# Nano underlayer effect on the CoFe magnetic film for GHz frequency use

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The  $Co_{35}Fe_{65}$  magnetic thin films with Cu underlayer were prepared and characterized using RF magnetron sputtering. Nano underlayer effects on the magnetic properties and crystalline structure of the film were investigated for the GHz frequency use. It was found that the underlayer was very effective in inducing elongation of lattice parameter, preferred crystalline orientation, and decrease of crystallite size. These effects were attributable to a reduction of surface energy in the underlayer and led to improve remarkably the magnetic properties for the GHz frequency use above 10 GHz.

Key words: nano underlayer, Cu, uniaxial magnetic anisotropy field, crystalline structure

### **1. INTRODUCTION**

Nano underlayer is the one candidate for optimizing crystallographic texture and surface roughness for soft magnetic thin film materials [1]. Generally, soft magnetic properties for the high frequency use mean the combination of low coercivity  $H_c$  and large in-plain uniaxial magnetic anisotropy field  $H_{k}$  which are essential conditions for coherent spin rotation (precessional rotation) under the high frequency magnetic field [2]. Both the  $H_c$  and  $H_k$  are closely related to the structural and compositional characteristics, such as crystallite size, crystallographic texture, through magnetocrystalline anisotropy and magnetoelastic anisotropy [1].

The CoFe magnetic thin film with high saturation magnetization and large  $H_k$  has attracted great interest from applied view points for the GHz frequency use, because the high saturation magnetization can bring the higher permeability and the higher ferromagnetic resonance (FMR) frequency [3]. With respect to the nano underlayer effect on  $H_c$  of the CoFe film, it is well known that  $H_c$  can be remarkably reduced due to decrease of crystallite size and surface roughness[1][4]. On the other hand, as for the nano underlayer effect on  $H_k$ , there is still not well known mechanism for explaining.

In this work, the  $Co_{35}Fe_{65}$  magnetic thin films with Cu underlayer were prepared and characterized using RF magnetron sputtering. Nano underlayer effects on the magnetic properties ( $H_k$  and  $H_c$ ) and crystalline structure of the film were investigated.

#### 2. EXPERIMENTAL

 $Co_{35}Fe_{65}$  single layers and  $Co_{35}Fe_{65}/Cu$  double layers were prepared on glass substrate (Matsunami #7050, 1×45×45mm) using RF-magnetron sputtering method. All of specimen films were at argon pressures of  $3\times10^{-3}$ Torr. The thickness of the  $Co_{35}Fe_{65}$  layer was 5000 Å fixed in all films. The thickness of the underlayer of Cu was varied from 0 to 200 Å. Fig.1 shows a schematic top view of the sputtering apparatus. The sputtering targets for depositing the CoFe film were Fe and Co with 4 in.diam. and 5mm thick. In this sputtering apparatus [2], the glass substrate was mounted on the side of the cylindrical electrode and was water-cooled.



Fig.1 Schematic top view of the sputtering apparatus.

The cylindrical electrode was rotated about its central axis at 140 r.p.m. While target electrodes were placed opposite to the side of the cylindrical electrode. Then, sputtered atoms from each target were deposited and mixed on the substrates by rotating the cylindrical electrode. Atomic composition in the CoFe film was adjusted by changing input RF -power to each target. For depositing the underlayer, Cu target with the same size as the Co and the Fe target was used.

The values of  $H_c$ ,  $H_b$ , and  $4\pi M$ , in as-deposited state were measured using a vibrating sample magnetometer (VSM). The structural characterization was carried out by an X-ray diffraction (XRD), using Cu-K $\alpha$  radiation, the  $\theta$ -2 $\theta$  and the 2 $\theta$  scanning technique. The lattice parameter was measured by multiple crystal method in XRD [3]. Surface roughness of the single and double layer was measured by 3 dimensional scanning electron microscope (3DSEM).

#### 3. RESULTS AND DISCUSSION

The  $4\pi M_r$  of Co<sub>35</sub>Fe<sub>65</sub> films prepared in this study was 23 kG Fig.2 shows the *M*-H loops of CoFe single layer (a), and CoFe/Cu double layer (b) with the



Fig.2 M-H loop along easy and hard axes of magnetization for (a) CoFe single layer, and (b) CoFe/Cu double layer with the underlayer thickness of 50 Å.

underlayer thickness of 50 Å.  $H_k$  is defined as the DC magnetic field which the M-H loop in the direction of hard magnetic axis of the film saturates, and means the necessary applied DC magnetic field which rotates the magnetic spin by 90 deg., from the easy magnetic axis direction to the hard magnetic axis direction in the film plane. The M-H loops indicated the effects of introduction of an Cu underlayer. The  $H_k$  in the double layer increased drastically. A very large  $H_k$  of above 1000 Oe was observed in the CoFe/Cu double layer. This value was about 3 times larger than that of the Co-Fe single layer. The underlayer was effective in the appearance of in-plane uniaxial magnetic anisotropy. Furthermore, in the same way, the introduction of the underlayer was effective in the reduction of  $H_c$ . The coercivity  $H_{ch}$  with the M-H loop measured in the direction of hard magnetic axis decreased from 56 Oe for the no underlayer to 11 Oe for the Cu underlayer. The coercivity  $H_{ce}$  with the *M*-H loop measured in the direction of easy magnetic axis also decreased from 99 Oe for the no underlayer to 41Oe for the Cu underlayer. The introduction of the underlayer exhibited very large uniaxial anisotropy field along with excellent soft magnetic properties. This anisotropy field was 2-50 times larger than reported values of CoFe and CoFeB thin films [1-2][4-5]. It is expected that the film shows a relative permeability of about 20 in the frequency (above 10 GHz) corresponding to the ferromagnetic resonance frequency.

Fig.3 shows the change of  $H_c$  and  $H_k$  of the CoFe single layer and the CoFe/Cu double layer as a function of the Cu underlayer thickness d. A 0 Å thick underlayer (d = 0 Å) means 5000 Å thick FeCo single layer. Both the  $H_c$  and  $H_k$  for these layers exhibited strong dependence on d in the range from d = 0 to d = 50



Fig.3 Dependence of the Hc and Hk of the CoFe single layer and CoFe/Cu double layer on the Cu underlayer thickness d.

Å. With increasing d,  $H_k$  increased totally with taking a local minimum at d = 5 - 15 and a local maximum at d =20-30 Å. It was worth while noting that the large  $H_k$ above 1000 Oe was observed in the range below d = 50Å, as shown in Fig.3. In the range over d = 50 Å,  $H_k$ decreased gradually. The observed uniaxial magnetic anisotropy was not induced by magnetic field and was associated with deposition geometry of the sputtering apparatus, as reported in previous paper [2]. In other words, the hard magnetic axis in this uniaxial anisotropy was always in the film plane and parallel to the rotational direction of the cylindrical electrode, independently by applied magnetic field.

Meanwhile, the  $H_c$  ( $H_{ch}$  and  $H_{ce}$ ) changed corresponding with the change of  $H_k$ , namely, the increase and decrease of  $H_c$  were along with those of  $H_k$ . This fact suggests that the  $H_c$  is mostly caused by the coherent spin rotation under the uniaxial anisotropy field [6].



CoFe/Cu double layer by the  $\theta - 2\theta$  scanning.

Fig.4 shows the XRD diagram of the CoFe single layer and CoFe/Cu double layer, measured by the  $\theta$ -2 $\theta$ scanning. For this scan, the only peak corresponding to the crystal plane parallel to the film plane is to be observed. The XRD diagram of the CoFe/Cu double layer exhibited only peaks of (110), (220), which indicated that these crystalline planes are oriented to the film plane. On the contrary, the XRD diagram of Co-Fe single layer for d = 0 Å exhibited very low intensity of peaks of (110), (200), (220), and (311). In this film, the structural orientation was not observed. In the previous paper [7], we confirmed that the single layer film deposited on glass substrate exhibited the non-oriented crystal structure. The underlayer was effective in the appearance of the crystal face orientation. It is well known of bcc  $\alpha$ -Fe thin films that the same orientation was caused by reducing surface free energy in the (110) plane, where the atoms under the bcc ( body centered cubic) structure were most close-packed. This reduction of surface energy was identified with that of the underlayer, because the surface roughness was quite different between the CoFe single layer and CoFe/Cu double layer, as shown in the 3DSEM surface roughness profile of Fig.5 (a) and (b). The surface roughness for the CoFe/Cu double layer in the Fig.5(b) was very low of  $\pm 2$  Å compared with that of  $\pm 6$  Å for the CoFe single layer in the Fig.5(a).



(b) CoFe/Cu double layer

Fig.5 3DSEM surface roughness profile of the CoFe single layer (a), and the CoFe/Cu double layer (b).

Fig.6 shows the XRD diagram of the CoFe single layer and CoFe/Cu double layer, measured by the  $2\theta$  scanning with  $\theta$  fixed on 1 deg. In this XRD, the projection of the incident and diffracted X-ray on the film plane was along the easy magnetic axis (a) and the hard magnetic axis (b), respectively. In both figures, (110) peak was observed. It suggested that the (110) crystalline plane orientation observed in Fig.4 was not so strict and had



Fig.6 XRD diagram of CoFe single layer, and CoFe/Cu double layer by the  $2\theta$  scanning along with the easy magnetic axis (a) and the hard magnetic axis (b).

some angle distribution to the film plane. As shown in Fig.6 (a), the XRD diagram of CoFe/Cu double layer for d = 5-200 Å exhibited peaks with the disappearance of (200) peak, while the XRD diagram of the Co-Fe single layer exhibited the (200) peak. The disappearance of (200) peak indicated that the incident X ray was not diffracted by the (200) plane, because this crystalline plane was oriented preferentially perpendicular to the film plane, and also preferentially perpendicular to the direction of the incident X-ray along with the easy magnetic axis. On the other hand, as shown in Fig.6 (b), the (200) peak appeared remarkably together the (311) peak. Furthermore, the (110) and (220) peak intensity decreased by more than 60 % compared with that in Fig.6 (a). It means that (110) and (220) plane are oriented more preferentially, and that the (200) and (311) plane was oriented less preferentially, in the film plane and in the direction of the incident X-ray along with the hard magnetic axis.

The introduction of the underlayer was effective in the crystalline plane orientation, however this orientation did not change remarkably depending on d.

Fig.7 shows the dependence of the CoFe lattice parameters for the CoFe single layer and the CoFe/Cu double layer on the d. The original bcc structure has been already changed to the tetragonal like structure (body centered tetragonal structure: bct) at d = 0, in other words, only the lattice parameter in the c direction increased by about 0.8 % of the changeless lattice parameters in each direction of a and b, and then exhibited a maximum of about 1.0 % of the changeless lattice parameters in the vicinity of d = 20-30 Å. This elongation should be caused by the effect of the Cu



Fig.7 Dependence of CoFe lattice parameter of the CoFe single layer, the CoFe/Cu double layer on the underlayer thickness of d.

underlayer The maximum of elongation of the lattice parameter corresponded to the minimum of  $H_c$  and the maximum of  $H_k$  in Fig.3. The change of the lattice parameter (equivalent to the crystalline plane space) is strongly associated with  $H_c$  and  $H_k$  through magnetoelastic anisotropy and crystalline anisotropy [1]. If this elongation causes internal strain in the direction parallel with the easy magnetic axis, the very large  $H_k$  in the vicinity of 1000 Oe is possible to be explained by magnetoelastic anisotropy.

It is well known that  $H_c$  of polycrystalline magnetic thin film is decreased steeply with decreasing crystallite size D below 1000 Å, because of spin exchange interaction between the crystallites becoming dominant[8].



Fig.8 Dependence of the D of CoFe single layer and the CoFe/Cu double layer on the underlayer thickness of d.

Fig.8 shows the dependence of crystallite size D of the CoFe single layer and the CoFe/Cu double layer on the d, where the D was estimated from the half width of CoFe (110) peaks in the XRD diagram by using the Scherrer equation [3]. The D of the CoFe/Cu double layers exhibited a drastic decrease of 1500 to several hundred Å and a minimum in the range of d = 5 to d =40 Å. In this range,  $H_c$  shown in Fig.3 also exhibited a minimum corresponding to the minimum of the D. It should be noted that the underlayer was effective in decreasing  $H_c$  because of crystallites size decreasing to less than several hundred Å [8]. The D of Co-Fe/Cu double layer was 200-300 Å with being almost independent of d. Furthermore, taking the results of Fig.4 and Fig.5 into consideration, this underlayer effect is thought to be caused by the change of surface energy in the underlayer, which affects on nuclear crystal growth for the CoFe crystallite.

We suppose a crystalline model which can explain the large  $H_k$  of above 1000 Oe and the low coercivity  $H_{ch}$  of less than 10 Oe, taking above-mentioned results into consideration. In this model, a bct CoFe unit lattice is drawn in the film plane, where the (110) plane of the unit lattice is preferentially parallel to the film plane and the elongated lattice in the c direction is oriented preferentially to the easy magnetic axis. The observed elongation is about 10<sup>3</sup> times larger than that of the magnetostriction of CoFe crystal phase [9]. Namely, it has possibility that the elongation induces the spontaneous strain in the unit lattice and the large  $H_k$  by the medium of magnetoelastic effect. For more quantitatively detailed discussion, more work is needed to clarify the angle distribution of the crystalline plane orientation and nano crystallographic texture associated with the underlayer.

## 4. CONCLUSION

The nano underlayer was effective in inducing the elongation of lattice parameter, the preferred crystalline orientation, and the decrease of crystallite size. The elongation of lattice parameter was about 1%, which was brought by transforming the bcc CoFe to the bct CoFe. The preferred crystalline orientations were found with (110) plane parallel to the film plane and with (200) plane perpendicular to the film plane. The crystallite size was decreased from 1500 Å ( for no underlayer) to several hundred Å. This effect was thought to be caused by the reduction of surface energy in the underlayer. As a result, the introduction of the underlayer led to improve the magnetic properties of the Co35Fe65 thin film for the GHz frequency use, in association with increasing the uniaxial magnetic anisotropy field above 1000 Oe and decreasing the coercivity to about 10 Oe.

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