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Increase of Saturation Magnetization of Ba Ferrite Films Deposited by Adding Xe Into Sputtering Gas Mixture

K. Noma, N. Matsushita, S. Nakagawa and M. Naoe

Department of Physical Electronics, Tokyo Institute of Technology, 2-12-1 O-okayama, Meguro, Tokyo 152, Japan

Abstract. Ba ferrite thin films with well *c*-axis orientation were deposited by means of three sputtering systems and the relationships among their microstructure, magnetic characteristics and plasma parameters were discussed. It was clearly confirmed that the suppression of heavy bombardment of energetic particles to the surface of the growing film was the most important for attaining the excellent crystallographic and magnetic characteristics.

1. INTRODUCTION

Since the bulk materials of a magnetoplumbite type of Ba ferrite ($\text{BaM: BaO} \cdot 6\text{Fe}_2\text{O}_3$) exhibit the magnetic characteristics such as saturation magnetization $4\pi M_s$ of 4.8 kG and uniaxial anisotropy constant K_u of $3.3 \times 10^5 \text{ J/m}^3$, they have been used widely as components in microwave devices as well as permanent magnets. Recently, the Ba ferrite films with well *c*-axis orientation and large $4\pi M_s$ are required for the applications to recording media and milliwave isolators. In this study, the plasma-free sputtering techniques were developed for the strictly control of microstructure in Ba ferrite films and then, they were able to possess the excellent crystallinity and magnetism by adjusting the plasma parameters even at the substrate temperature T_s as low as 475 °C.

2. EXPERIMENTS

All specimens were deposited on thermally oxidized Si (SiO_2/Si) wafers with the thickness of 200 nm. The sintered ferrite plates with the compositions of $\text{BaO} \cdot n\text{Fe}_2\text{O}_3$ ($n = 4.0, 5.5, 6.5$) were used as the sputtering targets and the mixture of Ar and O_2 (or Xe, Ar and O_2) were used as the sputtering gas. The crystallographic and magnetic characteristics of the as-deposited films were evaluated by X-ray diffractometry, vibrating sample magnetometer and torque magnetometer, respectively. The plasma parameters such as the plasma potential V_p and the floating potential at substrate V_f were measured by Langmuir's single probe method.

3. RESULTS AND DISCUSSION

3.1. Deposition with the heavy bombardment of energetic particles

The depositions were carried out by means of the conventional RF Diode Sputtering (RFDS) apparatus using the mixture of Ar and O_2 as the sputtering gas at the total pressure p_{total} of 0.50 Pa. In this case, V_p took a very large value of +30 V and V_f was -8 V. Therefore, Ar ions bombarded the surface of the growing film at the energy of 38 eV, of which the value was large enough to cause the selective resputtering of the deposited Ba atoms from the film surface, and it led to the remarkable decrease of Ba content in the

Figure 1: Schematic illustrations of three sputtering systems and their plasma distributions.

Figure 2: T_s dependences of $\Delta\theta_{50(00n)}$ and $4\pi M_s$ for three sputtering systems.

film. In addition, the γ -electrons ejected from the sputtering target were accelerated across the sheath region and bombarded the film surface. The energy of bombarding electrons reaches to several hundreds eV and it caused the elevation of the substrate temperature T_s and the deconstruction of crystal. So, the Ba ferrite films with well c -axis orientation were deposited at T_s only in very narrow range from 620 °C to 760 °C using the target with a Ba-excessive composition of $\text{BaO} \cdot 4.0\text{Fe}_2\text{O}_3$. The (00n) dispersion angle of BaM crystallites $\Delta\theta_{50(00n)}$ was 1.8° at T_s of 700 °C. However, the saturation magnetization $4\pi M_s$ and the uniaxial magnetic anisotropy constant K_{u1} were as comparatively small as 4.1 kG and $1.9 \times 10^5 \text{ J/m}^3$, respectively. These results suggest that the heavy bombardment of energetic particles during deposition may cause the resputtering of Ba atoms and the formation of thick grain boundaries with the segregated non-magnetic phase.

3.2. Deposition with the unnegligible bombardment of recoiled Ar atoms

In the Facing Targets Sputtering (FTS) apparatus, the DC glow plasma is confined between the two facing targets and the bombardment of charged particles such as Ar ions, γ -electrons and negative oxygen ions to the surface of the growing film was extremely reduced. While the films were deposited in FTS apparatus with the mixture of Ar and O₂ as sputtering gas at the p_{total} of 0.27 Pa, V_p and V_t were as small as 2 V and -8 V, respectively and T_s at which the films with well c -axis orientation were attained was as relatively low as 550 °C and the $\Delta\theta_{50(00n)}$ was as very small as 1.0°. However, the target composition for depositing the films with the stoichiometric composition ($\text{BaO} \cdot 6\text{Fe}_2\text{O}_3$) was a little Ba-excessive ($\text{BaO} \cdot 5.5\text{Fe}_2\text{O}_3$) and $4\pi M_s$ took the maximum value of 4.7 kG at T_s of 600 °C. The decrease of Ba content in the film seemed to be caused by the unnegligible bombardment of Ar atoms recoiled from target surface and the smaller $4\pi M_s$ seemed to be caused by the incorporation of Ar atoms into the film body.

3.3. Deposition without any significant bombardments

The recoil of Ar atoms is intrinsic phenomena in the sputtering of Ba ferrite because of the large difference in atomic mass between Ar and Ba atoms and the energy of recoiled Ar atoms reaches to several hundreds eV. This phenomena seemed to be sufficiently suppressed by adding Xe into the sputtering gas, since Xe and Ba have almost same atomic mass. V_p was maintained at the constant value of 2 V but V_t increased gradually from -8 to -5 V and the electron temperature T_e at the center of plasma column decreased from 3.4 to 2.6 eV with increase of the partial Xe pressure p_{Xe} . These results suggest that the addition of Xe was effective not only for suppressing the recoil of Ar atoms but for minimizing the ejection of the energetic species from the plasma. XRD diagrams indicated that the films deposited at T_s of 600 °C using the targets with the Fe-excessive composition of $\text{BaO} \cdot 6.5\text{Fe}_2\text{O}_3$ were composed of the crystallites of almost single phase Ba ferrite and that $4\pi M_s$ and K_{u1} reached to the values of 5.1 kG and $3.3 \times 10^5 \text{ J/m}^3$, respectively. The significant increase of $4\pi M_s$ and K_{u1} seemed to be correlated to the increase in the number of atomic layers in S-blocks.

In addition, the Ba ferrite films with well c -axis orientation and $4\pi M_s$ of 4.7 kG were deposited even at T_s as low as 475 °C. The decrease of T_s was owing to the elimination of significant bombardments of energetic particles to the surface of the growing films. Such a low T_s enabled us to use the glass slides and the semiconductor sheets as substrates instead of SiO₂/Si wafers.

4. CONCLUSION

The microstructural and magnetic characteristics of Ba ferrite films deposited by means of three sputtering systems were investigated in detail on several sputtering parameters. It was cleared that the heavy bombardments to the surface of the growing films influence on the crystallization and the decomposition in the films and the excellent magnetic characteristics of $4\pi M_s$ of 4.7 kG were attained at the optimized sputtering conditions such as the T_s of as low as 475 °C, using the targets with the Fe-excessive composition of $\text{BaO} \cdot 6.5\text{Fe}_2\text{O}_3$ in the FTS apparatus and the mixture of Xe, Ar and O₂ as sputtering gas.

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