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Optimizing high-frequency properties of stripe domain ferrite doped CoFe thin films by means of a Ta buffer layer

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Abstract
The permeability μ and resonant frequency f_r of stripe domain ferrite doped CoFe thin films are enhanced from 14 to 22 and 6.0 GHz to 6.4 GHz, respectively, by using Ta buffer layer. The observed enhancement of both permeability and resonant frequency seems to deviate from Snoek's law which expects the value of μ decrease as the f_r increases. Compared to the case of using Permalloy ($Ni_{80}Fe_{20}$) buffer layer where μ is enhanced to 25 and f_r is reduced to 4.1 GHz, the present result suggests that the employment of Ta buffer layer may be useful in the quest for high permeability magnetic thin films at GHz frequency range.

(Some figures may appear in colour only in the online journal)

1. Introduction

Soft magnetic thin films with high permeability at GHz frequency range have been extensively utilized in many high-frequency applications such as microwave noise filters, microwave absorbers, thin-film inductors and so on [1–6]. The requirement for high resonance frequency beyond 5 GHz can be fulfilled through the usage of exchange bias coupling between ferromagnet (FM) and antiferromagnet (AF) [7], or gradient composition deposition [8, 9]. However, based on the works of Snoek [10] and Acher [11], there exists a trade-off between permeability and resonance frequency, thus posing a challenge for research scientists to realize a film with resonant frequency beyond 5 GHz, simultaneously with static permeability still high enough [12]. In this paper, we show that by employing rotatable anisotropy rather than other traditional kinds of magnetic anisotropy such as unidirectional anisotropy or uniaxial anisotropy, it is possible to obtain high permeability at high frequency [13, 14]. The rotatable anisotropy normally is found in FM–AF exchange-biased systems [15, 16] and thin films with stripe domains [17, 18].

In the previous work [19], we reported that the rotatable anisotropy can be found in FM doped ferrite thin films similar to FM–AF exchange-biased systems. We also extended our work to thicker ferrite doped CoFe thin films deposited on Si substrate with both stripe domain structures and FM–AF exchange coupling and possesses excellent thermal stability [20]. However, we note that the static permeability of the ferrite doped CoFe thin films deposited on Si substrate is around 14, which is in low side in order to meet the stringent requirements for microwave applications where high permeability is essential. It is due to the fact that the stripe domain structures lead to some out-of-plane component magnetization, which results in decreasing in-plane magnetization and hence decreases the value of μ. In this work, we found that when Ta is used as an under layer, which was often used to optimize the structure and magnetic properties of magnetic thin films, to optimize the high-frequency properties of stripe domain ferrite doped CoFe thin films, then the in-plane permeability was enhanced from 14 to 22 keeping the resonant frequency around 6 GHz with Ta buffer layers.

2. Experimental details

A radio frequency (rf) magnetron sputtering system is used to deposit different thicknesses of Ta and permalloy ($Ni_{80}Fe_{20}$)

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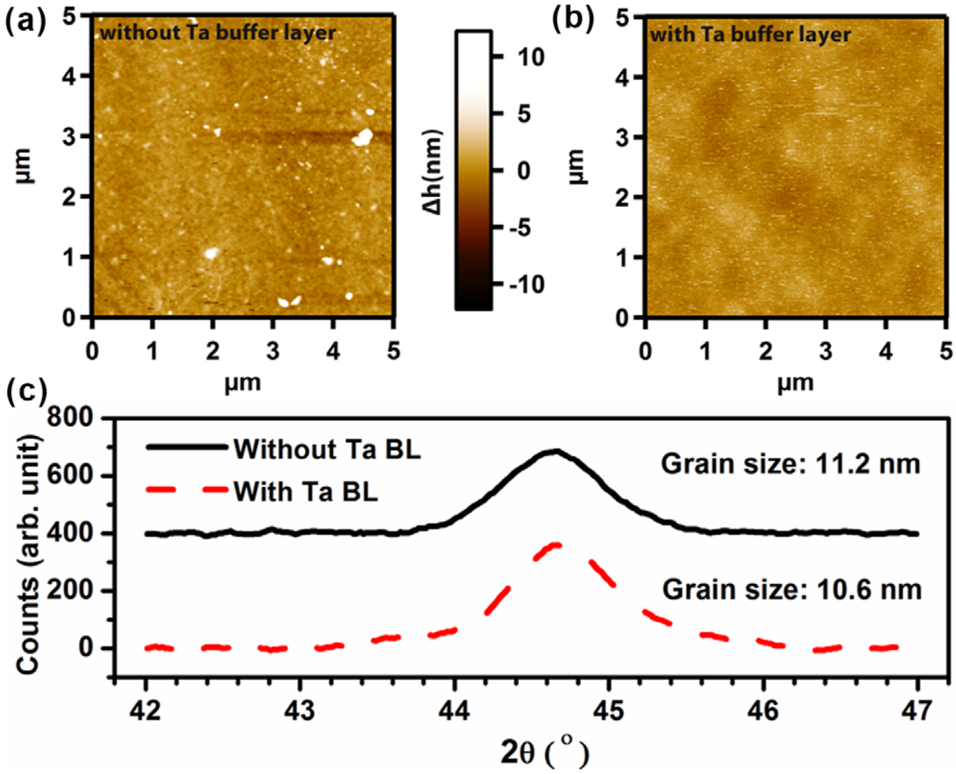

Figure 1. (*a*) and (*b*) the AFM images of ferrite doped CoFe thin films without (*a*) and with (*b*) Ta buffer layers. The colour code represents the surface roughness of the films. (*c*) The XRD profiles of ferrite doped CoFe thin films without and with Ta buffer layers; the calculated average grain sizes are also shown in the figure.

buffer layers onto 5 mm × 10 mm × 0.50 mm Si(1 0 0) substrates, then 180 nm thick ferrite doped CoFe magnetic thin films at ambient temperature are grown on it. The background pressure is lower than 5×10^{-7} Torr. The 3 inch Ta target, permalloy and $Co_{50}Fe_{50}$ targets with varying numbers of equal-sized $Ni_{0.5}Zn_{0.5}Fe_2O_4$ ferrite chips attached are used to fabricate Ta under layer, permalloy buffer layer and ferrite doped CoFe films. During sputtering, an Ar flow rate of 16 SCCM (SCCM denotes cubic centimetre per minute at STP) is needed to maintain an Ar pressure of 2×10^{-3} Torr, and the rf power density is 2.7 W cm^{-2}. The thickness of the buffer layers is controlled both by the deposition time and by keeping the deposition rate constant, which is verified by a thickness profile meter. The concentrations of the elements of the films studied in this work are Co : Fe : Ni : Zn = 46.6 : 46.0 : 3.4 : 4.0, which is determined by energy dispersive x-ray spectroscope (EDS). The crystalline structure is characterized by grazing incidence x-ray diffraction (GIXRD, Panalytical Empyrean with Cu $K\alpha$ radiation). The surface morphology and magnetic domain state of the ferrite doped CoFe magnetic thin films are characterized by atomic force microscopy (AFM)/magnetic force microscopy (MFM). The saturated magnetization M_s and M–H loops were obtained by the vibrating sample magnetometer (VSM). The static permeability was obtained in the following way. (1) Apply a saturating magnetic field in easy axis. (2) Turn to perpendicular direction, measure the small-field M–H loops. (3) The static permeability can be obtained by linear fitting the small loop. High-frequency permeability measurements were carried out with a vector network analyser at room temperature by using the shorted microstrip transmission line method from 500 MHz to 10 GHz [21].

3. Results and discussion

Figure 1 shows the AFM images of a ferrite doped CoFe thin film directly deposited on Si substrate and a ferrite doped CoFe film with Ta buffer layer with the colour code representing the roughness of the films. From the AFM images, we can estimate that the roughness of the thin film without buffer layer is around 10 nm, while the thin film with Ta buffer layer has a smoother surface with roughness less than 5 nm. Normally a magnetic thin film with smoother surface has less surface energy leading to a reduction of coercivity and thus enhancing the permeability. From the XRD results shown in figure 1(*c*), the grain sizes of thin films can be estimated on the basis of Scherrer equation [22]. Compared with the film without Ta buffer layer having an average grain size of 11.2 nm, the film with Ta buffer layer has a significantly smaller average grain size of 10.6 nm. According to random anisotropy model [23, 24], a film with smaller grain sizes may have a lower effective magnetocrystalline anisotropy, which also leads to the enhancement of the permeability of the film.

The magnetic domain structures were characterized by using the MFM technique. The results are shown in figure 2. The colour code represents Δf value which indicates the magnitude of the z-component (i.e. perpendicular component) of the magnetization in the films (a positive Δf value means

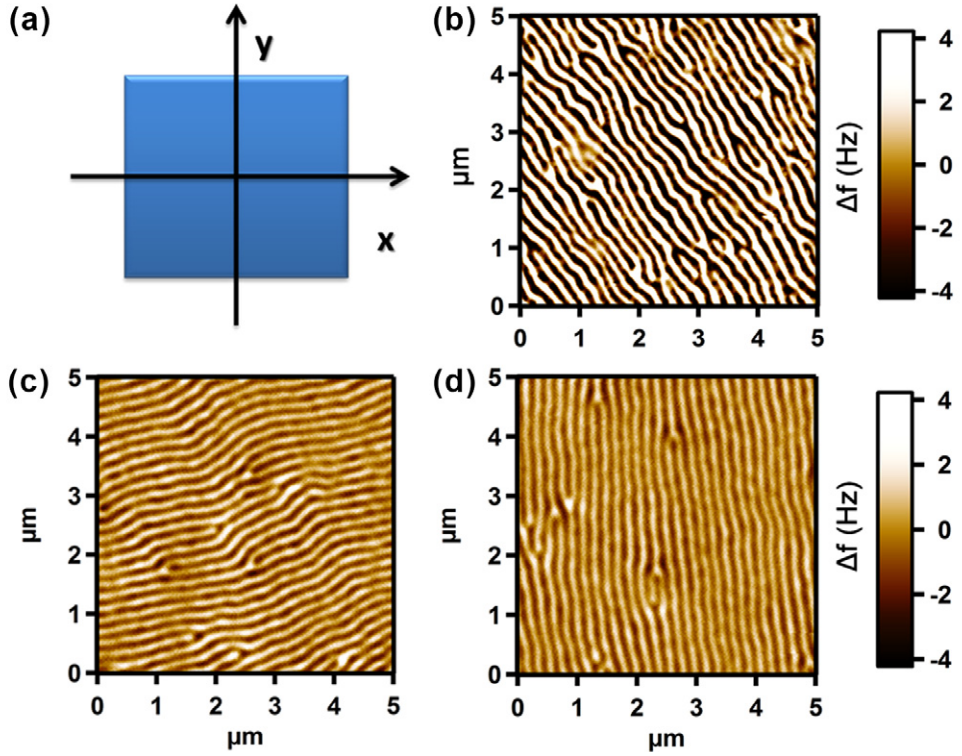

Figure 2. (*a*) Sketch map of the direction definition of the thin films. (*b*) The MFM image of ferrite doped CoFe thin films without buffer layer in as-deposited state taken in an arbitrary direction. (*c*) The MFM image of ferrite doped CoFe thin films with Ta buffer layer in as-deposited state taken in an arbitrary direction. (*d*) MFM image of ferrite doped CoFe thin films with Ta buffer layer in remanent state after the sample was placed under a field with 5 kOe along the *y*-axis.

that the magnetization is pointing upward while a negative Δf indicates that the magnetization is pointing downward). The MFM patterns are similar to the results in other soft magnetic films [18, 25]. The sketch of the magnetization configuration of the domain structure thin films can be found in figure 3. The magnetization in each population being represented in spherical coordinates by θ_1, φ_1, θ_2 and φ_2 as sketched in figure 3. The free energy function may be written as [25]

$$E = \frac{1}{2}K^{\mathrm{OP}}(\sin^2\theta_1 + \sin^2\theta_2) + \frac{1}{2}K^{\mathrm{IP}}(\sin^2\theta_1 \sin^2\varphi_1 + \sin^2\theta_2 \sin^2\varphi_2) + \frac{\pi}{2}N_{zz}M_{\mathrm{s}}^2(\cos\theta_1 - \cos\theta_2)^2 + \frac{\pi}{2}N_{yy}M_{\mathrm{s}}^2(\sin\theta_1\sin\varphi_1 - \sin\theta_2\sin\varphi_2)^2 + \frac{\pi}{2}M_{\mathrm{s}}^2(\cos\theta_1 + \cos\theta_2)^2 + A\left(\frac{2}{d}\right)^2\left(\frac{\theta_2-\theta_1}{2}\right)^2, \quad (1)$$

where K^{OP} is the out-of-plane anisotropy (including perpendicular anisotropy, surface anisotropy, etc) and K^{IP} is the in-plane anisotropy, N_{zz} and N_{yy} are demagnetization factors at z and y directions, A is the exchange constant, d is the domain width. The first and second terms of equation (1) are out- of-plane and in-plane anisotropy, the third and fourth terms are demagnetization energies, the fifth term is the static magnetic energy and the sixth term is the exchange energy. The equilibrium position is that $\theta_1 = \pi - \theta_2 = \theta$, $\varphi_1 = -\varphi_2 = 0$.

Compared with the film without Ta buffer layer (figure 2(*b*)), the films with Ta buffer layer (figure 2(*c*)) have smaller z-component of the magnetization (larger θ), implying that the films with Ta buffer layer have greater in-plane

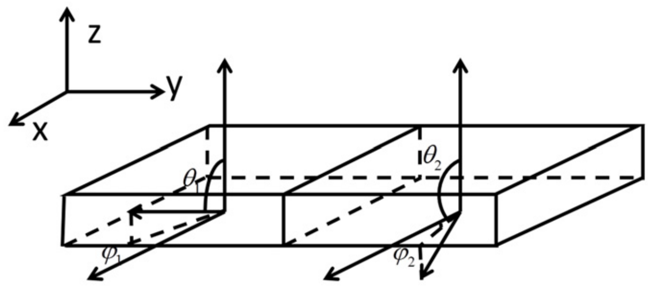

Figure 3. Sketch of the magnetization configuration in the domain structure magnetic thin films.

component of magnetization. Figure 2(*d*) shows the MFM image of ferrite doped CoFe thin films with Ta buffer layer in remanent state after the sample was placed under a field with 5 kOe along the y-axis. First, the MFM image in figure 2(*c*) was taken with an arbitrary referenced direction for the film in remanent state as in figure 2(*c*). After that, a sufficiently large magnetic field of about 5 kOe was applied in the y-axis and reduced to zero so that the sample was in remanent state along the y-axis. Then the MFM image was taken again as in figure 2(*d*) which shows that the stripe domains are now aligned along the y-axis following the direction of applied external magnetic field. This observation clearly demonstrates that the direction of stripe domains can be rotated under a 5 kOe external magnetic field [26].

Figure 4 shows the M–H loops of the ferrite doped CoFe thin films without a Ta buffer layer and with Ta buffer layers of

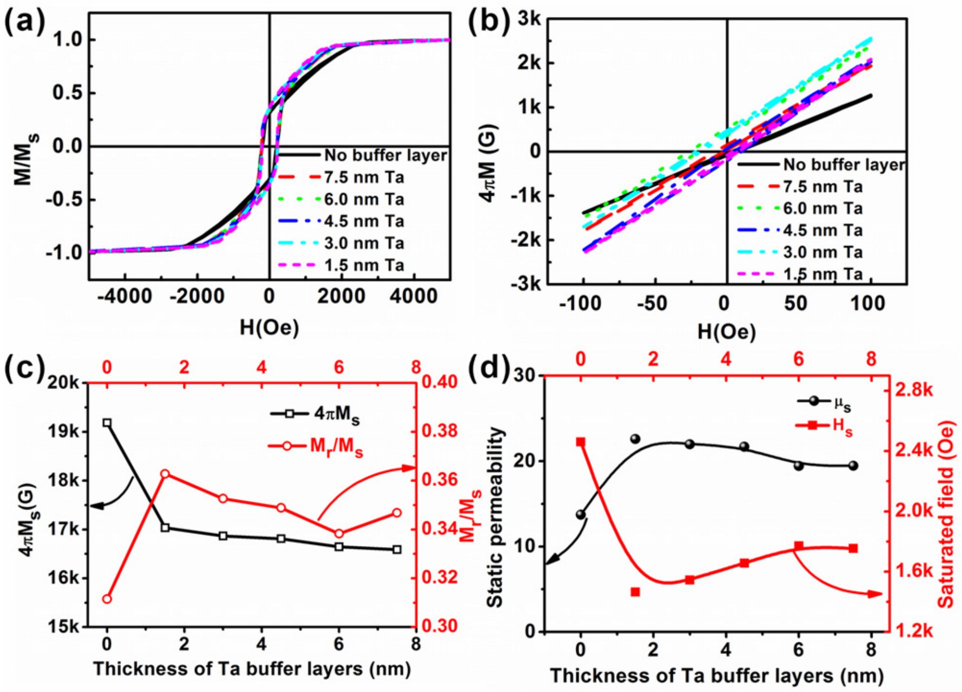

Figure 4. (*a*) M–H loops of ferrite doped CoFe thin films without and with Ta buffer layers of different thicknesses. (*b*) Small-field M–H loops measured after saturating the films at in-plane perpendicular direction. (*c*) The saturation magnetization and remanent magnetization of ferrite doped CoFe thin films without and with Ta buffer layers of various thicknesses. (*d*) The saturation fields of the samples obtained from M–H loops and the static permeability obtained by fitting the small-field M–H loops.

different thicknesses. Those loops are all superimposed curves with a linear decrease of magnetization from its saturation value and a moderate remanence [20, 26]. This is a clear signature of the formation of stripe domains with the presence of out-of-plane magnetization component, which can also be found in figure 4(*a*) [27]. When a large enough field is applied along y-axis, the domain structure disappears and all the spins will rotate to be aligned with the y-axis [27]. The film will split into new stripe domain structure with the wall parallel to y-axis when the field reduces from saturated field to 0 as shown in figure 2(*d*). The saturation magnetization $4\pi M_s$ and remanent magnetization M_r of these thin films obtained from the M–H loops are shown in figure 4(*c*). It is found that $4\pi M_s$ is decreased with Ta buffer layers. This behaviour can be understood by the formation of the smaller grain sizes in CoFe doped ferrite films when deposited with Ta buffer layers, which introduces more frozen spins at the interface of the ferromagnetic grains. Consequently, the magnetization is reduced due to the reduced ferromagnetic spins. However, the remanent magnetization is increased for thin films deposited on Ta buffer layers. This may be attributed to the smoother surface, which reduces the surface anisotropy leading to more magnetic spins lying in the film plane. The static permeability μ_s of these thin films estimated from the minor M–H loop measurement as shown above can be found in figure 4(*b*). The slopes of the linear loops represent the values of μ_s. The results show that M–H loops of thin films deposited on Ta buffer layers are steeper than that of the one without buffer layer, indicating that the static permeability is increased when using Ta buffer layer.

In soft magnetic thin films, out-of-plane anisotropy field $H_k^{OP} \ll 4\pi M_s$ and in-plane anisotropy field $H_k^{IP} \ll 4\pi M_s$, the static permeability $\mu_s \approx \mu_{yy}$ can be written as [28]

$$\mu_s \approx 1 + \frac{4\pi M_s}{H_k^{IP}} - \frac{4\pi}{N_{zz}} \frac{H_k^{OP}}{H_k^{IP}}. \tag{2}$$

The increasing of the static permeability is due to the decreasing of the out-of-plane anisotropy. The variation of permeability as a function of Ta buffer thickness is presented in figure 4(*d*). Based on the Stoner–Wohlfarth model [29], the saturation field is dependent on the effective anisotropic field (H_k^{IP} and H_k^{OP}) in stripe domain structure thin films [27]. The saturated fields H_s shown in figure 4(*d*) are decreased with increasing Ta buffer layers thickness, implying that the energy of the stripe domain in films deposited on Ta buffer layers is decreased. These results are in good agreement with the MFM images shown in figure 2 where weaker stripe domains were found in the films deposited on Ta buffer layers.

The high-frequency properties of these thin films investigated by VNA microstrip method are displayed in figure 5. Based on the LLG equation [30], the permeability spectrum of an in-plane magnetized thin film can be expressed as [18, 31, 32]

$$\mu = 1 + \frac{f_m(f_0 + f_m + i\alpha f)}{f_r^2 - f^2 + i f \Delta f_r}, \tag{3}$$

where $f_m = \gamma 4\pi M_s/2\pi$, γ is the gyromagnetic ratio, $f_0 = \gamma H_k^{dyn}/2\pi$, $H_k^{dyn} = H_k^{IP} - H_k^{OP}$, $f_r^2 = f_0^2 + f_m f_0$, and $\Delta f_r = \alpha(2f_0 + f_m)$. The resonant frequency f_r and the effective dynamic anisotropic fields H_k^{dyn} can be obtained by fitting the experimental curves with equation (3). The f_r and H_k^{dyn} as function of Ta buffer thickness are demonstrated in

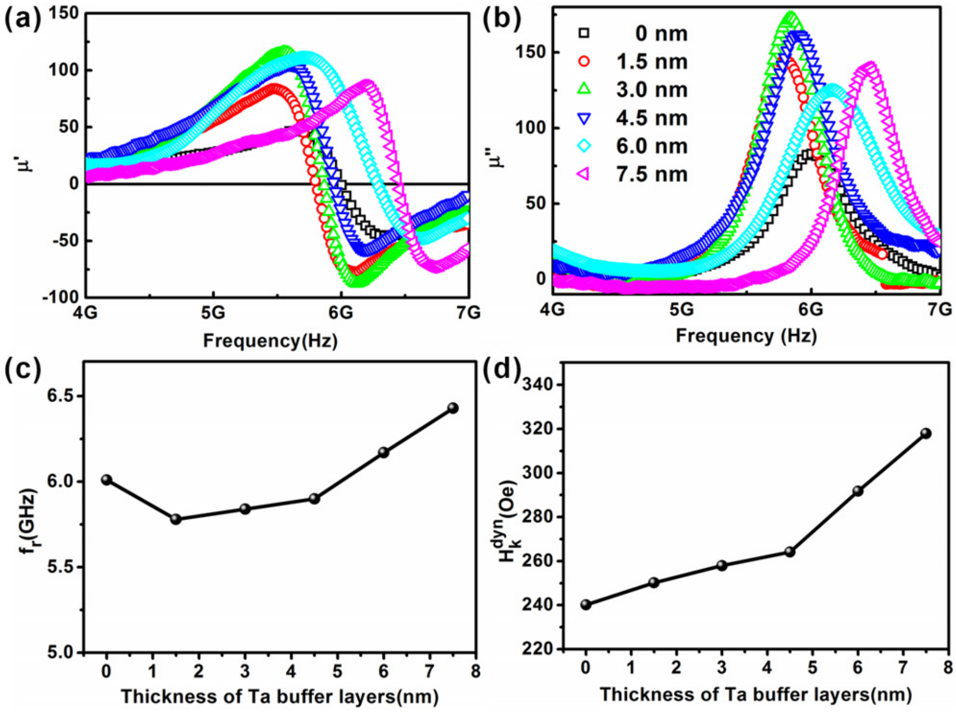

Figure 5. (*a*) Real and (*b*) imaginary parts of the high-frequency permeability spectra of ferrite doped CoFe thin films without and with Ta buffer layers of different thicknesses. (*c*) Resonant frequency and (*d*) dynamic anisotropy field as a function of the thickness of Ta buffer layers.

figures 5(*c*) and 4(*d*), respectively. The resonant frequency f_r decreases from 6.0 GHz to 5.8 GHz with Ta buffer layer thickness less than 1.5 nm and then increases to 6.4 GHz with further increasing of Ta buffer layer. However, the effective dynamic anisotropic fields H_k^{dyn} increase monotonously from 240 to 319 Oe. This behaviour is due to the fact that, according to Kittel's equation, the resonant frequency is not only related to dynamic anisotropic field but also related to the saturation magnetization of the thin films. From figure 4(*b*), we can see that the saturation magnetization of the thin film first decreases then keeps constant with the Ta buffer layer thickness increases. Hence, the decreasing of the resonant frequency as observed in figure 5(*c*) is likely due to the dramatic decrease of the saturation magnetization for buffer layer thickness less than 1.5 nm before becoming stable with further increasing of Ta buffer layer thickness.

Now we turn to the discussion of the effect of the Ta buffer layers on high-frequency properties of the ferrite doped CoFe thin films. Based on the above results, the main influence of Ta buffer layer are: (i) the permeability is increased; (ii) the saturation magnetization is reduced; (iii) the resonant frequency is increased for thicker Ta buffer layers. It is very interesting to observe that the permeability and resonant frequency are both increased as the saturated magnetization is decreasing, which are in contradiction with the Acher's law [11]. The static permeability μ_s can be obtained by equation (2), which is smaller than μ obtained in in-plane anisotropy films $\mu_s = 1 + M_s/H_k^{IP}$. As shown in figure 4(*b*), the remanent magnetization M_r is increased with the increasing of Ta buffer layer thickness which means the H_k^{OP} decreasing. As shown in figure 4(*d*), the saturation field of the thin films decreases with the increasing of Ta buffer layer thickness. Due to the decreasing of effective anisotropy, the static permeability is increased and the saturated magnetization is reduced with the Ta buffer layer thickness increasing. Based on the Kittel equation [33], the resonant frequency f_r in stripe domain thin films should be rewritten as [18]

$$f_r = \frac{\gamma}{2\pi}\sqrt{H_k^{IP}[H_k^{IP} + (4\pi M_s - H_k^{OP})]}. \tag{4}$$

The saturation magnetization $4\pi M_s$ is first decreased then keeps constant with the increasing of Ta thickness. Thus, the resonant frequency decreases with the addition of a very thin buffer layer. As discussed above, weaker stripe domain and less are also found in the films with the Ta buffer layer, indicating that the H_k^{OP} for the films is decreased. Hence, from equation (4), it can be deduced that the resonant frequency of thin films with thicker Ta buffer layer is higher than that of the films without buffer layers. Based on equations (2) and (4), Acher's limit should be rewritten as

$$(\mu - 1) f_r^2 = (4\pi M_s + H_k^{IP} - H_k^{OP})(4\pi M_s - H_k^{OP}). \tag{5}$$

So the Acher's limit will enhance in thin films with Ta buffer layers comparing with the films without buffer layers due to the decreasing of the H_k^{OP}.

As a comparison, the effects of permalloy buffer layers are also studied. The results can be found in figure 6. The resonant frequency f_r and the effective dynamic anisotropic fields H_k^{dyn} as a function of permalloy buffer thicknesses are demonstrated in figures 6(*c*) and (*d*), respectively. The resonant frequency f_r decreases from 6.0 GHz to 4.1 GHz with permalloy buffer layer thickness less than 3.0 nm and then increases to 4.7 GHz

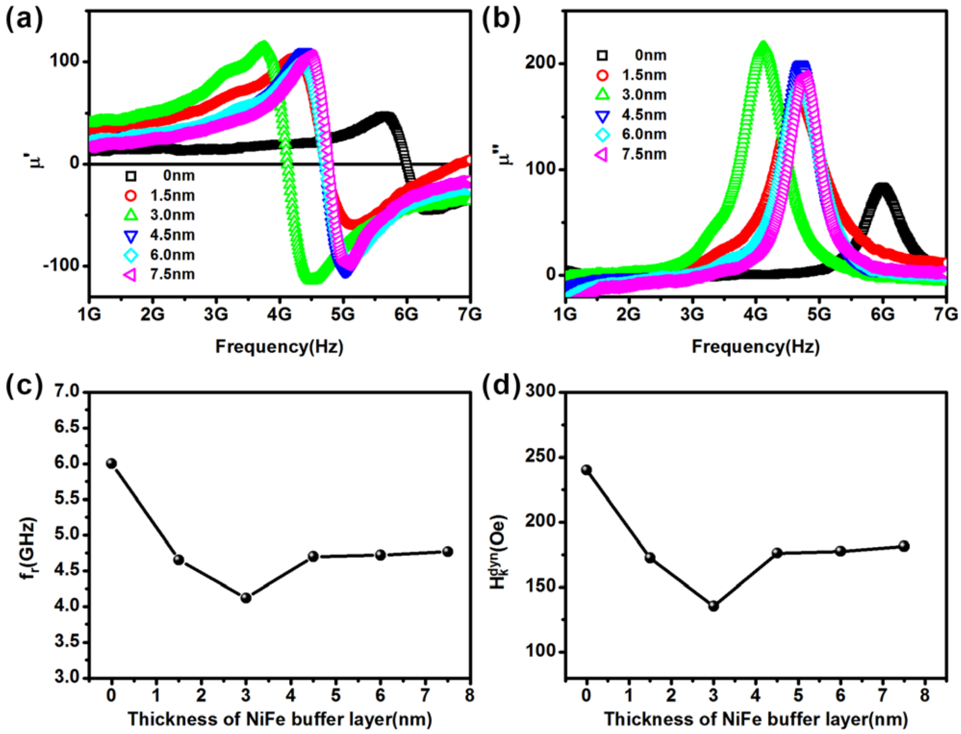

Figure 6. (*a*) Real and (*b*) imaginary parts of the high-frequency permeability spectra of ferrite doped CoFe thin films without and with permalloy buffer layers of different thicknesses. (*c*) Resonant frequency and (*d*) dynamic anisotropy field as a function of the thickness of Permalloy buffer layers.

with further increasing the thickness of permalloy buffer layer. The effective dynamic anisotropic fields H_k^{dyn} show the same behaviour first reducing from 240 to 135 Oe then increasing to 180 Oe. These behaviours are due to the very soft magnetic properties of permalloy buffer layer. Unlike Ta buffer layer, the permalloy is a soft magnetic material with M_s about 1 T and very small anisotropy less than 10 Oe. So the M_s of the whole film does not change much, but the total effective anisotropy reduces to a small level due to the exchange coupling between the permalloy buffer layers and the main ferrite doped CoFe layer. Hence, according to the Kittel equation, the resonant frequency should show a similar behaviour to the effective anisotropy field.

4. Conclusions

In summary, we have investigated the effect of Ta and Permalloy buffer layers on the static and high-frequency properties of ferrite doped CoFe thin films. For the better choice with Ta buffer layers, the static permeability of the thin film is increased due to less out-of-plane anisotropy. The resonant frequency of the thin film is also increased due to the smaller perpendicular anisotropic field with Ta buffer layers. This work proposes a novel way to optimize the high-frequency properties of magnetic thin films with stripe domain structures.

Acknowledgment

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References

[1] Li X L, Chai G Z, Guo D W, Gao B, Zhang Z M and Xue D S 2009 *Microelectron. Eng.* **86** 2290
[2] Gardner D S *et al* 2008 *J. Appl. Phys.* **103** 07E927
[3] Chai G Z, Xue D S, Fan X L, Li X L and Guo D W 2008 *Appl. Phys. Lett.* **93** 152516
[4] Chai G Z, Guo D W, Li X L, Zhu J Y, Sui W B and Xue D S 2009 *J. Phys. D: Appl. Phys.* **42** 205006
[5] Phuoc N N, Xu F and Ong C K 2009 *Appl. Phys. Lett.* **94** 092505
[6] Chai G Z, Yang Y C, Zhu J Y, Lin M, Sui W B, Guo D W, Li X L and Xue D S 2010 *Appl. Phys. Lett.* **96** 012505
[7] Phuoc N N, Chai G Z and Ong C K 2012 *J. Appl. Phys.* **112** 113908
[8] Li S D, Huang Z G, Duh J G and Yamaguchi M 2008 *Appl. Phys. Lett.* **92** 092501
[9] Phuoc N N and Ong C K 2013 *Adv. Mater.* **25** 980
[10] Snoek J L 1948 *Physica* **14** 207
[11] Acher O and Adenot A L 2000 *Phys. Rev.* B **62** 11324
[12] Ledieu M, Schoenstein F, Le Gallou J H, Valls O, Queste S, Duverger F and Acher O 2003 *J. Appl. Phys.* **93** 7202
[13] Stiles M D and McMichael R D 1999 *Phys. Rev.* B **59** 3722
[14] McCord J, Mattheis R and Elefant D 2004 *Phys. Rev.* B **70** 094420
[15] Stiles M D and McMichael R D 1999 *Phys. Rev.* B **60** 12950
[16] McCord J, Kaltofen R, Gemming T, Huhne R and Schultz L 2007 *Phys. Rev.* B **75** 134418
[17] Prosen R J, Holmen J O and Gran B E 1961 *J. Appl. Phys.* **32** 91S
[18] Youssef J B, Vukadinovic N, Billet D and Labrune M 2004 *Phys. Rev.* B **69** 174402
[19] Chai G Z, Phuoc N N and Ong C K 2012 *Sci. Rep.* **2** 832

[20] Chai G Z, Phuoc N N and Ong C K 2013 *Appl. Phys. Lett.* **103** 042412
[21] Liu Y, Chen L F, Tan C Y, Liu H J and Ong C K 2005 *Rev. Sci. Instrum.* **76** 063911
[22] Patterson A L 1939 *Phys. Rev.* **56** 978
[23] Herzer G 1990 *IEEE Trans. Magn.* **26** 1397
[24] Xue D S, Chai G Z, Li X L and Fan X L 2008 *J. Magn. Magn. Mater.* **320** 1541
[25] Acher O, Boscher C, Brulé B, Perrin G, Vukadinovic N, Suran G and Joisten H 1997 *J. Appl. Phys.* **81** 4057
[26] Wang G X, Dong C H, Wang W X, Wang Z L, Chai G Z, Jiang C J and Xue D S 2012 *J. Appl. Phys.* **112** 093907
[27] Saito N, Fujiwara H and Sugita Y 1964 *J. Phys. Soc. Japan* **19** 1116
[28] Buznikov N A and Rozanov K N 2005 *J. Magn. Magn. Mater.* **285** 314
[29] Stoner E C and Wohlfarth E P 1948 *Philos. Trans. R. Soc. Lond.* A **240** 599
[30] Gilbert T L 2004 *IEEE Trans. Magn.* **40** 3443
[31] Sun N X, Wang S X and Silva T J 2002 *IEEE Trans. Magn.* **38** 146
[32] Zhang Z M, Lin M, Zhu J Y, Guo D W, Chai G Z, Fan X L, Yang Y C and Xue D S 2010 *J. Appl. Phys.* **107** 083912
[33] Kittel C 1948 *Phys. Rev.* **73** 155