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Research Paper

Thermal Stability and Dynamic Magnetic Properties of NiO/Fe Multilayered Thin Films Prepared by Oblique-Angle Sputtering Technique

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ABSTRACT

In this work, the bilayer NiO/Fe thin films compared with single-layer Fe film were deposited on Si (100) substrate using the sputtering technique at deposition angles of 0° and 31.5°. Structure, the static magnetic properties, and the temperature dependence of the dynamic magnetic properties in the range from 300 K to 420 K have been investigated. The results show that the nanocrystalline BCC phase of Fe with the average crystallite size of 11-12 nm and (110) preferred orientation is formed during the deposition process. The resonance frequency is found to rise from 1.03 GHz to 1.13 GHz by employing the NiO sublayer for the typically deposited Fe film. Moreover, the resonance frequency increases for the NiO/Fe films from 1.13 GHz to 1.67 GHz as the deposition angle increases from 0° and 31.5° as a result of the increase in the magnetic anisotropy from 16 Oe to 45 Oe. The permeability values decrease for both as-deposited films with increasing temperature; however, the higher values of the permeability are observed for the film obtained at a deposition angle of 31.5°.

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1. Introduction

Nanocrystalline Fe thin films due to their high saturation magnetization can be a good candidate for use in microwave applications such as wireless communication devices, electromagnetic noise absorbers, magnetic recording media, inductive devices, etc. [1-3]. Nevertheless, the application of the as-deposited Fe thin films is rather restricted because of their poor dynamic magnetic properties; hence, several attempts have been made to improve their high-frequency magnetic properties, including using oblique deposition [4], inserting antiferromagnetic underlayer (AF) [5], composition modification [6], and so on. Oblique deposition is an effective and simple technique to modify dynamic magnetic properties [7]. During the oblique deposition process, when an external magnetic field is applied in the substrate plane parallel to the easy axis direction, a high in-plane uniaxial magnetic anisotropy is induced, leading to an increment in the ferromagnetic-resonance frequency (f_{FMR}). Therefore, the as-deposited magnetic thin film can keep the high permeability in the gigahertz frequency range, which is desirable for high-frequency applications [1, 8, 9]. Up to now, several antiferromagnet sublayers such as MnIr [5, 10] and NiO [11] have been used to grow magnetic thin films by sputtering oblique deposition techniques to achieve the desired performance for high-frequency applications. However, there are few works in the literature focusing on the improvement of high-frequency magnetic via simultaneous use of inserting AF sublayer and oblique deposition. The present work aims to prepare Fe thin film using NiO AF sublayer through the oblique-angle sputtering. Regarding the literature in this area, no attempts have been made to optimize the resonance frequency of Fe thin films by the simultaneous use of inserting NiO sublayer and oblique deposition.

2. Material and methods

The bilayer NiO (~25 nm)/Fe (~100 nm) thin films were deposited on 10 × 5 × 0.5 mm^3 Si (100) substrate at deposition angles of 0° and 31.5° using an 80 W radio-frequency magnetron sputtering. The base pressure of the sputtering chamber and the Ar working pressure were maintained around 10^{-7} Torr and 10^{-3} Torr, respectively. During deposition, a magnetic field of 200 Oe was applied in the substrate plane parallel to the easy axis direction. To compare the effect of the NiO sublayer, the regular deposited single-layer Fe thin film was prepared under similar conditions.

The phase analysis was carried out via X-ray diffraction (XRD, Philips Analytical X-ray Diffractometer with Cu Ka radiation). The static magnetic properties of the as-deposited film were measured using an M–H loop tracer. The high-frequency magnetic properties of the as-deposited film were obtained using a vector network analyzer (Agilent N5230A) in the temperature range of 330-420 K and the frequency range of 0.1 to 10 GHz.

3. Results and discussion

Fig. 1 shows the XRD patterns of the bilayer Fe thin films with the employment of the NiO sublayer at the deposition angles of 0° and 31.5°, as well as the regular deposited single-layer Fe thin film. In all cases, the isolated diffraction peak observed at around 2θ ≈44.3° (JCPDS card no. 01-085-1410) shows that the BCC phase of Fe has been growing during the deposition process. The observed diffraction line has been matched with the (110) characteristic line of Fe. On the other hand, the other characteristic diffraction lines of the BCC phase have not appeared indicating that a crystalline anisotropy (directionality dependence of properties) has been developed. It means that the magnetic easy axis lies along a particular direction [12].

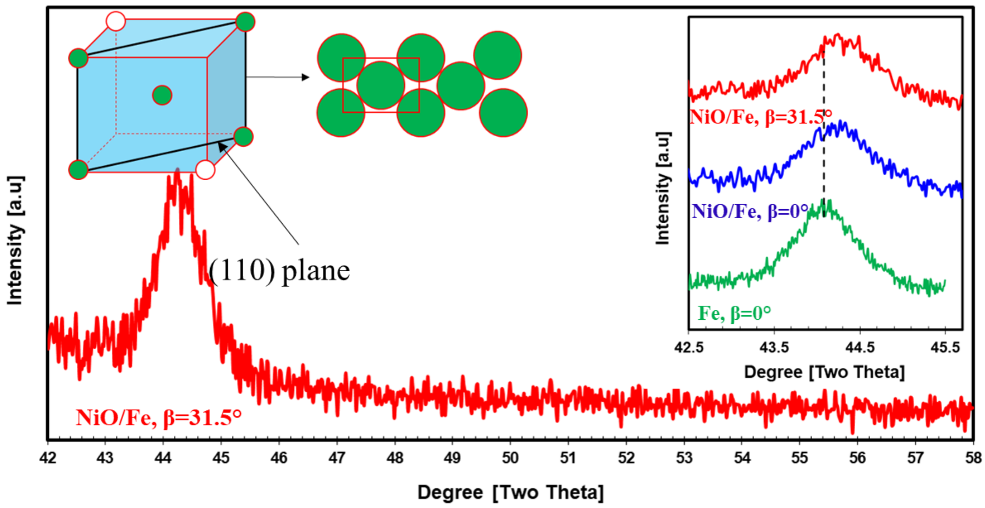

Fig. 1. X-ray diffraction patterns of the Fe film at deposited at oblique angles of 0° as well as the bilayer NiO/Fe films deposited at oblique angles of 0° and 31.5°.

As it can be seen, the characteristic diffraction peaks of the NiO sublayer cannot be observed in the bilayer NiO/Fe films, probably owing to restricted penetration of the primary X-ray beam, very thin NiO sublayer, and low degree of crystallinity in the NiO sublayer. Likewise, in some of the published papers, the characteristic diffraction lines of the underlayers were not observed in the corresponding XRD patterns. For example, Zhong et al. [1] fabricated FeCo thin film with the employment of Co underlayer using an oblique deposition. In contrast to the achievement of excellent microwave performance by Zhong et al. [1] via both oblique deposition and Co underlayer, the individual diffraction lines of the Co underlayer were not observed.

However, the direct effect of the magnetic thin film appears on the XRD pattern and magnetic properties of the obtained films. In comparison to the single-layer Fe film, a slight peak shift of (110) diffraction line towards higher diffraction angles can be observed for the normally deposited NiO/Fe film, indicating a contraction in the lattice spacing. This lattice spacing shrinking may be ascribed to the compressive residual stress developed inside the Fe film due to the employment of the sublayer [13]. The residual stress development in sputter-deposited thin films has been reported previously in magnetic film [14]. Concerning literature data, thin-film residual stresses are formed as a result of the dissimilar thermal-expansion coefficients of the substrate, underlayer, and magnetic film [13].

As reported in the literature, a gradual shift in the diffraction angle may be observed with increasing deposition angle [4]. Nonetheless, the diffraction angle is not changed significantly with the oblique angle, indicating that a shrinking or expansion of the interplanar spacing has not occurred for the bilayer NiO/Fe films when the deposition angle increases from 0° to 31.5°. Accordingly, a BCC phase with the lattice parameter of 2.889 nm in both films grows during the deposition process. Moreover, a significant (110) diffraction line broadening is observed owing to the reduction of crystallite size to the nanometer level.

Based on Scherer's equation, the average value of crystallite size lies in the range of 11-12 nm, showing a good agreement with those reported for the as-deposited soft magnetic films [15, 16]. As a result, a nanocrystalline BCC phase of Fe with the preferred orientation is obtained in both cases. Hysteresis loops of the as-deposited films along the hard axis and easy axis are displayed in Figure 2. The coercivity along hard (H_{ch}) and easy (H_{ce}) axes, as well as the uniaxial anisotropy field (H_k), were extracted from hysteresis loops and tabulated in Table 1.

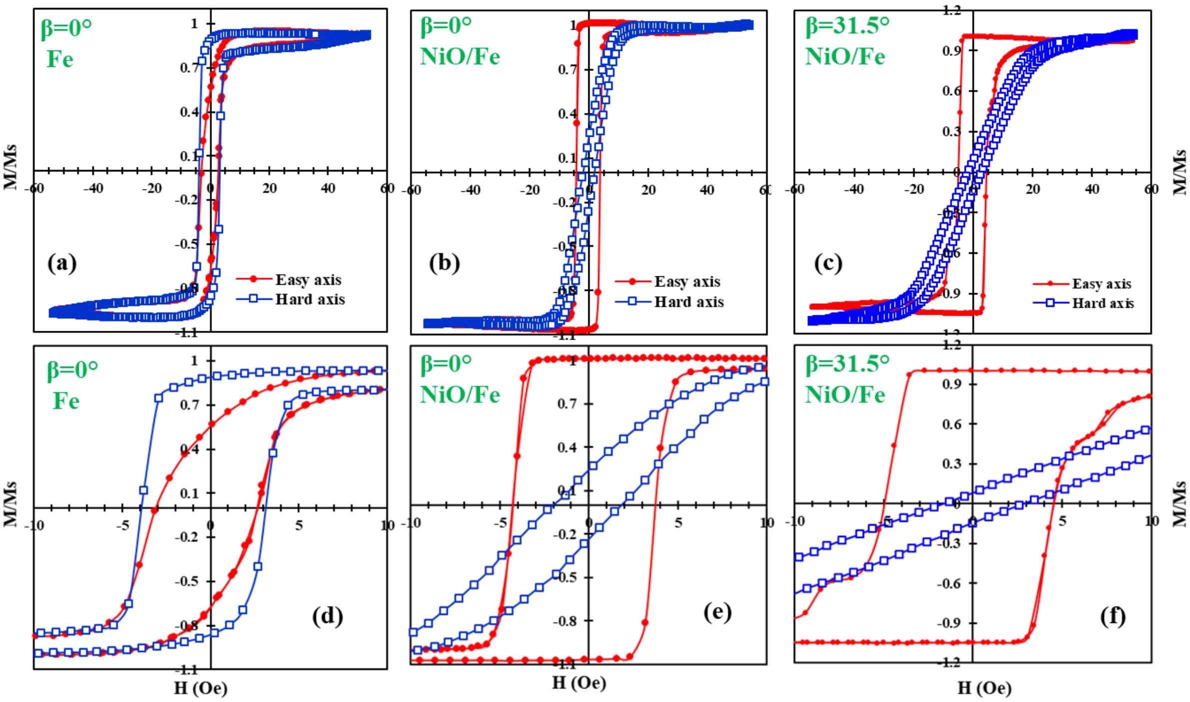

Fig. 2. (a, b, c) M/Ms–H loops along easy and hard axes of the Fe film and the bilayer NiO/Fe films deposited at various oblique angles. (d, e, f) A magnified view of M/Ms–H loops extracted from (a, b).

Table 1. Coercivity along hard axis H_{ch} and easy axis H_{ce}, anisotropy field H_k, and resonance frequency f_{FMR} for the as-deposited films.

Sample	β(°)	H_{ce}(Oe)	H_{ch}(Oe)	H_k(Oe)	f_{FMR}(GHz)
Fe	0	2.9	3.5	9	1.03
NiO/Fe	0	4.0	1.9	16	1.13
NiO/Fe	31.5	4.7	2.2	45	1.67

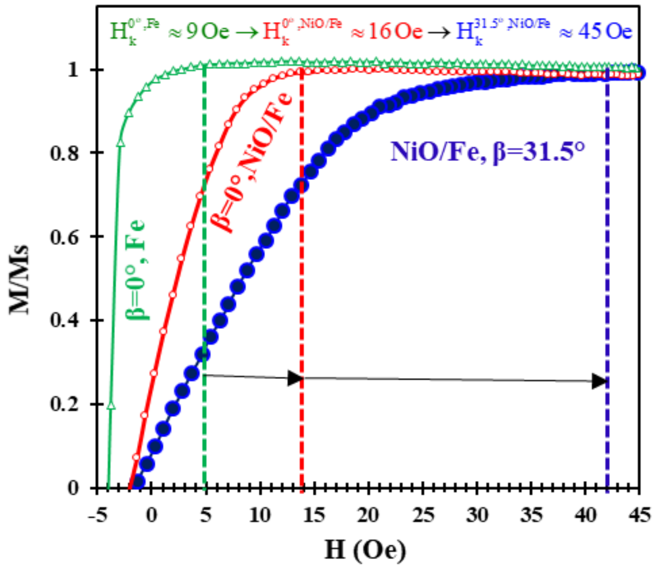

Fig. 3. Estimation of the static uniaxial anisotropy field H_k from hard axis M/Ms–H loops for the Fe film and the bilayer NiO/Fe films deposited at the various oblique angles.

As shown, in contrast to the hard M-H loop of the Fe film that exhibits a square shape, the slanted ones are observed for those obtained in NiO/Fe films, indicating that the NiO sublayer results in an in-plane anisotropy during the deposition process of Fe films on the NiO layer. It is worth noting that a square M-H loop along the hard axis was also detected for the 15 nm Fe film deposited on the Si wafer (100) by Belusky et al. [17]. However, the magnetic properties are improved by of NiO underlayer as a result of the induced in-plane anisotropy. In-plane magnetic anisotropy can be ascribed to the formation of the columnar microstructure in the oblique-angle deposition [4]. Furthermore, this figure also shows that a more slanted hard M-H loop is detected for NiO/Fe film at the deposition angle of 31.5°, representing that the anisotropy field qualitatively rises with an oblique angle.

The static uniaxial anisotropy field H_k can be measured quantitatively from the saturation field along the hard axis [18], as shown in Fig. 3. Accordingly, the anisotropy field increases from 9 Oe to 16 Oe when the Fe film normally deposited on the NiO sublayer. Further improvement is achieved for NiO/Fe films in such a way that H_k risers from 16 Oe to 45 Oe as the deposition angle increases from 0° to 31.5°. Enhancement of the H_k with the deposition angle has been reported previously [1, 2, 4, 5, 7, 10, 19].

It is worthwhile to state that in-plane magnetic anisotropy is generated owing to the formation of the columnar microstructure in such a way that the tilt angle of the columnar grains toward the deposition incidence increases as the deposition angle increases [10], increasing the anisotropy field. This magnetic anisotropy causes not only by the magnetocrystalline anisotropy of the tilted columnar grains but also by the shape anisotropy of the elongated columnar morphology [20]. However, the low values of the coercivity (1.9-4.7 Oe) exhibit good soft magnetic properties of the as-deposited film. The low values of coercivity can be attributed to the nanocrystalline structure of as-deposited films. Based on Herzer's random anisotropy model, in the nanocrystalline structure, when crystallite size *D* is smaller than the magnetic exchange length, the coercivity values are controlled by the D^6-power law. Consequently, with decreasing the crystallite size, the coercivity value decreases considerably. On the other hand, since the crystallite sizes of the as-deposited films here (11-12 nm) are lower than the magnetic exchange length of Fe (~15 nm) [21], low coercivity values are obtained. The dynamic magnetic properties containing the real and imaginary components of, permeability (μ', μ'') are displayed in Fig. 4. The typical behavior of the permeability spectra, including rapidly rising with frequency followed by a sudden drop, can be observed in all ad-deposited films. However, the ferromagnetic-resonance frequency (f_{FMR}), which shows a sharp peak, increases with deposition angle. It should be mentioned that f_{FMR} is well-known as the most important parameter of dynamic magnetic properties. After that, the magnetic permeability approaches the order of unity and restricts the applicable range of microwave devices [9].

The increase of the f_{FMR} results from the increase of H_k. Concerning Kittel's equation, the f_{FMR} is proportional to the square root of the H_k [10]; consequently, the increase of H_k positively affects the value of f_{FMR}, as shown in Fig. 4. Employment of NiO sublayer for normally deposited Fe film leads to a slight increase of f_{FMR} from 1.03 GHz to 1.13 GHz. However, simultaneous use of both oblique deposition and NiO underlayer as an appealing approach to fabricate magnetic thin films rises f_{FMR} to 1.67 GHz. Similar findings through a similar approach have been reported for FeCo thin films to obtain high resonance frequency [1].

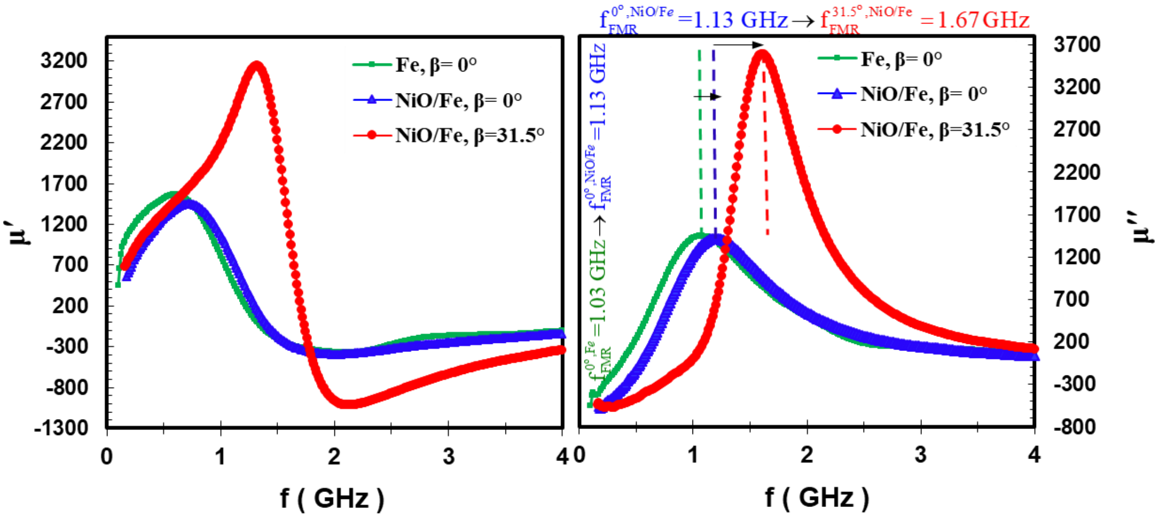

Fig. 4. Permeability spectra of the Fe film and the bilayer NiO/Fe films deposited at the various oblique angles.

The permeability spectra of the as-deposited thin films obtained at room temperature are shown as a function of the external magnetic fields (H_{ex}) in Fig. 5. As expected, the f_{FMR} shifts to a higher frequency with rising the H_{ex}. With increasing H_{ex} from 0 to 150 Oe, f_{FMR} increases from 1.13 to 5.45 GHz and from 1.67 to 5.58 GHz for the film deposited at β=0° and β=31.5°, respectively. This finding shows that a f_{FMR} indicates a directional dependent on H_{ex} which is consistent with the results found in the literature [22]. Furthermore, with increasing the H_{ex}, the initial permeability decreases, which in turn, leads to an increase in the resonance frequency. This behavior shows the permeability spectra follow well Snoek's law [4].

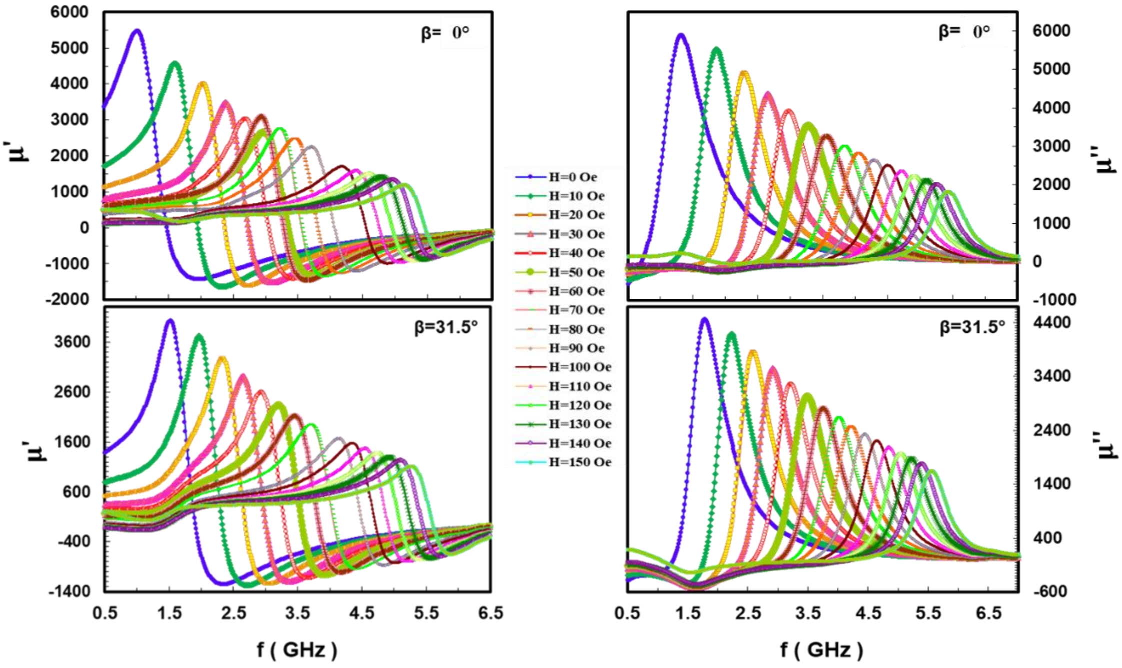

Fig. 5. Permeability spectra as a function of the external magnetic fields for the bilayer NiO/Fe films deposited at oblique angles of 0° and 31.5°.

The permeability spectra as a function of the working temperature measured in the range of 300-420 K are shown in Fig. 6. The value of both components of permeability decrease with the increase in temperature. The downward trend of the permeability with temperature results from the decrease of the saturation magnetization with temperature [23]. In fact, because of the exacerbation of thermal perturbation of magnetization at higher temperatures, the saturation magnetization decreases, and hence, the magnetic permeability starts to decrease. However, both films display good thermal stability up to 330 K. Although, the magnetic permeability destroys sharply at the higher temperature. Generally, the film deposited at oblique angles of 31.5° shows a higher magnitude of permeability even at a higher temperature.

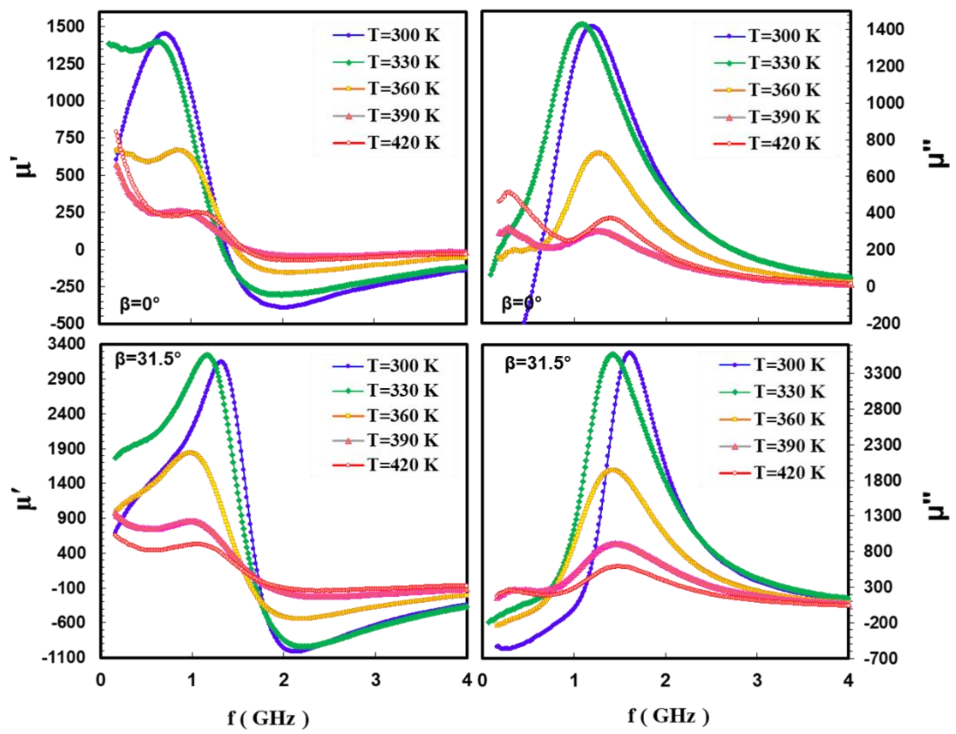

Fig. 6. Permeability spectra as a function of temperature for the bilayer NiO/Fe films deposited at oblique angles of 0° and 31.5°.

4. Conclusions

In this work, the bilayer sputtered NiO/Fe thin films were deposited on Si (100) substrate at deposition angles of 0° and 31.5°. To compare the effect of the NiO sublayer, the regular deposited single-layer Fe thin film was also fabricated under similar conditions. It is found that the NiO underlayer plays an important role in the magnetic properties of the nanocrystalline Fe thin film in such a way that the anisotropy field rises from 9 Oe to 16 Oe and the ferromagnetic resonance frequency rises from 1.03 GHz to 1.13 GHz by the employment of the NiO sublayer for the normally deposited film. Moreover, in NiO/Fe films, with an increasing deposition angle from 0° to 31.5°, the anisotropy field increases abruptly from 16 Oe to 45 Oe; consequently, the resonance frequency increases from 1.13 GHz to 1.67 GHz. Due to the decrease of saturation magnetization with temperature, the permeability values decrease with temperature; however, the film obtained at a deposition angle of 31.5° exhibits higher values of permeability at all temperatures.

Acknowledgments

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