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# Enhancing exchange bias and tailoring microwave properties of FeCo/MnIr multilayers by oblique deposition

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A systematic study of the static and dynamic magnetic properties with regards to temperature for FeCo/MnIr multilayered thin films fabricated by oblique sputtering technique was carried out. Compared with the film produced by conventional non-oblique sputtering method, those films grown by oblique deposition show a significant increment of exchange bias. The magnetic anisotropy field and the ferromagnetic resonance frequency can also be enhanced and tailored by changing oblique deposition angle. In addition, thermal stability of the dynamic characteristics of the films was presented and discussed from application-oriented perspective. © *2012 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4768280]

## I. INTRODUCTION

Exchange bias refers to a shift of the magnetic hysteresis loop when a ferromagnet (FM) is coupled with an antiferromagnet (AF).[1,2] This effect has received much attention and has been widely studied by research scientists all over the world owing to its intriguing physical origin[1–3] and extensive applications in magnetic data storage.[4,5] Another potential application of exchange bias that should not be undermined is in the field of microwave devices based on magnetic thin films as this effect can be used to push the ferromagnetic resonance frequency to a higher range.[6–13] Nevertheless, one of the drawbacks for the employment of this effect in microwave devices is its thermal stability as many applications are required to operate at high temperatures while their microwave properties are still maintained.[8,13] For example, our recent study in exchange-biased FeMn/NiFe multilayers revealed that for a better thermal stability the FM thickness should be larger but this larger FM thickness will bring about a significant reduction of ferromagnetic resonance frequency.[13] Hence, to possess an exchange-biased system that can have good thermal stability and at the same time have a high resonance frequency is a challenging issue. In this paper, we demonstrate that by incorporating exchange bias with oblique deposition technique one can attain a system satisfying both requirements of good thermal stability and high resonance frequency. In addition, through a systematic investigation of the magnetic and microwave properties of FeCo-MnIr multilayers we show that exchange bias can also be enhanced substantially by oblique deposition. Oblique deposition is a popular technique in thin film fabrication which has also been studied extensively in the literature[14–22] as an effective way to tune the magnetic anisotropy and consequently govern ferromagnetic resonance frequency.[16–22] To our best knowledge, up to now only few works in the literature have investigated exchange-biased films fabricated by oblique deposition technique.[23,24] Even in those few works, the influence of oblique deposition on exchange bias effect has not as yet been studied systematically and/or fully clarified. Hence, a detailed investigation as in the present paper on magnetic and microwave properties of exchange-biased FM/AF multilayered films with regards to the effect of oblique deposition is crucially essential for both fundamental knowledge and application-oriented research.

## II. EXPERIMENT

Samples with the structure of $Mn_{75}Ir_{25}$(15 nm)/[$Fe_{30}Co_{70}$(40 nm)/$Mn_{75}Ir_{25}$(15 nm)]$_{10}$/$SiO_2$(10 nm) were fabricated onto Si(100) substrates at ambient temperature using a radio-frequency (RF) magnetron sputter-deposition system with the base pressure of $7 \times 10^{-7}$ Torr. The first layer deposited onto the substrate is MnIr (which serves as AF layer) so that each FeCo (which serves as FM layer) has two interfaces.[12] Both of the targets used in the present study are alloy targets. A capping layer of $SiO_2$ with the thickness of 10 nm was deposited on the top of the samples to protect them from oxidation. The argon pressure was kept at $10^{-3}$ Torr during the deposition process by introducing argon gas at the flow rate of 16 SCCM (SCCM denotes cubic centimeter per minute at STP). The deposition setup is shown in Fig. 1, where the substrates were put at an oblique angle ranging from 0° to 30°. With this arrangement, the easy axis of the films induced by oblique deposition is perpendicular to the incident plane. A magnetic field of about 200 Oe was applied during the deposition process along the easy axis induced by oblique deposition in order to assist the inducement of magnetic anisotropy. The composition was determined by energy dispersive x-ray spectroscope (EDS), and the thickness of each layer was controlled both by the deposition time and by keeping the deposition rate constant, which was verified by a thickness profile meter. For the structural properties of the films, an X-ray diffractometer using CuKα radiation was employed to characterize. A vibrating sample magnetometer (VSM) was used for the measurement of magnetization

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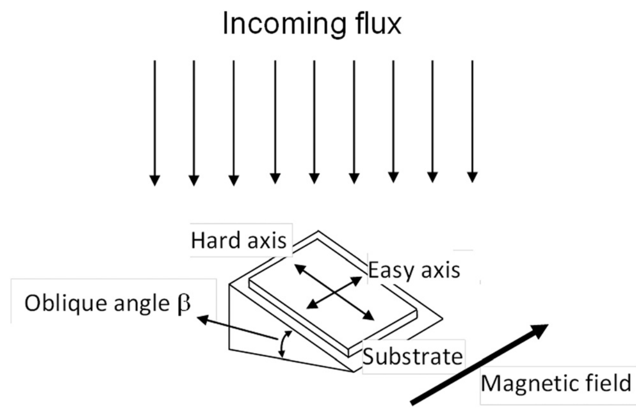


FIG. 1. Sketch of the oblique sputtered deposition system used in the present study.

curves at room temperature. In order to get permeability spectra of the films at high temperatures, we modified the setup of our previous measurement based on shorted microstrip transmission-line perturbation method[25] by attaching a heater to the fixture. The temperature is meticulously controlled by changing the voltage applied to the heater and a thermo-couple sensor attached to the fixture to monitor the temperature.[26] For the sake of the accuracy, the measurement for each sample was only carried out after the sample placed in the fixture and let the temperature stabilized for at least 10 min. The observed variation of the temperature during the measurement is less than 1 °C. By using this modified setup, we can measure permeability spectra of the films from 0.1 GHz to 10 GHz in the temperature range varied from 25 °C to 150 °C.

## III. RESULTS AND DISCUSSION

The X-ray diffraction profiles of the multilayers with different oblique angles are presented in Fig. 2. Prominent peaks of MnIr fcc (111) and (002) and FeCo bcc (110) were clearly observed for all the samples. However, as the oblique angle is increased the peaks are shifted to higher diffraction angle range indicating the contraction of the corresponding lattice spacing. Detailed summary of the change of lattice spacings with the oblique angle are presented in Fig. 3 confirming the reduction trend. This change of the microstructure of the films is possibly due to the formation of the columnar grains which are tilted due to the self-shadow effect[14–16] of the oblique deposition method. With the increasing of the oblique angle, the tilting of the columnar grains is increased and this tilting may cause some distortion of the lattice of the films in such a way that the lattice spacing is reduced.

Figure 4 shows some representative hysteresis loops of MnIr-FeCo multilayers with $\beta = 0°$, 16°, and 30° measured at room temperature along the easy axis and hard axis. The easy axis loops reveal typical exchange-biased curves with the shift of the loops from the original point which is defined as exchange bias field. It is interesting to observe the loop shift for $\beta = 0°$ is significantly less than those with $\beta = 16°$ and 30° indicating that exchange bias can be enhanced by using oblique deposition. Moreover, the hard axis loops

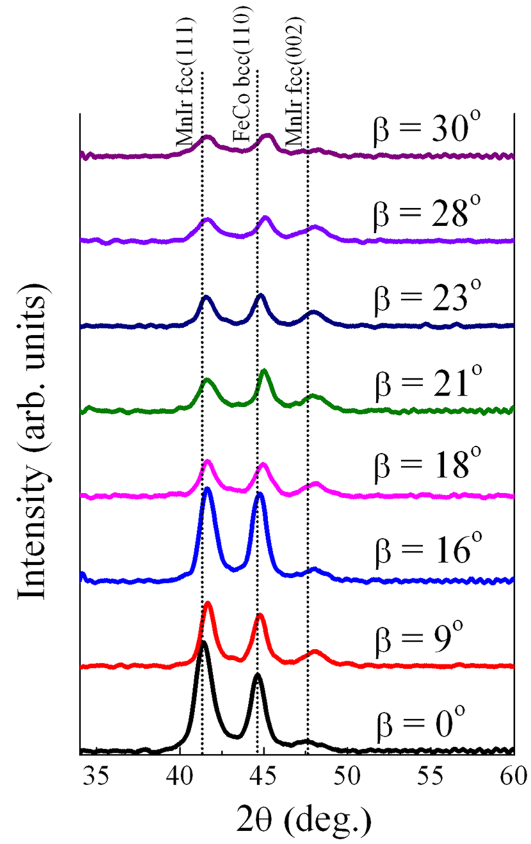


FIG. 2. X-ray diffraction profiles for FeCo-MnIr multilayers deposited at various oblique angles.

become more slanted with the increasing of oblique angle implying that the magnetic anisotropy is increased with oblique angle. It is observed in Fig. 4 that there is a two-step or even multiple-stage reversal in the magnetization curves along the easy axis. This is presumably due to the different unidirectional anisotropy constants ($J_K$) at different interfaces, which results in different exchange bias fields ($H_E$) acting on each FeCo layer.[10] Owing to the overlap of several loops with different exchange bias fields, the overall magnetization curve manifests itself as a multiple-stage reversal as in Fig. 4.[10] Similar behavior has previously been observed in

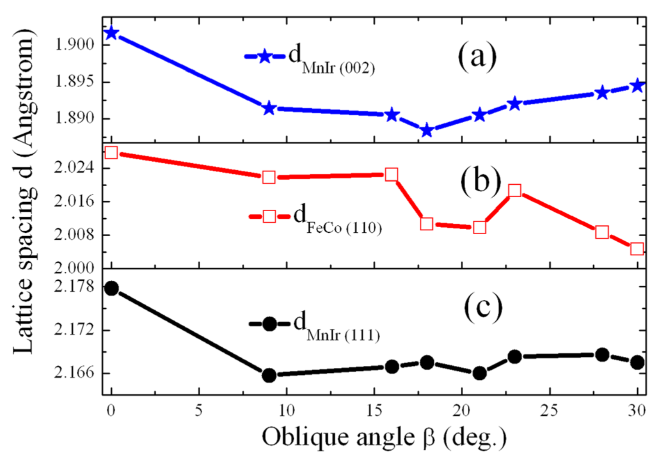


FIG. 3. Variations of lattice spacings of (a) MnIr (002), (b) FeCo (110), and (c) MnIr (111) peaks as a function of oblique angle.

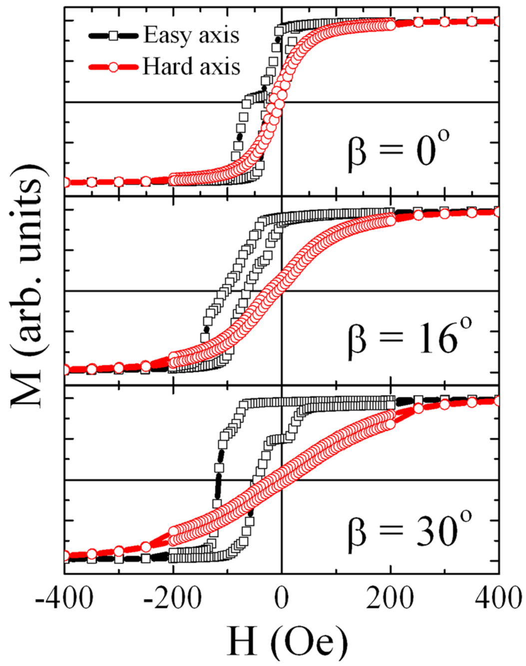


FIG. 4. Representative magnetization hysteresis loops for FeCo-MnIr multilayers deposited at various oblique angles ($\beta = 0°$, 16°, and 30°) measured at room temperature with the external magnetic field applied in easy and hard axes.

several exchange biased systems,[10,27–29] and some of the authors argued that the bottom interface normally has a better unidirectional anisotropy constant and the reason for that is that the top interface is rougher than the bottom interface.[27]

The dynamic magnetic properties of these samples as presented in Fig. 5 also show an interesting behavior. The typical permeability spectra in Fig. 5 were measured at room temperature showing that the peak of the imaginary part are shifted to higher frequency range when the oblique angle is increased. This behavior indicates that the frequency is increased with the increasing of oblique angle. For more elaborate discussion of the behaviors of both static and dynamic magnetic characteristics, we plot in Fig. 6 a summary of the oblique angle dependences of the exchange bias field $H_E$, the intrinsic uniaxial magnetic anisotropy $H_K^{int}$, the effective magnetic anisotropy field $H_K$, and the ferromagnetic resonance frequency $f_{FMR}$. The exchange bias field is determined from the loop shift of the magnetization curves along the easy axis as in Fig. 4.[6–12,30,31] The effective magnetic anisotropy field $H_K$, which is the total magnetic anisotropy determined by static magnetic measurement, can be estimated from the hard axis hysteresis loops as shown in Fig. 4.[30,31] According to McCord *et al.*,[30,31] this effective magnetic anisotropy $H_K$ is the sum of the unidirectional anisotropy field (i.e., exchange bias field $H_E$) and the intrinsic uniaxial magnetic anisotropy field $H_K^{int}$ ($H_K = H_E + H_K^{int}$).[30,31] Hence, one can determine the intrinsic uniaxial magnetic anisotropy field $H_K^{int}$ by subtraction of $H_E$ value from $H_K$ value ($H_K^{int} = H_K - H_E$). As seen in Fig. 6(a), the exchange bias field $H_E$ was found to be increased nearly doubly from 50 Oe to 95 Oe when the oblique angle is increased from 0° to 23°, and it was then slightly decreased with further increment of oblique angle from 23° to 30°. The enhancement of exchange bias with oblique angle may tentatively be interpreted in terms of the change in microstructure with oblique deposition. As discussed earlier, due to the so-called self-shadow effect, a columnar structure may form in the films grown by oblique deposition[14–22] that results in

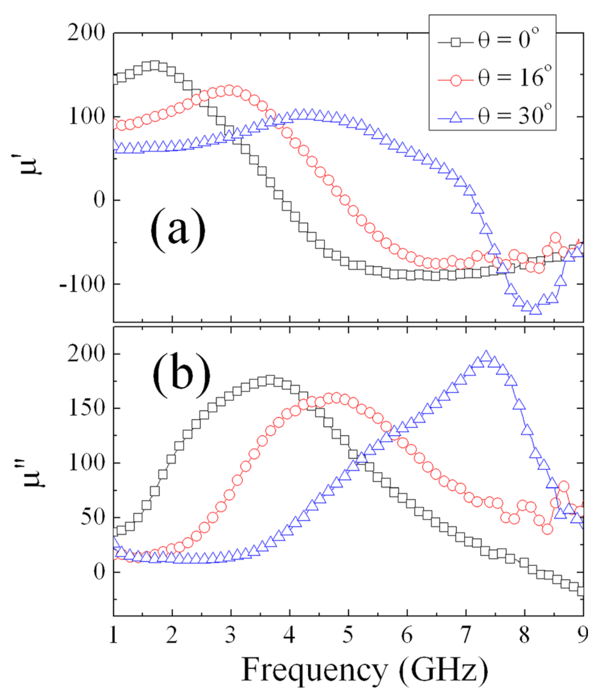


FIG. 5. Permeability spectra of FeCo-MnIr multilayers deposited at various oblique angles ($\beta = 0°$, 16°, and 30°) measure at room temperature. (a) Real part. (b) Imaginary part.

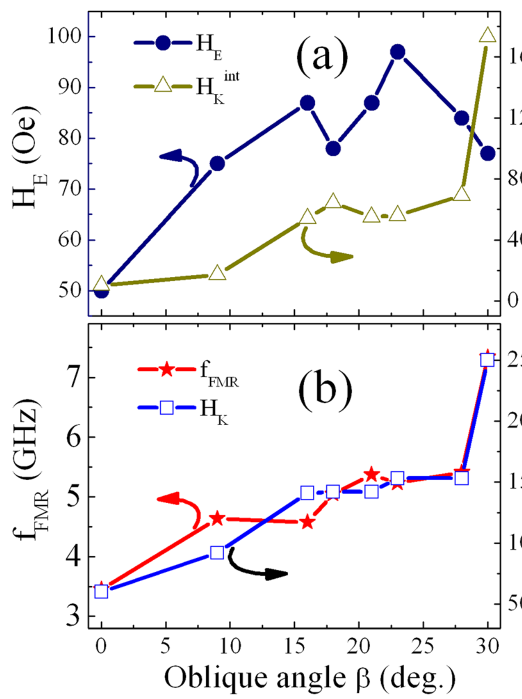


FIG. 6. Dependences of (a) exchange bias field ($H_E$) and intrinsic uniaxial magnetic anisotropy field ($H_K^{int}$), and (b) ferromagnetic resonance frequency ($f_{FMR}$) and effective magnetic anisotropy field ($H_K$) on the oblique angle.

some distortion of the lattice of the films as evident in Fig. 3. This distortion may help to create more pinning sites at the FM/AF interfaces which are important for emerge of the frozen AF spins accounting for exchange bias. This scenario may thus explain why exchange bias is significantly enhanced when the oblique angle is increased. However, since the characteristics of our FM/AF interfaces cannot be probed in a more detailed manner, this explanation is just tentative, and further study may be needed to confirm this argument. Similar to other oblique-deposited thin film systems,[14–22] our samples also show an increase of intrinsic uniaxial magnetic anisotropy field $H_K^{int}$ with the increasing of oblique angle as seen in Fig. 6(a). The variation of intrinsic uniaxial magnetic anisotropy field $H_K^{int}$ in our case is exactly the same as the case of single-layered FM films prepared by oblique sputtering. Therefore, it is increased with oblique angle as well owing to the same physical origin as reported in the literature for this kind of fabrication method[14–22] which can be interpreted in terms of the formation of the tilted columnar structure of the grains and/or the tilted elongated columns owing to the so-called self-shadow effect.[14–16] The tilted columnar structure of the grains assists the inducement of magneto-crystalline anisotropy while the elongated columns play a key role in inducing shape anisotropy.[16] Although there is a slight decrease of $H_E$ when oblique angle is increased from 23° to 30°, the increment of $H_K^{int}$ in this range is quite drastic from 56 Oe to 173 Oe making the overall magnetic anisotropy $H_K$ is monotonically increased in the whole range of oblique angle from 0° to 30° as observed in Fig. 6(b). Due to the increasing of magnetic anisotropy field with oblique angle, the ferromagnetic resonance frequency $f_{FMR}$ is also increased with oblique angle in the same manner as presented in Fig. 6(b). According to Kittel's equation,[32] the ferromagnetic resonance frequency $f_{FMR}$ is related to the effective magnetic anisotropy field $H_K$ and the saturation magnetization $M_S$ as in the following formula:

$$f_{FMR} = \frac{\gamma}{2\pi}\sqrt{H_K(H_K + 4\pi M_S)}. \tag{1}$$

Here, $\gamma$ is the gyromagnetic ratio. Note that the saturation magnetization $M_S$ is not changed with oblique angle. Hence, the variation trend of the ferromagnetic resonance frequency $f_{FMR}$ should be similar with that of the total effective magnetic anisotropy field $H_K$ as observed in Fig. 6(a).

Now we turn to the discussion of the experimental results of thermal stability of dynamic magnetization for these samples. Figure 7 presents the permeability spectra of FeCo-MnIr multilayers fabricated at oblique angle of 30° measured in the temperature range from 25 °C to 150 °C. As observed in Fig. 7(b), when the temperature is raised from 25 °C to 150 °C the peak of the imaginary part of permeability spectra is shifted towards the lower frequency range indicating the reduction of ferromagnetic resonance frequency from 7.3 GHz down to 6.2 GHz. The temperature dependences of the ferromagnetic resonance frequency for different oblique deposition angle are summarized in Fig. 8(a) showing a clear reduction of $f_{FMR}$ with temperature for all the oblique angles. The reduction of ferromagnetic resonance

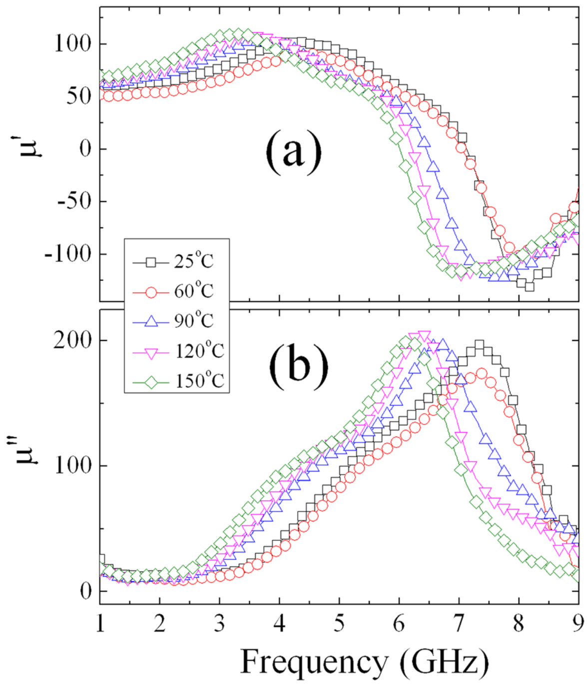


FIG. 7. Permeability spectra of FeCo-MnIr multilayers with oblique angle $\beta = 30°$ measured at different temperature from 25 °C to 150 °C. (a) Real part. (b) Imaginary part.

frequency with temperature was previously reported by several groups[8,13,26,33,34] and explained in terms of the decreasing of both the effective magnetic anisotropy field $H_K$ and the saturation magnetization $M_S$ as the temperature is increased.[8,13,26,33,34] The reduction of $M_S$ is mostly due to the thermal fluctuation of the spins when the temperature is

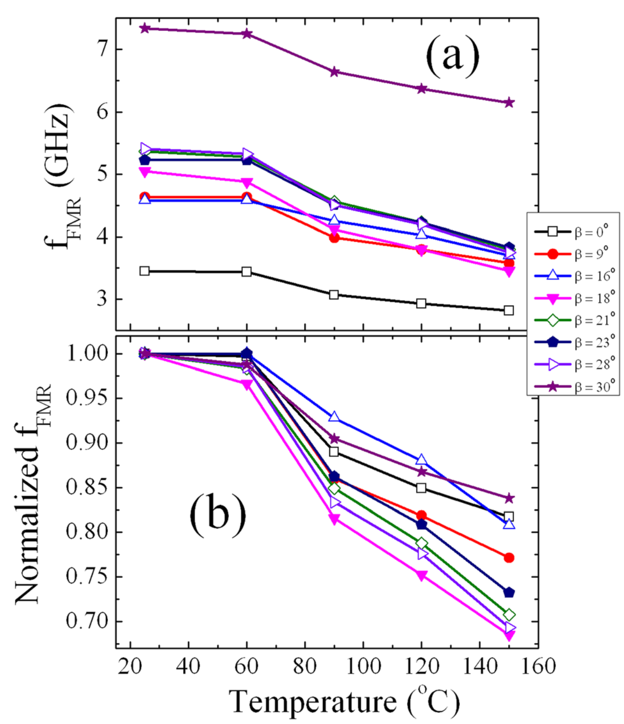


FIG. 8. Temperature dependences of (a) ferromagnetic resonance frequency and (b) normalized ferromagnetic resonance frequency for FeCo-MnIr multilayers deposited at various oblique angles.

raised. Regarding the reduction of $H_K$, as discussed above, this effective magnetic anisotropy field $H_K$ is composed of two contributions; one is the intrinsic uniaxial anisotropy field $H_K^{int}$ and the other is the unidirectional anisotropy field $H_E$. When the temperature is increased, $H_E$ is expected to decrease due to the thermal fluctuation of the AF spins at the interface which causes a substantial reduction of unidirection anisotropy.[1,8,13] The intrinsic uniaxial anisotropy $H_K^{int}$ is also expectedly decreased with temperature because when the temperature is increased, the local magnetization direction fluctuates around the mean directions, which results in the raising of energy of the easy axis and lowering the energy of the hard axis and consequently leads to the decrement of magnetic anisotropy.[34] Naturally it may be expected that the AF spins at the interface are more vulnerable to thermal fluctuation than the bulk FM spins. Hence, $H_E$ may presumably be reduced more drastically than $H_K^{int}$. However, further experimental evidence is still needed to confirm this point, especially experimental data that allow one to distinguish the thermal behavior of these two contributions. Yet such an investigation may perhaps be beyond the scope of this paper and may be left for future plan. In any way, the bottom line here is that due to the decrease of these two contributions ($H_E$ and $H_K^{int}$) with temperature, the total effective anisotropy field $H_K$ is resultantly expected to be reduced when the temperature is raised.

In order to evaluate the thermal stability of the films with various oblique angles, we plotted in Fig. 8(b) the normalized values of ferromagnetic resonance frequency as a function of temperature. The result shows that in general for the samples with low oblique angles the resonance frequency is more thermally stable than for the ones with higher angles. In particular, for $\beta = 0°$ the ferromagnetic resonance frequency is reduced around 17% while it falls off about 30% for $\beta = 28°$ when heating up from 25 °C up to 150 °C. However, an interesting observation is that when $\beta = 30°$ the thermal stability of the film becomes better with the reduction of only 16% when the temperature is increased from 25 °C up to 150 °C. This result is of crucial importance from application point of view as it demonstrates that employing oblique deposition method in exchange bias system may result in magnetic thin films with good thermal stability while still keeping a very high resonance frequency beyond 6 GHz.

## IV. SUMMARY AND CONCLUSION

In summary, we have investigated systematically the influence of oblique deposition on the magnetic and microwave characteristics with respect to temperature in FeCo-MnIr multilayered films. Our study revealed that oblique deposition technique can be used to enhance exchange bias with the value of the exchange bias field almost double compared to films prepared by non-oblique sputtering. This enhancement is tentatively suggested to be due to the creation of pinning sites at the FM/AF interfaces arising from the formation of tilted columnar structures often found in thin films fabricated by oblique sputtering. The intrinsic uniaxial magnetic anisotropy is also increased due to the formation of these tilted columnar structures thus leading to the total magnetic anisotropy is increased with the oblique angle as well. As a result, the ferromagnetic resonance frequency can be enhanced substantially by increasing oblique angle. Moreover, our films show good thermal stability with the ferromagnetic resonance frequency reduced by only 16% when the temperature is raised from 25 °C to 150 °C. The high performance of our FeCo-MnIr films suggests that one can incorporate exchange bias effect into oblique deposition technique to obtain thin films promisingly suitable for future requirements in both terms of thermal stability and high operation frequency.

## ACKNOWLEDGMENTS

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[1] J. Nogués, J. Sort, V. Langlais, V. Skumryev, S. Suriñach, J. S. Muñoz, and M. D. Baró, Phys. Rep. **422**, 65 (2005).
[2] F. Radu, R. Abrudan, I. Radu, D. Schmitz, and H. Zabel, Nat. Commun. **3**, 715 (2012).
[3] H. Ohldag, A. Scholl, F. Nolting, E. Arenholz, S. Maat, A. T. Young, M. Carey, and J. Stohr, Phys. Rev. Lett. **91**, 017203 (2003).
[4] B. Dieny, V. S. Speriosu, S. S. P. Parkin, B. A. Gurney, D. R. Wilhoit, and D. Mauri, Phys. Rev. B **43**, 1297 (1991).
[5] C. Chappert, A. Fert, and F. N. Van Dau, Nat. Mater. **6**, 813 (2007).
[6] B. K. Kuanr, R. E. Camley, and Z. Celinski, J. Appl. Phys. **93**, 7723 (2003).
[7] S. Queste, S. Dubourg, O. Acher, K. U. Barholz, and R. Mattheis, J. Appl. Phys. **95**, 6873 (2004).
[8] Y. Lamy, B. Viala, and I. L. Prejbeanu, IEEE Trans. Magn. **41**, 3517 (2005).
[9] D. Spenato, S. P. Pogossian, D. T. Dekadjevi, and J. Ben Youssef, J. Phys. D: Appl. Phys. **40**, 3306 (2007).
[10] N. N. Phuoc, F. Xu, and C. K. Ong, Appl. Phys. Lett. **94**, 092505 (2009).
[11] C. Jiang, D. Xue, and W. Sui, Thin Solid Films **519**, 2527 (2011).
[12] N. N. Phuoc and C. K. Ong, Physica B **406**, 3514 (2011).
[13] H. Y. Chen, N. N. Phuoc, and C. K. Ong, J. Appl. Phys. **112**, 053920 (2012).
[14] T. G. Knorr and R. W. Hoffman, Phys. Rev. **113**, 1039 (1959).
[15] R. D. McMichael, C. G. Lee, J. E. Bonevich, P. J. Chen, W. Miller, and W. F. Egelhoff, Jr., J. Appl. Phys. **88**, 3561 (2000).
[16] F. Tang, D. L. Liu, D. X. Ye, Y. P. Zhao, T. M. Lu, G. C. Wang, and A. Vijayaraghavan, J. Appl. Phys. **93**, 4194 (2003).
[17] E. Yu, J. S. Shim, I. Kim, J. Kim, S. H. Han, H. J. Kim, K. H. Kim, and M. Yamaguchi, IEEE Trans. Magn. **41**, 3259 (2005).
[18] X. L. Fan, D. S. Xue, M. Lin, Z. M. Zhang, D. W. Guo, C. J. Jiang, and J. Q. Wei, Appl. Phys. Lett. **92**, 222505 (2008).
[19] N. N. Phuoc, F. Xu, and C. K. Ong, J. Appl. Phys. **105**, 113926 (2009).
[20] Z. M. Zhang, X. L. Fan, M. Lin, D. W. Guo, G. Z. Chai, and D. S. Xue, J. Phys. D: Appl. Phys. **43**, 085002 (2010).
[21] X. Zhu, Z. Wang, Y. Zhang, L. Xi, J. Wang, and Q. Liu, J. Magn. Magn. Mater. **324**, 2899 (2012).
[22] I. Barsukov, P. Landeros, R. Meckenstock, J. Lindner, D. Spoddig, Z. A. Li, B. Krumme, H. Wend, D. L. Mills, and F. Farle, Phys. Rev. B **85**, 014420 (2012).
[23] B. Diouf, L. Gabillet, A. R. Fert, D. Hrabovsky, V. Prochazka, E. Snoeck, and J. F. Bobo, J. Magn. Magn. Mater. **265**, 204 (2003).
[24] R. L. Rodríguez-Suázez, L. H. Vilela-Leão, T. Bueno, A. B. Oliveira, J. R. L. de Almeida, P. Landeros, S. M. Rezende, and A. Azevedo, Phys. Rev. B **83**, 224418 (2011).
[25] Y. Liu, L. F. Chen, C. Y. Tan, H. J. Liu, and C. K. Ong, Rev. Sci. Instrum. **76**, 063911 (2005).
[26] L. T. Hung, N. N. Phuoc, X. C. Wang, and C. K. Ong, Rev. Sci. Instrum. **82**, 084701 (2011).
[27] Y. S. Lee, S. Hong, C. G. Kim, and C. O. Kim, J. Magn. Magn. Mater. **272–276**, e943 (2004).

[28]B. T. Bolona, M. A. Haugena, A. Abin-Fuentesa, J. Deneena, C. B. Cartera, and C. Leighton, J. Magn. Magn. Mater. **309**, 54 (2007).
[29]N. N. Phuoc, S. L. Lim, F. Xu, Y. G. Ma, and C. K. Ong, J. Appl. Phys. **104**, 093708 (2008).
[30]J. McCord, R. Mattheis, and D. Elefant, Phys. Rev. B **70**, 094420 (2004).
[31]J. McCord, R. Kaltofen, T. Gemming, R. Huhne, and L. Schultz, Phys. Rev. B **75**, 134418 (2007).
[32]C. Kittel, Phys. Rev. **71**, 270 (1947).
[33]M. Ledieu, F. Schoenstein, J.-H. Le Gallou, O. Valls, S. Queste, F. Duverger, and O. Acher, J. Appl. Phys. **93**, 7202 (2003).
[34]N. N. Phuoc, G. Chai, and C. K. Ong, J. Appl. Phys. **112**, 083925 (2012).