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Optimized soft magnetic properties and high frequency characteristics of obliquely deposited Co–Zr thin films

Zhengmei Zhang, Xiaolong Fan[1], Min Lin, Dangwei Guo, Guozhi Chai and Desheng Xue[1]

Key Laboratory for Magnetism and Magnetic Materials of the Ministry of Education, Lanzhou University, Lanzhou 730000, People's Republic of China

E-mail: xueds@lzu.edu.cn and fanxiaolong@lzu.edu.cn

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Abstract

$Co_{100-x}Zr_x$ ($x \leqslant 16.7$) films with designable in-plane uniaxial magnetic anisotropy have been prepared by oblique sputtering at room temperature. It has been found that the introduction of a small amount of Zr into obliquely deposited Co films results in excellent soft magnetic properties and high frequency responses. We have qualitatively explained that such an optimization comes from the enhancement of exchange coupling between the Co grains in the films by using the random anisotropy model. Thus, the magnetocrystalline anisotropy of grains can be suppressed and the 'obliquely induced' anisotropy can become dominant in determining the static and dynamic properties of the obliquely deposited films. Therefore, films with designable static and dynamic properties can be easily obtained by controlling the deposition angles, as illustrated in our data.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Recently, there has been an increasing demand for soft magnetic films in high frequency applications such as soft underlayers for perpendicular media, magnetic recording write heads and thin film wireless inductor cores [1–14]. For these films, a sufficiently large in-plane anisotropy field H_k is required to keep a high ferromagnetic resonance frequency f_r, since f_r is proportional to $(4\pi M_s H_k)^{1/2}$, where $4\pi M_s$ is the saturation magnetization. On the other hand, when the in-plane anisotropy field is very large, the permeability μ is low, because in that case it is difficult to rotate the magnetization out of the anisotropy direction. Therefore, an optimized balance between f_r and μ is essential for the application of soft magnetic films at different working frequencies.

It is well established that magnetic transition metal films deposited at oblique angles exhibit a spontaneous in-plane uniaxial magnetic anisotropy (IPUMA) [15]. The in-plane easy axis (EA) of such an 'obliquely induced' anisotropy is found to be perpendicular to the projection of the oblique deposition direction [16], and its intensity depends on the angle of deposition [17, 18]. However, in these films, the magnetocrystalline anisotropy of individual column grains will degrade the soft magnetic properties [19–22]. Therefore, we have attempted to weaken such an effect of magnetocrystalline anisotropy of column grains by means of an enhancement of exchange coupling between the grains by doping the element Zr, so that the 'obliquely induced' anisotropy can play a dominating role in determining the magnetic properties of the films deposited at oblique angles. As a result, an '*in situ*' manipulation of the IPUMA as well as the dynamic properties can be achieved in the as-deposited films without any post-treatment.

2. Experiment

$Co_{100-x}Zr_x$ ($x \leqslant 16.7$) thin films with thickness ~ 100 nm were prepared by radio frequency (rf) sputtering onto Si (1 1 1) substrates ($10 \times 10\,mm^2$) attached to a water cooling system.

[1] Authors to whom any correspondence should be addressed.

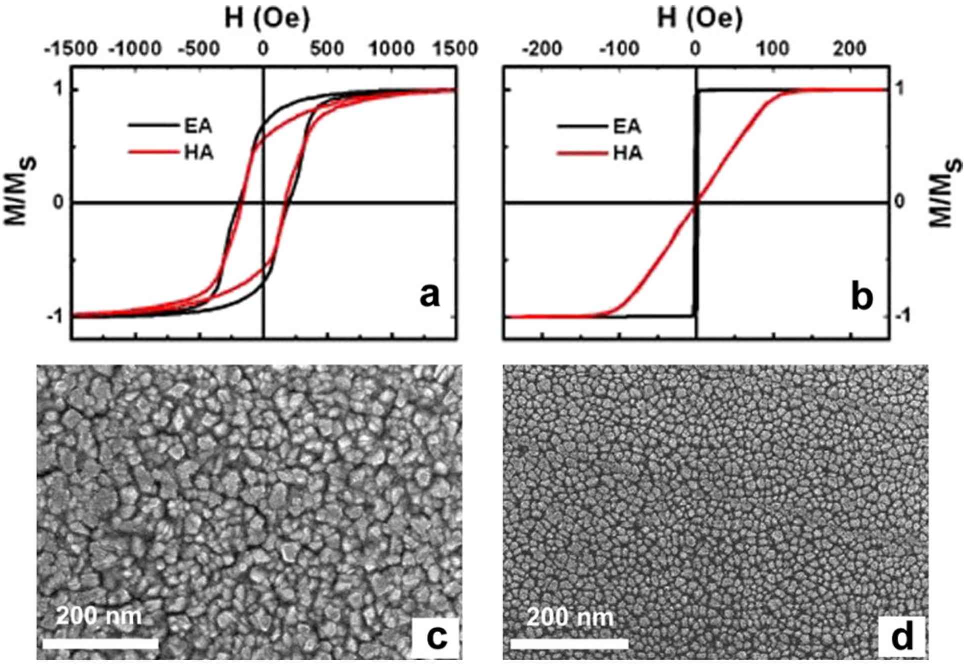

Figure 1. In-plane hysteresis loops ((*a*) and (*b*)) and surface images ((*c*) and (*d*)) for Co film (left column) and $Co_{93.5}Zr_{6.5}$ film (right column). Both the samples were deposited at 23°.(Colour online.)

A Co target, 70 mm in diameter and 3 mm in thickness, on which Zr chips were placed in a regular manner, was used. The composition of the deposited films was adjusted by controlling the number of Zr chips. The oblique sputtering angle is defined as the angle between the normal direction of the target and the line from the target centre to the substrate centre, as shown in figure 1 of [23]. The films were deposited with an oblique angle ranging from 0° to 37° without any applied field, and the direction of the in-plane EA was found to be perpendicular to the deposition direction. The background pressure was less than 2×10^{-5} Pa, the working Ar pressure was 0.15 Pa with an Ar flow rate of 20 SCCM (SCCM denotes cubic centimetre per minute at STP) and the rf power density was 1.7 W cm^{-2}. The compositions and the crystallographic properties of the films were measured by energy dispersive x-ray spectroscopy (EDS) and field emission scanning electron microscopy (SEM, Hitachi S-4800), respectively. The static magnetic measurements were performed using a vibrating sample magnetometer (Lakeshore model 7304). The microwave permeability measurements of the films were carried out with a PNA E8363B vector network analyzer using the microstrip method from 100 MHz to 5 GHz. All the above measurements were performed on the as-deposited samples at room temperature.

3. Results and discussion

Figure 1(*a*) shows the hysteresis loops for a 100 nm Co film deposited obliquely on Si(1 1 1) at 23°. The difference between the hysteresis loops measured along the EA and hard axis (HA) exhibits an in-plane magnetic anisotropy. The saturation magnetization of this film is about 1.6 T, which is slightly less than the bulk value ∼ 1.7 T in Co. The coercive force H_c is

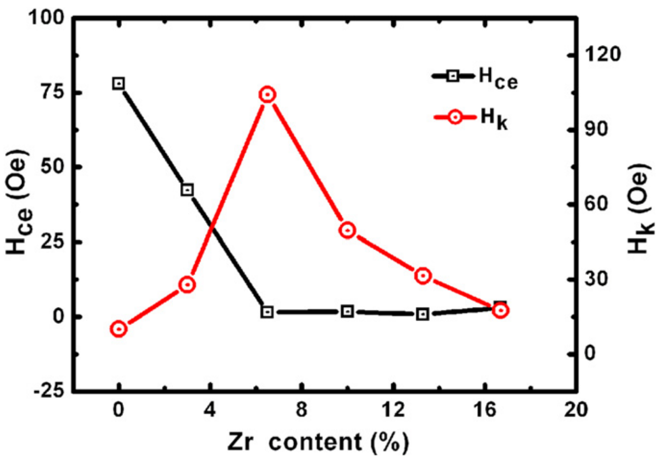

Figure 2. EA coercivity H_{ce} and IPUMA field H_k of Co–Zr films versus Zr concentration. All samples were deposited at 16°. (Colour online.)

large, 193.5 Oe along the EA and 169 Oe along the HA, and the anomalous shape of the hysteresis loops indicates a complexity of the domain structure and magnetization reversal process. However, after doping a small amount of Zr, the Co–Zr films display excellent soft magnetic properties and well-defined spontaneous IPUMA, as shown in figure 1(*b*). The loop along the EA is basically a rectangle with a residual magnetization ratio $M_r/M_s \sim 1$, which indicates that in the remanence state (at zero field) the magnetization aligns almost along the EA direction. The relatively small H_c along the EA (H_{ce}), less than 1.5 Oe, means that the re-orientation of magnetization easily occurs by applying a relatively small field along that direction. The HA loop is basically a closed curve without hysteresis, indicating that the change in magnetization takes place through its rotation by the field. The angle θ between

M_s and the EA is given by $\sin\theta = H/H_k$ (when $H < H_k$), where H_k is the in-plane magnetic anisotropy field. For this film, H_k is about 115 Oe, and the saturation magnetization is about 1.3 T. The results shown in figure 1 demonstrate that the introduction of Zr into the obliquely deposited Co films can significantly decrease H_{ce} and simplify the domain structure and the magnetization process.

It is speculated that such a profound improvement of soft magnetic properties results from the small effective magnetocrystalline anisotropy in an assembly of column grains, which arises by the effect of exchange coupling between the grains. According to the random anisotropy model [24], the effective anisotropy constant K_{eff} decreases with decreasing crystal size D according to the equation $K_{eff} = K_1(D/L_{ex})^6$ as long as D is smaller than the exchange length L_{ex}, where K_1 is the magnetocrystalline anisotropy constant of an individual grain. For our case, the average grain size of the films decreases from 30 nm to less than 10 nm on doping 6.5% Zr, as shown in figures 1(*c*) and (*d*). Supposing that the $L_{ex} \sim 10$ nm for Co grains is insensitive to the Zr doping [25], it is, therefore, considered that the magnetocrystalline anisotropy is substantially weakened, and the shape anisotropy of the thin film (makes the magnetization arrange in-plane) and the obliquely induced anisotropy (makes the magnetization arrange along the in-plane EA) become dominant. As a result, the soft magnetic properties are optimized.

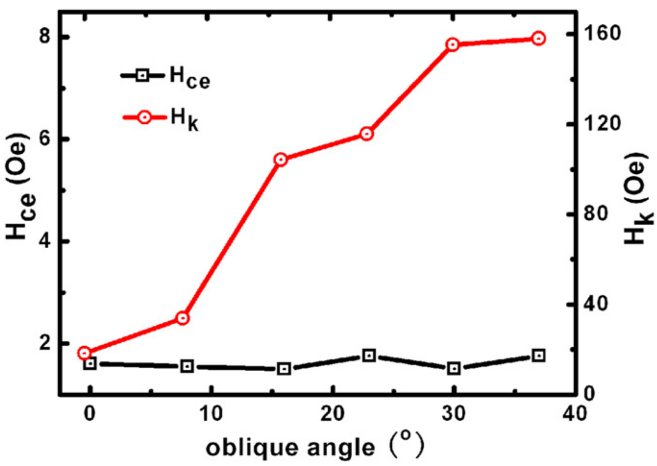

Figure 3. EA coercivity and IPUMA field H_k of $Co_{93.5}Zr_{6.5}$ soft magnetic films as a function of oblique angle. (Colour online.)

The static magnetic property of the 16° deposited Co thin film with different Zr concentrations x was studied systematically, as shown in figure 2. H_{ce} of the Co–Zr films is significantly reduced from ~78 Oe for the Co film to ~1 Oe when x is about 6.5 at%, while it remains constant when x is above 6.5%. H_k shows a maximum ~105 Oe at $x = 6.5\%$ and then decreases with x. As a result, we have chosen $Co_{93.5}Zr_{6.5}$ as the preferable composition, and studied the oblique angle dependence of the static properties, as shown in figure 3. As expected, a continuous increase in H_k as a function of oblique angle can be observed. H_k for the $Co_{93.5}Zr_{6.5}$ film deposited at 37° is about 158 Oe, almost ten times larger than 16 Oe found for the film deposited at normal incidence. H_{ce} of the films maintains low values of ~1 Oe which is insensitive to the

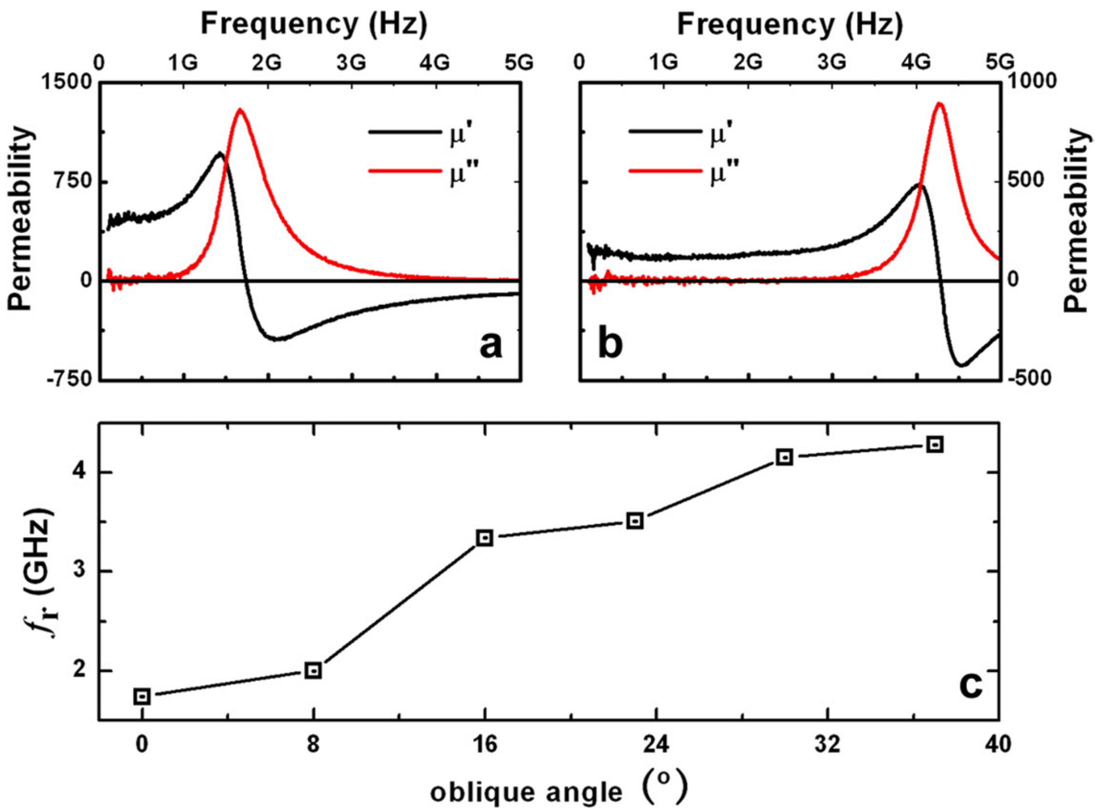

Figure 4. Frequency dependence of the relative complex permeability of $Co_{93.5}Zr_{6.5}$ films deposited at oblique angles of ~0° (*a*) and ~37° (*b*), respectively. Oblique angle dependence of the resonance frequency of Co–Zr films (*c*). (Colour online.)

deposition angle. Therefore, we can adjust the oblique angle to design the IPUMA in those films.

The increase in H_k results in a significant change in the high frequency performance of these films. Frequency dependences of the real (μ') and imaginary (μ'') components of the relative permeability of the $Co_{93.5}Zr_{6.5}$ films are shown in figures 4(*a*) and (*b*). For the film deposited at $\sim 0°$ with a weak IPUMA, μ' maintains a high value of $\sim$ 400 up to 1 GHz, as shown in figure 4(*a*), and f_r is around 1.7 GHz which causes a significant magnetic loss and a sharp decrease in μ' and increase in μ''. However, for the film deposited at $\sim 37°$ with a strong IPUMA, f_r can attain a value as high as 4.3 GHz but μ' remains at a moderate value of $\sim$150 up to 3.5 GHz, as shown in figure 4(*b*). Therefore, by a flexible adjustment of the oblique angle, the resonance frequency can be continuously manipulated, as shown in figure 4(*c*). Thus, an optimal balance between permeability and resonance frequency can be achieved, which is important for soft magnetic films in high frequency applications.

4. Conclusion

In summary, the following facts are pointed out: by doping a small amount of Zr, the static and dynamic magnetic properties of a Co thin film deposited by oblique sputtering can be significantly optimized. Such an optimization is obtained, most likely, from the fact that when the size of Co grains decreases to a value lower than their exchange length, the exchange coupling between those grains will significantly weaken the magnetocrystalline anisotropy of the individual grain. Therefore, the 'obliquely induced' in-plane anisotropy, which can be easily manipulated by controlling the oblique deposition angle, can play a dominant role in determining the static and dynamic magnetic properties of the films. Thus, the intensity of the IPUMA as well as the high frequency performance of these films can be *in situ* designed, which is fundamental for the application of soft magnetic films at different working frequencies.

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