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Soft magnetic properties and high frequency characteristics of Fe - O nanocrystalline alloy films

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Abstract: The effect of oxygen - doping and thickness has been studied on soft magnetic properties and high - frequency characteristics of Fe - O alloy thin films prepared by a helicon - plasma - enhanced RF magnetron sputter - deposition at room temperature. A reduction in coercivity due to grain refinement was achieved using very low dose of oxygen which did not lead to the formation of crystalline Fe oxides with the low saturation magnetization. The real part (μ') of permeability has a high value of 1100 and is maintained up to 1 GHz below 150 nm for the relative O_2 flow ratio of R_{O_2} = 2.4%.

Key words: oxygen - doping; thickness; soft magnetic properties; high frequency characteristics
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Fe - O纳米晶合金膜的软磁和高频特性

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摘要:采用等离子体增强射频磁控溅射沉积方法,在室温下制备了 Fe - O合金薄膜。研究了氧的掺杂量和薄膜厚度对薄膜软磁和高频特性的影响。结果发现少量氧的掺杂不导致低饱和磁化强度铁氧化物的形成,但可使薄膜晶粒细化,矫顽力下降。在薄膜厚度低于 150 nm且氧气与氩气相对流量比为 2.4%的条件下,薄膜的实部磁导率高达 1100且能够维持到 1GHz

关键词:氧掺杂;厚度;软磁特性;高频特性

0 Introduction

There are growing demands of magnetic thin film materials for high - radio - frequency (GHz) use: magnetic recording heads, integrated thin - film inductors, and electromagnetic noise absorbers[1-6]. Traditional Fe oxide films are widely used in high - frequency applications by light of high resistivity, while the low magnetization, high temperature for preparation and complicated crystalline structure inhibit its use. Soft magnetic amorphous ferromagnetic materials such as CoZrNb [7] and CoZrTa [3] have been typical for boosting the inductance and enhancing the quality fac-

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tor of integrated inductors [8]. However, the low electrical resistivity and multiple magnetic resonances in the permeability spectra of these materials restrain the enhancement of the inductor characteristics [9,10]. To overcome these difficulties and increase the efficiency of micromagnetic devices, a number of approaches have been taken and developed. For example, soft magnetic $Ni_{0.81}Fe_{0.19}$/($Fe_{0.7}Co_{0.3}$)0.95$N_{0.0}$/$Ni_{0.81}Fe_{0.19}$ thin film which has a sandwich structure[11], and Co_xFe_{100-x} multilayer which consists of discontinuous metal layers with native oxide surfaces[12], have been reported. Important progresses have also been attained by granular metal/insulator composites, in which Fe-rich crystalline nanoparticles with high magnetic moments are embedded in a nonmagnetic insulator matrix, such as Al_2O_3 [13], ZrO_2 [4], HfO_2 [5,14], SiO_2 [15], Si_3N_4 [16]. However, soft magnetic properties, large magnetization, and high resistivity are not simultaneously compatible because high electrical resistivity comes out with increasing oxide fraction and at the expense of the volume-averaged saturation magnetization.

In pure Fe films prepared by the conventional thermal evaporation or sputtering method, a high saturation magnetization (M_s = 2.15 Wb/m^2) is realized, but good soft magnetic features cannot be obtained because of their large magnetocrystalline anisotropy and magnetostriction which lead to large magnetic coercivity. To improve soft magnetic properties and further increase electrical resistivity for high-frequency applications, the doping effect of light elements (B, C, N and Si) has been extensively investigated [17-19], although the reduction in M_s is an unwanted trade-off. Until now, little research has been done on the improvement of soft magnetic properties and high-frequency characteristics of Fe-O thin films (not Fe-oxide thin film) with changing amounts of oxygen.

In this paper, we describe a preparation method of soft magnetic Fe-O alloy thin films which reveal a good high-frequency magnetic response and elucidate the relationship between the magnetic properties and oxygen gas flow ratio. The film thickness effects on the high-frequency characteristics of these films have also been studied.

1 Experiment

A helicon plasma enhanced RF magnetron sputtering system were used to prepare Fe-O thin films. This technique was developed based on conventional RF magnetron sputtering and the helicon waves produced by an RF induced coil which was placed on the top part of an RF magnetron cathode. It has some advantages over conventional RF magnetron sputtering, e.g. smooth dense films can be deposited at low substrate temperatures, because of its high plasma density (10^{12} - 10^{13} cm^3) and high ionization efficiency. Also, plasma damage of the film surface is minimized owing to the large distance between the substrate and the target (220 mm) and the RF coil attached to the planar magnetron cathode which can restrict plasma in the target area. Fe-O thin films of 150 nm thickness were prepared by helicon plasma enhanced RF magnetron sputtering (Ar+O_2 atmosphere) with a target power of 100 W and a RF coil power of 40 W. The sputtering chamber was evacuated down to lower than 5x10^{-5} Pa (as the base vacuum level) with a turbo-molecular pump and the deposition pressure was 0.1 Pa. A pure Fe (99.99%) target (50 mm in diameter) was sputtered in high purity argon and oxygen atomsphere. For obtaining Fe-O thin films with different oxygen contents, the relative O_2 flow ratio, R_{O_2} = [O_2 flow rate]/[Ar flow rate + O_2 flow rate] (in %), was changed by fine control of mass flow meters of O_2 and Ar gases. The Si wafers and glass plates were used as the substrates which were maintained at room temperature during the deposition. The pretreatment of the Si wafers and glass substrates was carried out by ultrasonic cleaning in an ethanol solution. The in-plane magnetic anisotropy of as-deposited Fe-O thin films was induced by a static magnetic field of about 24 kA/m (300 Oe) which was applied parallel to the substrate surface during deposition.

The structure of the thin films was analyzed by a X-ray diffraction equipment (XRD) in the standard Bragg-Brentano geometry using a rotor-type Cu target. The magnetic properties were investigated using a superconducting quantum interference device (SQUID) magnetometer. The frequency dependence of initial permeability at room temperature was evaluated using a high - frequency permeameter (Ryowa PMF - 3000). The electrical resistivity was measured with a conventional four - point probe method.

2 Results and discussion

The dependence of the magnetic and electrical properties of Fe - O thin films on R_{O_2} was investigated. With R_{O_2} increasing from 0% to 3. 6%, the saturation magnetization shows a slight decrease, while the magnetic coercivity, H_{ce} (easy axis) and H_{ch} (hard axis), exhibit a significant change. Simultaneously, the electrical resistivity monotonically increases. At R_{O_2} = 2 4%, H_{ce} and H_{ch} reveal a minimum of 3 Oe with a resisitivity of about 126μΩ cm.

XRD measurement results of Fe - O thin films with R_{O_2} = 0 - 3. 6% indicate that all films reveal an Fe(110) texture and no other diffraction peak is observed in the wide 2θ range of 35 – 100°. No evidence was found for the formation of Fe oxides up to R_{O_2} = 3. 6 %. This means that the obtained films exist as a form of single alloy phase and oxygen atoms are dissolved in the bcc Fe lattice. It is not found that the (110) peak shifts to a lower 2θ side up to R_{O_2} = 2 4 %, and this peak position is still consistent with that of pure Fe film. When the R_{O_2} increases up to 3. 6 %, the (110) peak clearly shifts to a lower 2θ side (the lattice expands) and a uniform stress appears in this film. To further characterize the film crystal structure, we calculated the crystallite size (D) of the films from the Scherrer's equation [20]. D is 6. 7, 6. 0, 5. 8, and 5. 6 nm for R_{O_2} = 0, 1. 2, 2. 4, and 3. 6 %, respectively. This indicates that the thin films are composed of small nanograins. The film thickness of 150 nm is much larger than these D values; consequently, it is considered that many (110) oriented nanocrystals compose the highly oriented Fe - O alloy thin films.

It is also found that when R_{O_2} was about or less than 2. 4%, the obtained films had very good in - plane uniaxial anisotropy as demonstrated by the hysteresis loops along easy and hard directions shown in Fig. 1 (a and b). The introduction of a very small quantity of O_2 lead to the decrease of H_{ce} from 9 Oe for a pure Fe thin film (R_{O_2} = 0 %) to 3 Oe for a Fe - O alloy thin film (R_{O_2} = 2. 4 %). Moreover, a large anisotropy field (H_k = 18 Oe) is observed for the R_{O_2} of 2. 4 %, suggesting that this film has a high ferromagnetic resonance frequency (f_r). This soft magnetic feature can be attributed to the decrease of Fe magnetocrystallic anisotropy with the introduction of oxygen atoms into the Fe nanocrystalline lattices.

Figure 2 shows the frequency (f) dependence of permeability for the Fe - O alloy thin films prepared at R_{O_2} = 0 and 2. 4 %, repectively. Clearly, the f_r increases as R_{O_2} increases. For R_{O_2} = 2. 4 %, the f_r reaches a value of 1. 7 GHz while the real part (μ') of permeability still has a high of 1100 and flattens up to 1 GHz. Moreover, it was demonstrated that oxygen - doping also enhances the in - plane uniaxial magnetic anisotropy of Fe films and thus can be applied beneficially to increase high - frequency permeability which is needed in microwave application fields.

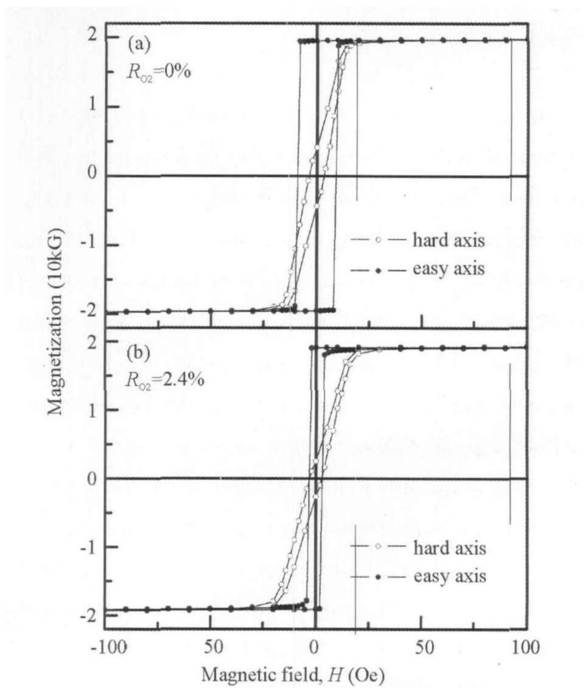

Fig. 1 Hysteresis loops and magnetic anisotropy field of Fe - O thin films deposited at R_{O_2} = 0 and 2. 4%

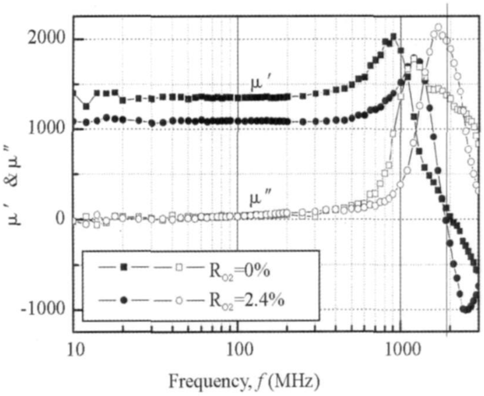

Fig. 2 High frequency characteristics of Fe - O thin films deposited at (easy axis) R_{O_2} = 0 and 2.4%

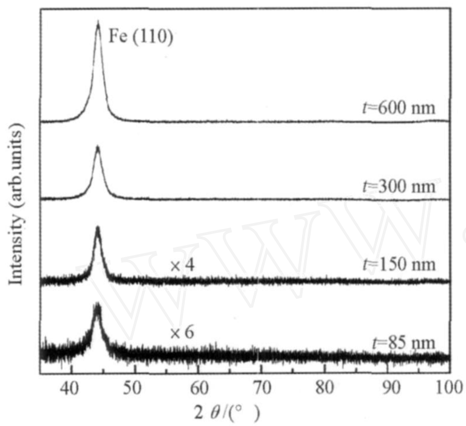

Fig. 3 X - ray diffraction patterns of Fe - O alloy thin films as a function of film thickness

In order to apply these films to high - frequency magnetic devices, it is necessary to understand the film thickness effects on their high - frequency characteristics. Figure 3 shows XRD measurements on Fe - O thin films with R_{O_2} = 2.4% and the film thicknesses from t = 85 to 600 nm. It is clearly seen that all films show only a Fe (110) peak, the position of which is also consistent with that of pure Fe film, and no other diffraction peak is observed in the wide 2θ range of 35 - 100°. This indicates that these films have the same Fe (110) texture with increasing t up to 600 nm.

Figure 4 shows the hysteresis loops along easy and hard directions and magnetic anisotropy field of Fe - O thin films with R_{O_2} = 2.4% and the film thicknesses from t = 85 to 600 nm. It is found that the film with t = 85 nm has larger H_c values (H_{ce} = 11 Oe and H_{ch} = 4Oe) and smaller H_k value (H_k = 14 Oe). When t ≥ 150 nm, the films have the values of H_c ≤3Oe and H_k ≥18Oe.

In order to further examine the high - frequency magnetic power loss in these films, we measured their initial permeability. Figure 5 shows the f dependence of permeability for the Fe - O alloy thin films with R_{O_2} = 2.4% and the film thicknesses from t = 85 to 600 nm. The real part (μ′) of permeability has a high value larger than 1100 and flattens up to 1 GHz for t = 85 and 150 nm, while μ′ starts to decrease at 0.7 GHz for t = 300 nm and at 0.3 GHz for t = 600 nm. On the other hand, for this thickness range of t = 85 to 600 nm, the effective permeability of these Fe - O alloy thin films is above 1000 and maintained up to 1GHz. In particular, in case of t = 150 and 300 nm, effective permeability is retained up to 1.6 GHz. Therefore, the Fe - O alloy thin films show excellent the high - frequency characteristics, which is interpreted to be due to the high electrical resistivity and magnetic anisotropy field.

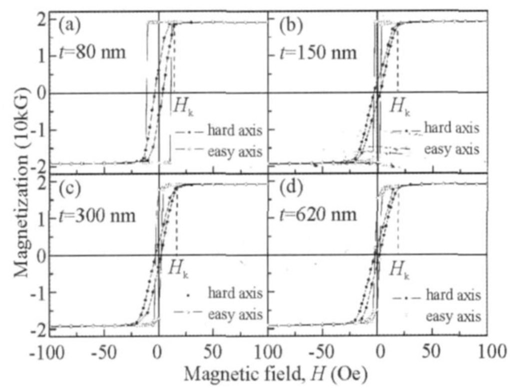

Fig. 4 Hysteresis loops and magnetic anisotropy field of Fe - O thin films as a function of film thickness

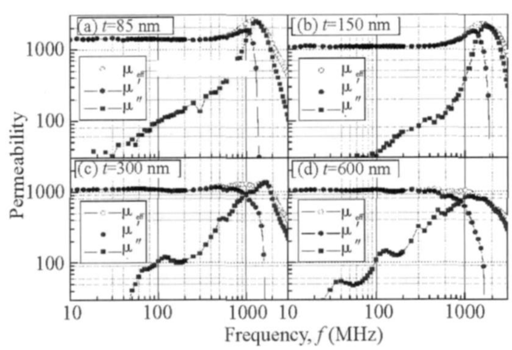

Fig. 5 High frequency characteristics of Fe - O thin films as a function of film thickness

3 Conclusion

Using the helicon plasma enhanced RF magnetron sputtering system, we prepared oxygen - doped magnetic Fe - O alloy thin films (not Fe - oxide thin film) by varying the R_{O_2} value during sputtering. The effect of oxygen - doping and film thickness has been studied on soft magnetic properties and high - frequency characteristics of as - deposited Fe - O alloy thin films. The incorporation of oxygen into the Fe film (lattice) refines grains, leading to the soft magnetic characteristics. For the thin films produced at R_{O_2} = 2.4 %, the real part (μ') of permeability has higher values than 1100 and is maintained up to 1 GHz below t = 150 nm. The effective permeability of these Fe - O alloy thin films is above 1000 and maintained up to 1 GHz below t = 600 nm.

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