### International Journal of Thin Film Science and Technology

| Volume 11<br>Issue 3 <i>Sep. 2022</i> | Article 6 |
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2022

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#### **Recommended Citation**

Gheisari, Kh. and K. Ong, C. (2022) "Magnetization Behaviour of Nanocrystalline Permalloy Thin Films Prepared Using Oblique-angle Magnetron Sputtering Technique," *International Journal of Thin Film Science and Technology*: Vol. 11 : Iss. 3 , PP -. Available at: https://digitalcommons.aaru.edu.jo/ijtfst/vol11/iss3/6

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International Journal of Thin Films Science and Technology

http://dx.doi.org/10.18576/ijtfst/110306

## Magnetization Behaviour of Nanocrystalline Permalloy Thin Films Prepared Using Oblique-angle Magnetron Sputtering Technique

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Received: 2 May 2022, Revised: 12 Jul. 2022, Accepted: 30 Jul. 2022. Published online: 1 Sep. 2022.

**Abstract:** In the current work, nanocrystalline Fe<sub>0.5</sub>Ni<sub>0.5</sub> magnetic thin films were deposited on a Si(100) substrate using the oblique-angle sputtering technique with the oblique deposition angle ranging from 11.5 to 45°. Structure, static magnetic properties, and dynamic magnetic characteristics were evaluated as a function of the deposition angle. The results indicate that the nanocrystalline FCC phase of FeNi with (111) preferred orientation and the average crystallite size of 6.3-9.3 nm was deposited successfully. The measured value of the uniaxial anisotropy field shows an increment from 7.65 to 16.71 Oe as the oblique angle rises from 11.5 to 45°, which in turn leads to an increase in the ferromagnetic resonance frequency from 0.63 to 0.88 GHz.

Keywords: FeNi Film, Ferromagnetic resonance frequency, Oblique-angle sputtering technique, Uniaxial anisotropy field.

### **1** Introduction

The structure and properties of the thin films have attracted thorough research works in recent years owing to their excellent high-frequency characteristics which make them especially applicable for microdevices such as electromagnetic noise absorbers, magnetic recording heads, micro-transformers, micro-inductors, etc. [1, 2, 3, 4].

Magnetic FeNi thin films generally called Permalloys have become a subject of great interest due to their high permeability in a weak magnetic field [4] which makes them useable in magnetic sensing devices and magnetic recording media [5].

FeNi alloys can be classified into different types according to the Ni content. 50 Permalloy, the FeNi alloy containing 50% Ni, exhibits the highest saturation magnetization as well as the relatively low coercivity at room temperature[4, 6]. FeNi thin films have been produced via several techniques; the most important are the electrodeposition [4, 7] and sputtering methods [8, 9].

The sputtering technique has been commonly used in many industrial productions owing to some noticeable merits such as high purity, uniform film formation, high adhesion between substrate and coating, and ease to apply to a large area [10]. In addition, the properties of the produced thin films can be effectively regulated by controlling deposition parameters such as deposition angle [2, 11], composition modification [12, 13], external field applied during the deposition [14], and multilayered components induced using multiple targets [15]. Among these processing parameters, deposition angle is well-known as an effective parameter that has a high contribution to the development of a high in-plane uniaxial magnetic anisotropy in the magnetic thin film, which in turn significantly improves the resonance frequency as the most important parameter for highfrequency applications [16, 17].

The present work aims to study the effect of deposition angle on the dynamic magnetic properties of  $Fe_{0.5}Ni_{0.5}$  thin films deposited by oblique-angle magnetron sputtering technique on a Si (100) substrate at room temperature.

### 2 Material and methods

Fe<sub>0.5</sub>Ni<sub>0.5</sub> thin films were deposited via a radio-frequency (RF) magnetron sputtering on a  $10 \times 5 \times 0.5$  mm<sup>3</sup> Si (100) substrate at room temperature. During the deposition process, the sputtering power was kept at 80 W and the deposition time was set to 20 min. The base pressure of the sputtering chamber and the pressure of Ar sputtering gas



were maintained around  $10^{-7}$  and  $10^{-3}$  Torr, respectively. Films were deposited at an oblique angle configuration using a wedge-shaped holder with a deposition angle ranging from 11.5 to 45°, while a magnetic field of 200 Oe was applied parallel to the shorter axis (easy axis) of the produced films. The phase analysis was performed using Xray diffraction (XRD, Philips Analytical X-ray Diffractometer with Cu Ka radiation). The static and dynamic magnetic properties of the as-deposited film were measured using an M–H loop tracer and a vector network analyzer, respectively. More details can be found elsewhere [11].

#### **3** Results and discussion

#### 3.1. Structure



**Fig. 1.** (a) X-ray diffraction patterns and (b) lattice parameter and diffraction angle of FeNi films deposited at various oblique angles.

Fig. 1 (a) exhibits the X-ray diffraction patterns of the asdeposited FeNi films at various deposition angles. Lattice parameters and diffraction angles deduced from Fig 1(a) are displayed in Fig 1(b). As expected, the (111) characteristic reflection of the FCC  $\gamma$ -(Fe, Ni) phase with space group Fm3m appears around  $2\theta = 44^{\circ}$  in all obliquely sputtered FeNi films. No individual peaks of BCC Fe are detected, confirming the absence of segregation of Fe. Furthermore, no other diffraction lines of FCC  $\gamma$ -(Fe, Ni) phase are observed in the wide angular range diffraction pattern, showing a predominant orientation of (111) planes has been successfully developed during the sputtering process. Concerning the literature, a crystalline anisotropy has been developed during the growth process of the film, as a result, the magnetic behaviour becomes dependent on the geometrical configuration of the film [18]. In addition, the (111) characteristic reflection shifts towards higher diffraction angles with increasing deposition angle, indicating a gradual decrease in the interplanar spacing and lattice parameter. The lower values of the lattice spacing at the higher oblique angles are attributed to the columnar grains formed at the oblique deposition condition. In fact, at

the normal incident deposition angle, no recognizable columnar grains can be detected. On contrary, tilted columnar grain morphology can be formed at the oblique-angle deposition in such a way that the tilting of the columnar grains rises with rising the oblique angle [19]. As a result, a lattice distortion is induced which is accompanied by a reduction in the interplanar spacing reduction [20]. Although, the angular shift observed in the (111) planes may be ascribed to the internal stress that may be developed during the growth process [21].

Fig. 1 (a) also indicates a noticeable diffraction line broadening due to the small crystallite size. The average size of the FeNi crystallites estimated using Scherer's equation lies in the range of 6.3-9.3 nm and shows a slight increase with the increase of the deposition angle. This finding is approximately in agreement with the results obtained by Quir et al (21.5-28.3 nm) [7] and Savin et al. (20 nm) [22] for FeNi thin film fabricated by the electrodeposition and sputtering techniques, respectively.

3.2. Magnetic properties



**Fig. 2**. M/M<sub>s</sub>-H loops along easy and hard axes for Fe films deposited at various oblique angles.



Fig. 3. Variation of easy axis coercivity  $H_{ce}$ , hard axis coercivity  $H_{ch}$  and uniaxial anisotropy field  $H_{ua}$  with oblique angle  $\beta$ .



**Fig. 4.** Room temperature permeability spectra of Fe thin films deposited at different deposition angles.



Fig. 5. Ferromagnetic resonance frequency  $f_{FMR}$  as a function of incident angle  $\beta$ .

In-plane hysteresis loops obtained along with the easy (parallel to the direction of the magnetic field applied during sputtering) and hard axis at room temperature are shown in Fig. 2. The values of coercivity along the easy axis (Hce) and hard axis (Hch) extracted from hysteresis loops are demonstrated in Fig. 3. Moreover, the uniaxial anisotropy field (Hua) was estimated from the saturation field along with the hard axis [23]. As it can be seen, more or less a typical square shape has appeared for the film produced at the oblique angle lower than 45°, indicating of formation a nearly isotropic magnetic film. On contrary, a slanted hard axis M–H loop is observed for the oblique angle of 45°. This means that a noticeable in-plane uniaxial magnetic anisotropy is induced in this condition. In fact, in-plane magnetic anisotropy is originated from the columnar microstructure developed at the deposition angle of 45°. In a

more detailed description, the magnetic anisotropy raised from the magnetocrystalline anisotropy of the tilted columnar microstructure as well as the shape anisotropy of the elongated columns [19].

In addition, a broader M–H loops along with the easy and hard axis can be observed for the film deposited at  $45^{\circ}$  oblique angle, indicating that the higher values of easy and hard axis coercivities are achieved at this deposition angle. However, coercivity values lie in the range 1.80-3.25 Oe, showing excellent soft magnetism due to the nanocrystalline microstructure of as-deposited FeNi films [11]. The results indicate that the coercivity of the as-deposited FeNi thin film is lower than that of Fe (2.9-14.3 Oe [11]) and Ni (10-40 Oe [24]) thin films, approving that FeNi composition displays a better soft magnetic behavior than Fe and Ni elements. Previous findings by other researchers [2, 25] exhibited that the coercivity can be further reduced by using an underlayer such as Co, Cu, or Fe.

The permeability spectra measured at room temperature are displayed in Fig. 4. As shown, the typical behaviour of dynamic properties of soft magnetic thin films has appeared in which the real component of permeability  $\mu'$  initially increases sharply with frequency to a maximum value and then decreases suddenly to a minimum value. Likewise, the imaginary component of permeability  $\mu''$  primarily increases quite abruptly with frequency to a maximum value at about where  $\mu'$  is very close to its inflection point followed by a quick drop to a minimum value.

However, ferromagnetic resonance frequency fFMR (where a sharp peak is observed in the  $\mu^{\prime\prime}$  spectrum) is dependent on the deposition angle. An upward trend of fFMR with the deposition angle can be observed in Fig. 5. fFMR rises from 0.63 to 0.88 GHz as the oblique angle increases from 11.5 to 45°. The higher the deposition angle, the higher the resonance frequency. ferromagnetic However. in comparison to other fabricated soft magnetic thin films, FeNi thin films show little dependence on the deposition angle due to the relatively slight variation of the magnetic anisotropy with the oblique deposition angle. As well known, **f**<sub>FMR</sub> is proportional the to square root of the product of the saturation magnetization of the film  $M_s$  and the in-plane magnetic anisotropy field  $H_K$ ,

$$f_{FMR} = (\gamma / 2\pi) \sqrt{4\pi M_s H_K}$$
, where  $\gamma$  is the gyromagnetic ratio [26]. According to the Fig. 3, the magnetic anisotropy increases from 7.65 Oe at  $\beta$ =11.5° to 16.71 Oe at  $\beta$ =45°. Assuming that the saturation magnetization does not change significantly with the deposition angle, the resonance frequency should be increased 1.48 times. Similarly, the experimental value of the resonance frequency shows an increment of 1.40 times as  $\beta$  rises from 11.5 to 45°.



#### 4 Conclusion

In summary, nanocrystalline FCC single phase of FeNi magnetic thin film with the average crystallite size of 6.3-9.3 nm and (111) preferred orientation is fabricated at various oblique angles using the oblique-angle sputtering technique. The lattice parameter of the as-deposited films decreases from 3.564 to 3.542 Å as the incident angle increases from 11.5 to  $45^{\circ}$ . This trend can be ascribed to the tilted columnar grain morphology developed at the high deposition angle. Accordingly, the uniaxial anisotropy field rises gradually from 7.65 to 16.71 Oe when the oblique angle increases from 11.5 to  $45^{\circ}$ . Consequently, the high-frequency magnetic characteristics exhibit a slight improvement in the ferromagnetic-resonance frequency from 0.63 to 0.88 GHz.

### Acknowledgments

This work was supported by the Shahid Chamran University of Ahvaz (Grant No. SCU.EM1400.222) and the National University of Singapore. The authors also would like to acknowledge Dr. Soh Wee Tee for his great assistance.

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