to films having poor uniaxial magnetic anisotropy, possibly due to the large mismatch between MgO and SiC ($\sim$3.01%). Alternatively, films grown with the graduated buffer layer have demonstrated superior uniaxial magnetic properties to those without this modification. X-ray photoelectron spectroscopy (XPS) depth profiling measurements show that this modified layer also provides a barrier to the diffusion of Si into the BaM film. We believe the diffusion of Si into the BaM leads to a reduced anisotropy field.

III. Results and Discussion

The substrate surface quality is critical to the epitaxial growth of high crystal quality films. Even in the case of a perfect surface, SiC and BaM still have a 4.38% lattice mismatch, which is expected to introduce in-plane stress creating copious stacking faults and assorted lattice defects. In an earlier

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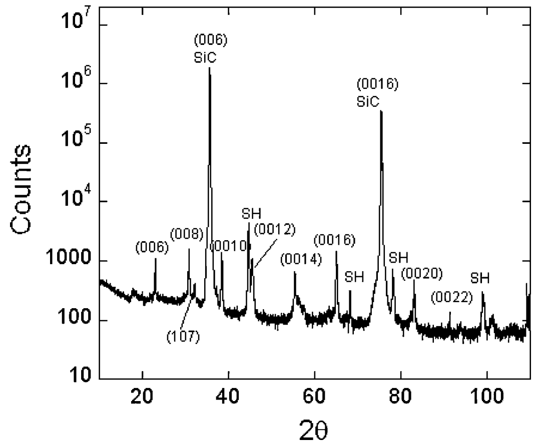

Fig. 1. X-ray $\theta - 2\theta$ diffraction pattern for Ba-hexaferrite (M-type) film. All significant diffraction features are referenced to (0,0,$2n$) indexes having space group P6/mmc (SH denotes substrate holder).

report [5], we measured a small grain microstructure having a large crystallite ($\sim$0.5 μm) embedded in a matrix of smaller grains ($<$ 0.1 μm diameter). Those films were measured to have a clear uniaxial anisotropy in the magnetic properties. The magnetic hysteresis loop squareness (i.e., Sq $= M_r/M_s$) approached 0.7. High Sq values are required, and highly desirable, for self-biased ferrite device applications [6]. For the highest quality films, we measured this squareness as low as 0.2. It is typical for hysteresis loop squareness values to track as the inverse of crystal quality metrics such as ferromagnetic resonance (FMR) linewidth [7]. The previously measured fine grain microstructure and high hysteresis loop squareness may account for the broad FMR linewidth ($\sim$2000 Oe) we reported in [5].

X-ray $\theta - 2\theta$ diffraction measurements using Cu kα radiation (see Fig. 1) reveal (0, 0, $2n$) diffraction peaks consistent with a pure phase magnetoplumbite crystal structure with strong c axis orientation perpendicular to the substrate plane.

Atomic force microscopy (AFM) images (a representative image is shown in Fig. 2) show 0.5–1-μm diameter grains, with no signs of the smaller grains as observed in the films reported in [5]. The crystallites appear to be densely packed and have their basal planes aligned parallel to the plane of the substrate. Upon close inspection, one sees that the basal planes are terraced, which may result from the layer-by-layer growth of the crystallites. This morphology may also result from crystal nucleation at screw dislocations on the surface of the SiC.

The dc magnetic properties were measured by vibrating sample magnetometry with a maximum magnetic field of 12 500 Oe aligned parallel and perpendicular to the sample plane. The hysteresis loops (see Fig. 3) confirm that the easy magnetic axis of the film was aligned perpendicular to the film plane, and the hard axis lies in the film plane. This is a required property for conventional circulator device applications. The anisotropy field is estimated by $16+/-1$ kOe, which is in reasonable agreement with the bulk $BaFe_{12}O_{19}$ value of 17 kOe. The FMR power derivative linewidths $(\Delta\ H)$ were measured to be 500 Oe in the as-deposited film without postdeposition annealing. This marks a significant decrease in ΔH from the results of [5].

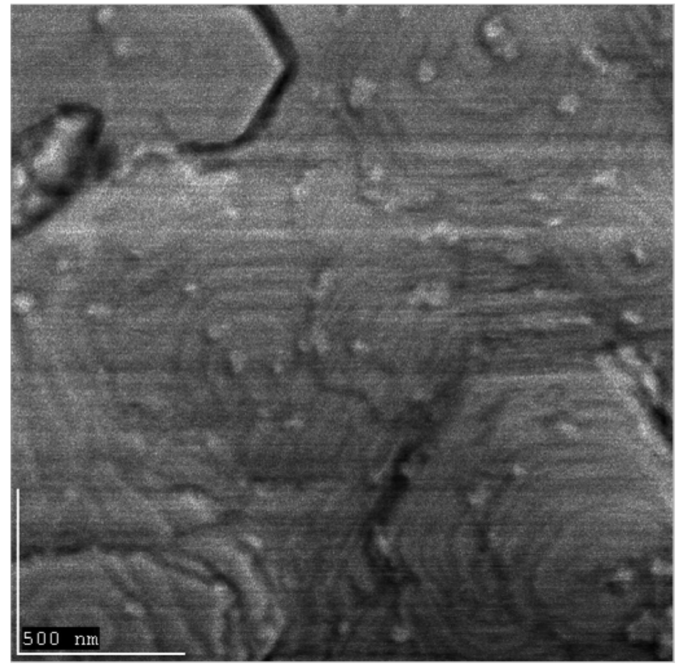

Fig. 2. AFM image processed in tapping mode illustrating hexagonal crystals oriented with c axis perpendicular to film plane.

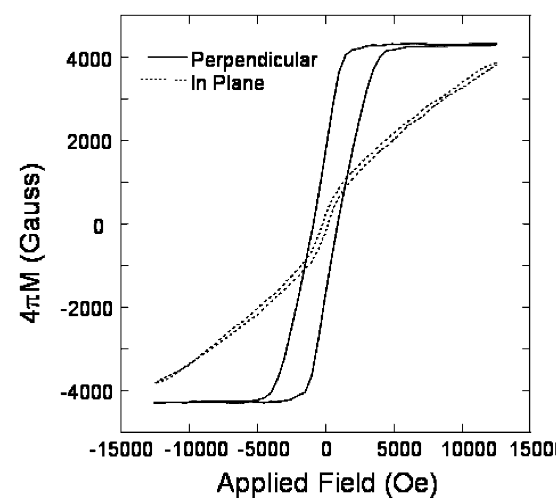

Fig. 3. Hysteresis loop obtained by vibrating sample magnetometry with maximum applied field of 12 500 Oe aligned parallel (dashed) and perpendicular (solid) to film plane. Data have not been corrected for demagnetization.

IV. Conclusion

High-quality epitaxial barium hexaferrite films have been successfully grown by PLD on single-crystal 6-H silicon carbide substrates. The films are characterized as being dense with 0.5 to 1-μm diameter hexagonal crystallites oriented with basal planes parallel to the substrate plane. The best of these films have a coercivity and FMR derivative power linewidth of 389 and 500 Oe, respectively. The use of a thin graduated MgO/BaM multilayer in the early stages of film growth proved to be essential in improving the crystallographic texture and improved magnetic properties ostensibly by reducing interfacial strain and diffusion of Si from the substrate into the film.

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