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Deposition of BaFe₁₂O₁₉ Thin Films by a New Injection-CVD Method

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Abstract : A new process of injection-MOCVD has been used to synthesize barium hexaferrite thin films on Al₂O₃ (0001) substrates. This new technique uses a liquid source of precursors dissolved in a convenient solvent. X-ray diffraction measurements have been performed to observe thin films lattices with respect to the orientation of the substrate : hexaferrite film epitaxy is observed with c-axis perpendicular to the substrate. Magnetic measurements have been performed in the plane and perpendicular to the film plane : they confirm the preferential orientation of films.

1. INTRODUCTION

Films of barium hexaferrite (BaM) are of great interest for magnetic recording [1] and for microwave circuit applications. In order to obtain BaM films several techniques have been tested [2] [3] [4] [5]. Here properties of BaFe₁₂O₁₉ thin films deposited by a new method called injection-CVD [6] are described. Results obtained for films grown on Al₂O₃ (0001) are presented.

2. EXPERIMENTAL PROCEDURE

In the new process presented here, solid precursors of Ba(thd)₂ and Fe(thd)₃ are dissolved in 1,2-dimethoxy-ethane ; the solution is placed in a hermetically closed vessel under argon which is connected to an injector. Droplets of few microliters are sequentially injected from this liquid source. Vapours of barium and iron precursors are produced by heating the injected droplets ; these vapours are then used in a conventional hot-wall CVD reactor.

Film growth has been performed under the following conditions :

Injection zone temperature	25 °C	Ba dilution	0.003 mol/l
Evaporator temperature	250 °C	Fe dilution	0.027 mol/l
Substrate temperature	800 °C		
Total pressure	5.0 hPa	Volume of droplets	5.0 µl
Oxygen flow	900 cm ³ /min	Number of droplets	700
Argon flow	300 cm ³ /min	Injection frequency	1 Hz

3. RESULTS AND DISCUSSION

3.1. X-Ray diffraction

Figure 1 displays the XRD pattern of a BaFe₁₂O₁₉ as-deposited film performed in the conventional $\theta/2\theta$ scanning mode ($\lambda_{Cu}=1.54056 \text{ \AA}$) ; the film is 2500 Å thick and the surface is 1×1 cm². For our films grown on Al₂O₃ (0001) the only intense peaks of the BaM phase are the (00 ℓ) indicating a very good orientation with the c-axis perpendicular to the film plane.

The $\theta/2\theta$ XRD scan indicates parallelism between the [001] direction of the BaM phase and the [001] direction of the sapphire substrate but reveals nothing about in-plane orientation. Figure 2 displays the {10 $\bar{1}$ 7} pole density of BaFe₁₂O₁₉ obtained for its Bragg angle. The six equivalent directions <107> appear for $\chi=33^\circ$ which indicates an epitaxial growth of the hexaferrite film. The full-width at half-maximum (FWHM) of the rocking curve performed on the 008 diffraction line of the Ba-ferrite is 0.79° showing the high orientation of the layer. This value of FWHM has to be compared with that of 0.32° obtained for the rocking curve performed on the 006 diffraction line of the sapphire substrate.

Figure 1 : XRD pattern of BaM film

Figure 2 : $\{10\bar{1}7\}$ pole density of BaM film

3.2. Magnetic measurements :

Figures 3 and 4 show the first magnetization curves performed with a vibrating sample magnetometer. The field was applied parallel and perpendicular to the film plane respectively. Barium hexaferrite has strong axial magnetocrystalline anisotropy ; the anisotropy of samples is determined by both the crystal anisotropy and the shape anisotropy of the thin film geometry. When the field is applied in the film plane the demagnetizing effect is negligible. This is not the case when H is perpendicular to the film, i.e. parallel to the easy direction of BaM ; the demagnetizing field factor has been evaluated using the formula for an oblate ellipsoid and has been corrected on figure 4.

Figure 3 : $H //$ to the film plane Figure 4 : $H \perp$ to the film plane

When the field is applied in the plane of the film, the observed anisotropy field is about 17000 Oe which is the value usually obtained on a single crystal. When the field is perpendicular to the film there is competition between shape and magnetocrystalline anisotropies. Until 1200 Oe the shape anisotropy is stronger so that the magnetization increases slowly. Then there is a strong increase in relation to the rotation and a quick development of magnetic domains. After 2000 Oe the magnetic moment increases more slowly by rotation mechanism of the domains until the saturation for a field of 10000 Oe. These two curves show that the easy direction of magnetization is the c-axis but also that the magnetic moments are not parallel to the c-axis but canted.

4. CONCLUSION

A new injection-CVD method has been successfully used to deposit epitaxial films of $\text{BaFe}_{12}\text{O}_{19}$ on sapphire (0001). Magnetic measurements show that the easy direction of magnetization is the c-axis which is perpendicular to the substrate, but that the magnetic structure is canted.

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