Effect of Zn substitution in (111)-textured $\text{Zn}_x\text{Fe}_{3-x}\text{O}_4$ thin films on magnetization dynamics

Honglei Yuan^a, Jian-Guo Zheng^b, Yuli Yin^a, Er Liu^a, Zhaocong Huang^a, Ya Zhai^{a,c*}, Jun Du^c, Hongru Zhai^c

^a Department of Physics and Jiangsu key laboratory of advanced metallic materials, Southeast University, Nanjing 211189, China

^b Irvine Materials Research Institute, University of California, Irvine, California 92697-2800, USA

^c National Laboratory of Solid State Microstructure, Nanjing University, Nanjing 210093, China

Abstract

The strong (111)-textured $\text{Zn}_x\text{Fe}_{3-x}\text{O}_4$ thin films were grown on SiO_2 -terminated silicon substrates using pulsed laser deposition. X-ray diffraction and high resolution transmission electron microscopy clearly show the column growth structure along the film thickness direction in [111] crystalline orientation for every grain column. With the introduction of texture induced magnetic anisotropy in the free energy density model, we quantitatively determined the doping effect on the various anisotropies in (111)-textured $\text{Zn}_x\text{Fe}_{3-x}\text{O}_4$ films. The mechanisms of ferromagnetic resonance linewidth of $\text{Zn}_x\text{Fe}_{3-x}\text{O}_4$ thin films are investigated by the theoretical fitting, in which considering the contributions from intrinsic spin-orbit coupling, extrinsic compositions from two-magnon scattering and inhomogeneous broadening. It is found that the Gilbert damping coefficient α increases with increasing Zn substitution x from 0 to 0.3, indicating that the spin-orbit coupling of the $\text{Zn}_x\text{Fe}_{3-x}\text{O}_4$ thin films increases with the introduction of Zn impurities.

Keywords: $\text{Zn}_x\text{Fe}_{3-x}\text{O}_4$ thin films, doping, FMR, magnetization dynamics

*Corresponding author.

Electronic mail: yazhai@seu.edu.cn.

1. Introduction

$\text{Zn}_x\text{Fe}_{3-x}\text{O}_4$ as a tunable ferromagnetic semiconductor has received considerable attention due to its strong electron correlation and high Curie temperature [1-3]. More desirably, low concentration of Zn substitution is an effective method for tuning the electronic and magnetic properties of Fe_3O_4 over a wide range without changing the half metallic properties [4,5]. These merits make $\text{Zn}_x\text{Fe}_{3-x}\text{O}_4$ system a promising functional material for spintronic devices such as spin-polarized current injection and tunneling magnetoresistance [6,7]. Recent progress of $\text{Zn}_x\text{Fe}_{3-x}\text{O}_4$ thin films was focused on their electrical and static magnetic properties. For instance, Venkateshvaran et al. [7] and Takaobushi et al. [8] studied the effect of Zn doping on the saturation magnetization (M_s) and electronic structures of $\text{Zn}_x\text{Fe}_{3-x}\text{O}_4$ spinel oxide thin films. Recently, Jedrecy et al. [9] presented a detailed magnetic and magnetotransport investigation of textured $\text{Zn}_x\text{Fe}_{3-x}\text{O}_4$ thin films grown on various substrates. In addition, theoretical analyses were also given with tunable electrical and magnetic properties of half-metallic $\text{Zn}_x\text{Fe}_{3-x}\text{O}_4$ by Cheng et al. [5]. On the other hand, the dynamic magnetic properties such as magnetic anisotropy and magnetization damping of $\text{Zn}_x\text{Fe}_{3-x}\text{O}_4$ thin films are less reported may be related to the difficulty of the ultrafast dynamic characterization. Fe_3O_4 thin film has large Gilbert damping and weak magnetic anisotropy in nature [10,11]. Zn dopant in $\text{Zn}_x\text{Fe}_{3-x}\text{O}_4$ thin film can induce extrinsic damping and additional magnetic anisotropy. In this regard, to determine the dynamic magnetic of $\text{Zn}_x\text{Fe}_{3-x}\text{O}_4$

thin film becomes even more difficult, considering the Zn doping effect on the local magnetic structure of Fe_3O_4 .

Ferromagnetic resonance (FMR) is a powerful experimental technique for studying the magnetic properties of thin films [12,13]. It can provide sufficient information describing the magnetic properties, such as effective magnetization, magnetic anisotropy, gyromagnetic ratio, especially magnetization dynamic damping from FMR linewidth. In previous work [14-16], many results have been achieved in the FMR study of the magnetic properties of the Fe_3O_4 films, which could offer a better understanding of the dynamic magnetic properties. Nevertheless, they neglect the other mechanisms in FMR linewidth including two-magnon scattering contribution and inhomogeneous broadening which are also important for the analysis of magnetic relaxation processes. Besides that, the correlation between Zn dopants and dynamic magnetic properties of $\text{Zn}_x\text{Fe}_{3-x}\text{O}_4$ film has not been systematically studied. From these viewpoints, we give a detailed study on the effect of diluted Zn doping on the magnetization dynamic behavior of (111)-textured $\text{Zn}_x\text{Fe}_{3-x}\text{O}_4$ thin films mainly using the magnetic field orientation dependence of FMR spectra. By theoretical fitting of FMR data, the effects of Zn impurities on the various contributions of FMR linewidth such as the intrinsic linewidth, two-magnon scattering broadening and inhomogeneous broadening have been separated and discussed. Dynamic magnetic parameters including magnetic anisotropy, splitting factor g , and Gilbert damping have been obtained, which are significant for oxide spintronic devices designs and applications.

2. Experimental

$\text{Zn}_x\text{Fe}_{3-x}\text{O}_4$ thin films were fabricated using a pulsed laser deposition (PLD) technique with varying Zn concentration ($x = 0, 0.05, 0.1$ and 0.3). The pulse repetition rate was set at 5 Hz and the energy density of the laser beam at the target was 2 J/cm^2 . The films were deposited at 400°C in the background pressure of 6×10^{-7} Torr on the SiO_2 -covered Si (100) substrate. Note the orientation of the Si substrate is here not meaningful since our crystals were terminated by approximately 3 nm- thick SiO_2 layer. Targets were prepared by sintering mixture of $\alpha\text{-Fe}_2\text{O}_3$ and ZnO powders according to stoichiometric ratio. Thin film thickness was fixed to about 40 nm. The crystal structure of the films was determined by x-ray diffraction (XRD) and high resolution transmission electron microscopy (HRTEM) measurements. Surface morphology was investigated by atomic force microscopy (AFM). Static magnetic properties were measured by vibrating sample magnetometer (VSM). The magnetic dynamic properties were investigated by FMR at room temperature using a Bruker Electron Spin Resonance (ESR) equipment (ER-200D-SRC, 9.78 GHz).

3. Results and discussion

Fig. 1. XRD patterns of as-prepared $\text{Zn}_x\text{Fe}_{3-x}\text{O}_4$ thin films. The inset shows the lattice constant of $\text{Zn}_x\text{Fe}_{3-x}\text{O}_4$ thin films.

XRD patterns of the $\text{Zn}_x\text{Fe}_{3-x}\text{O}_4$ thin films with different doping concentrations are shown in Fig. 1. It was confirmed that (111)-oriented spinel-type $\text{Zn}_x\text{Fe}_{3-x}\text{O}_4$ thin films were formed without any secondary phase. Lattice constants were systematically increased from 8.391 Å at $x = 0$ to 8.426 Å at $x = 0.3$, which obeyed Vegard's law, as shown in the inset of Fig. 1. This is expected because of the large ionic radius of Zn^{2+} (0.6 Å) compared to that of Fe^{3+} (0.49 Å) [1]. Therefore, the XRD results demonstrate that the $\text{Zn}_x\text{Fe}_{3-x}\text{O}_4$ thin films represent a complete series of a solid solution system.

Fig. 2. (a) Typical bright field HRTEM image of the Fe_3O_4 film. (b) and (c) HRTEM images and corresponding FFT patterns of marked areas A and B in Fig. 2a, respectively.

The HRTEM images in Fig. 2 provide more detailed information of the morphology and microstructure of the Fe_3O_4 thin film. Fig. The typical cross-sectional

HRTEM micrograph (Fig. 2a) of the Fe_3O_4 thin film, exhibiting the thin film is composed of nanograins with a columnar structure perpendicular to the substrate. The diameter of the columns is in the range of 20-30 nm, and the height of the columns is around 40 nm, i.e., the film thickness. Fig. 2b is the HRTEM image of the marked area A in Fig. 2a, showing one set of lattice fringes, as further confirmed by its fast Fourier transformation (FFT) pattern. However, the HRTEM image (Fig. 2c) of the area marked with B displays two-dimensional lattice fringes, as can be seen in its FFT pattern (inset in Fig. 2c). Interestingly, the columnar grains have different lattice fringes, but their growth direction is the same, that is, both of these areas have a parallel (111) lattice plane, as can be seen in the HRTEM images and FFT patterns. This is confirmed by the lattice spacing of the fringes parallel to the substrate which was measured to be 0.48 nm, corresponding to the (111) plane of Fe_3O_4 . This means that the film is highly (111)-textured, which is consistent with the XRD conclusion.

Fig. 3. Topography images with the same area of $2 \times 2 \mu\text{m}$ for $\text{Zn}_x\text{Fe}_{3-x}\text{O}_4$ thin films, (a) $x = 0$, (b) $x = 0.05$, (c) $x = 0.1$, (d) $x = 0.3$, respectively.

The surface morphology and surface roughness of the films with different Zn concentration are observed by AFM with $2 \times 2 \mu\text{m}$ scan scope, as shown in Fig. 3. We find that the average surface roughness shows an increasing trend after introducing the Zn impurities, the value is 1.3 nm, 1.8 nm, 2.5 nm, and 4.2 nm for increasing Zn concentration respectively. Although a small number of nanogains tend to enlarge size via Zn impurity doping, the average gain size of $\text{Zn}_x\text{Fe}_{3-x}\text{O}_4$ films is not significantly altered, which may be related to the same film thickness and fabrication conditions.

Fig. 4. The magnetic hysteresis loops of the samples with different doping concentrations.

The magnetic properties of the films were examined using VSM with magnetic fields applied in the film plane, as shown in Fig. 4. At room temperature, the magnetic hysteresis loops show that the saturation magnetization (M_s) of $\text{Zn}_x\text{Fe}_{3-x}\text{O}_4$ thin films strongly depends on the concentration of Zn substitution. The M_s shows an increasing trend with increasing Zn substitution concentration x from 0 to 0.1, the value is 349 emu/cm^3 , 368 emu/cm^3 and 465 emu/cm^3 for x at 0, 0.05 and 0.1, respectively. However,

the value is 253 emu/cm^3 at the higher concentration level $x = 0.3$, which significantly reduced compared to the Fe_3O_4 thin film. It is known that Fe_3O_4 is a ferrimagnet with the A-site Fe_A^{3+} and the B-site mixed-valent $\text{Fe}_B^{2+}/\text{Fe}_B^{3+}$ magnetic moments aligned antiparallel to each other, and the total moment is provided by the remaining B-site Fe_B^{2+} ions [7]. Nonmagnetic Zn^{2+} preferably occupies the A-site in the inverse spinel structure. In this way some of the antiferromagnetically coupled moments on the A-site are removed, leading to an initial increase in the saturation magnetization at low Zn substitution ($0 \leq x \leq 0.1$). However, at the higher doping-level, the moments on the A and B-sites no longer need to be rigidly antiparallel, resulting in a finite Yafet-Kittel angle [17]. This angle increases with increasing substitution, so that a reduction in the M_s is expected for a larger Zn concentration ($x = 0.3$).

Fig. 5. Room-temperature FMR spectra for the Fe_3O_4 film at $\theta_H = 0^\circ, 30^\circ, 50^\circ$, and 90° .

Inset: coordinate system used for FMR measurement and analysis.

FMR spectroscopy was used to measure the dynamic magnetic properties of our $\text{Zn}_x\text{Fe}_{3-x}\text{O}_4$ films at 9.78 GHz with various angles θ_H between the dc magnetic field H and the film normal (inset to Fig. 5). Fig. 5 shows four representative FMR spectra from

a Fe_3O_4 thin film at $\theta_H = 0^\circ, 30^\circ, 50^\circ$, and 90° . The resonance field (H_{res}) is defined as the field where the derivative of the FMR absorption crosses zero.

Fig. 6. Angular dependence of resonance field H_{res} on out-of-plane orientation θ_H for $\text{Zn}_x\text{Fe}_{3-x}\text{O}_4$ thin films.

The blue dots in Fig. 6 stand for the experimental angular dependences of FMR field on out-of-plane field orientation for films with different contents of Zn. It is easy to find that the value of H_{res} is minimum when the applied field is along the film plane ($\theta_H = 90^\circ$) and reaches maximum when along the film normal ($\theta_H = 0^\circ$), indicating that the magnetic easy axis is in plane. Furthermore, as the Zn concentration increases, the H_{res} perpendicular to the film plane increases obviously, while in parallel geometry H_{res} decreased slightly. The out-of-plane angular dependence of H_{res} can be fitted by Smit-Beljers approach [18,19]

$$\left(\frac{\omega}{\gamma}\right)^2 = \frac{1}{M^2 \sin^2 \theta} \left[\frac{\partial^2 F}{\partial \theta^2} \frac{\partial^2 F}{\partial \theta \partial \varphi^2} - \left(\frac{\partial^2 F}{\partial \theta \partial \varphi} \right)^2 \right], \quad (1)$$

where θ and φ are angles describing the orientation of the equilibrium magnetization with respect to the film normal and in-plane easy axes, respectively. $\gamma = g\mu_B/\hbar$ is the gyromagnetic ratio, M is the saturation magnetization, and F is the free energy density, including Zeeman energy and anisotropy contributions.

Considering the crucial effect of textured structure on the magnetic anisotropy in $\text{Zn}_x\text{Fe}_{3-x}\text{O}_4$ thin films, the free energy density of our system, introducing an additional expression of texture anisotropy energy (F_t) is [20]

$$F = -\vec{M} \cdot \vec{H} - 2\pi M^2 \sin^2 \theta + (K_{\perp 1} \sin^2 \theta + K_{\perp 2} \sin^4 \theta) + K_{t1} (\cos 2\theta + 7/4 \cos 4\theta) + K_{t2} (\cos 2\theta + 3/2 \cos 4\theta + \cos 6\theta). \quad (2)$$

The first term in Eq. (2) refers to Zeeman energy, the second term is the effective demagnetizing energy. The third term in bracket is related to the magnetic anisotropy energy, in which $K_{\perp 1}$ and $K_{\perp 2}$ are first order and second order perpendicular anisotropy constants, respectively. The remaining terms are defined as the first (K_{t1}) and second (K_{t2}) order magneto-texture energy (F_t), respectively. The equilibrium equation is obtained by minimizing the total free energy density. By combining equilibrium equation and Eq. (1), the dispersion relation for resonance field as a function of field orientation is obtained. We used a numerical procedure to fit the equilibrium angle at resonance condition. As shown in black dotted lines in Fig. 6, the fitting curves agree with the experimental data very well. However, without introduction of F_t related terms in the free energy density F , an obvious deviation of the fitting curves from experiment data (red dotted lines in Fig. 6) is observed for all samples. The deviations indicate the

strong (111)-textured structure related magneto-texture energy should be taken into consideration in the expression of free energy density.

Table 1. Parameters obtained from theoretical fitting.

Zn concentration (x)	0	0.05	0.1	0.3
$4\pi M_{eff}$ (10^3 A/m)	4479	4582	5476	3135
g	2.21	2.25	2.29	2.3
$K_{\perp 1}$ (10^3 J/m ³)	-9.2	-5.9	-2.2	1.5
$K_{\perp 2}$ (10^3 J/m ³)	-1	-1	0	0.5
K_{t1} (10^3 J/m ³)	-0.5	-0.3	-0.2	-0.2
K_{t2} (10^3 J/m ³)	0.1	0.45	0.55	0.6

The values of fitted magnetic parameters including the effective field ($4\pi M_{eff}$), $K_{\perp 1}$, $K_{\perp 2}$, K_{t1} , K_{t2} , and g factor for $Zn_xFe_{3-x}O_4$ films are listed in Table 1. The effective field $4\pi M_{eff} = 4\pi M_s - 2K_{\perp 1}/M_s$, where M_s is the saturation magnetization of $Zn_xFe_{3-x}O_4$ thin films measured from VSM. It is clear from Table I that $4\pi M_{eff}$ obtained from obtained from FMR fitting follows a similar variation as $4\pi M_s$ as a function of Zn doping concentration x. It is interesting that the evolution of the perpendicular anisotropy constants $K_{\perp 1}$ and $K_{\perp 2}$ with increasing Zn contents displays a spin-reorientation behavior. Both the $K_{\perp 1}$ and $K_{\perp 2}$ increase from negative to positive with increasing Zn concentration, which indicates that the easy axis of magnetic anisotropy changes from the film plane to the film normal. However, the preferred direction of the magnetization vector is always in plane for all films due to larger demagnetizing field. The magneto-texture energy constant $K_{t1} + K_{t2}$ of $Zn_xFe_{3-x}O_4$ films increases with increasing Zn doping concentration, which implies that the Zn dopants could increase the magnetic anisotropy induced by texture structure. The splitting factor

g increases slightly from 2.21 to 2.3 with increasing Zn concentration x from 0 to 0.3, indicating that the Zn impurities also affect the g factor of $\text{Zn}_x\text{Fe}_{3-x}\text{O}_4$ films.

The FMR linewidth is one of the most sensitive parameters of ferromagnetic resonance in thin films. It can provide information about relaxation processes of magnetization, which can be influenced by structural and magnetic inhomogeneities. In general, three contributions are considered for the peak to peak FMR linewidth, which include intrinsic Gilbert-type and extrinsic non-Gilbert-type mechanisms [21,22]

$$\Delta H = \Delta H_{\text{Gilb}} + \Delta H_{2\text{M}} + \Delta H_{\text{inhom}}. \quad (3)$$

Where the ΔH_{Gilb} is the intrinsic linewidth due to Gilbert damping (α) related to the dissipation of magnetic energy. The Gilbert damping is understood to be a spin relaxation process refer to spin-orbit coupling, which is important in determining the magnetization dynamics and the relaxation time. For out-of-plane angles, the ΔH_{Gilb} is given by [22]

$$\Delta H_{\text{Gilb}} = \frac{1}{\sqrt{3}} \frac{\alpha}{M_s} \left(\frac{\partial^2 F}{\partial \theta^2} + \frac{\partial^2 F}{\partial \varphi^2} (\sin^2 \theta)^{-1} \right) \left| \frac{d(\omega/\gamma)}{dH} \right|^{-1}. \quad (4)$$

The second term, $\Delta H_{2\text{M}}$, in Eq. (3) is the two-magnon scattering contribution to the linewidth. The scattering of the precession of the uniform mode (wave vector $k = 0$) into non uniform ($k \neq 0$) ones is mediated by sample defects and results in an additional contribution to the FMR linewidth. To describe the two-magnon contribution to the out-plane angles dependence of FMR linewidth in our films, we use the Arias-Mills formulation [23]

$$\Delta H_{2\text{M}} = \frac{2}{\sqrt{3}} \Gamma(H_0, \theta_H) \sinh^{-1} \sqrt{\frac{H_x}{H_x + M_{\text{eff}}} \frac{\cos(2\theta_M)}{\cos^2 \theta_M}}. \quad (5)$$

In the above expressions, the term $\Gamma(H_0, \theta_H)$ is given by

$$\Gamma(H_0, \theta_H) = \frac{8H_u^2 b^2 p / \pi D}{(H_x + H_y)^2} \frac{d\omega}{dH} \left\{ \left(\left\langle \frac{c}{a} \right\rangle - 1 \right) H_y^2 + \left(\left\langle \frac{a}{c} \right\rangle - 1 \right) \times [H_x \cos(2\theta_M) + \cos^2 \theta_M]^2 + [H_x \cos(2\theta_M) - H_y \sin^2 \theta_M]^2 \right\}. \quad (6)$$

Here, $D = 2A/M_s$ is the exchange stiffness of the ferromagnetic material. The defects are supposed to cover the fraction p of the film surface and to be rectangular in nature, with height b and lateral dimensions a and c randomly distributed. H_u is uniaxial out-of-plane anisotropy field, $H_x = H_0 \cos(\theta_H - \theta_M) - M_{eff} \sin^2(\theta_M)$ and $H_y = H_0 \cos(\theta_H - \theta_M) - M_{eff} \cos(2\theta_M)$. Therefore, by adjusting the value of $H_u^2 b^2 p / \pi D$ and $\langle \frac{c}{a} \rangle$, we can obtain information related to the extrinsic two-magnon process. For the FMR linewidth with main contributions from two-magnon scattering, there is a larger broadening in the parallel configuration of the film and no contribution to the perpendicular configuration, thus $\Delta H_{\perp} < \Delta H_{\parallel}$. As shown in Fig. 7, the measured linewidth in the perpendicular configuration $\Delta H_{\perp}$ is obviously smaller than that in the parallel configuration $\Delta H_{\parallel}$ for all samples, which indicates that the contribution of two-magnon scattering could not be neglected. The calculated FMR linewidth due to two-magnon scattering (blue dash lines in Fig. 7) was found to be comparable with the intrinsic composition (red dash lines). Besides, it is interesting to note that the contribution of two-magnon scattering has slightly change with increasing Zn concentration, which indicates that there is no correlation between Zn dopants and two-magnon scattering. The interface between amorphous SiO_2 and highly textured $\text{Zn}_x\text{Fe}_{3-x}\text{O}_4$ films may be responsible for the two-magnon scattering linewidth

broadening [24].

Fig. 7. Angular dependence of different linewidth contributions related to the intrinsic linewidth, two-magnon scattering and the inhomogeneous broadening for $\text{Zn}_x\text{Fe}_{3-x}\text{O}_4$ thin films.

The last term ΔH_{inhom} is the inhomogeneous linewidth broadening due to the inhomogeneity of the films, which can be expressed as [25]

$$\Delta H_{\text{inhom}} = \frac{1}{\sqrt{3}} \left| \frac{\partial H}{\partial 4\pi M_{\text{eff}}} \right| \Delta 4\pi M_{\text{eff}} + \frac{1}{\sqrt{3}} \left| \frac{\partial H}{\partial \theta_H} \right| \Delta \theta_H. \quad (7)$$

Where $\Delta 4\pi M_{\text{eff}}$ and $\Delta \theta_H$ represent the resonance linewidth broadening caused by the dispersion of magnitude and the direction of $4\pi M_{\text{eff}}$, respectively. By adjusting the value of $\Delta 4\pi M_{\text{eff}}$ and $\Delta \theta_H$, we can obtain information related to the inhomogeneity linewidth broadening. For inhomogeneous films, the inhomogeneous linewidth broadening exhibits a larger contribution to the linewidth in the perpendicular configuration than the contribution in the parallel configuration, which shows that $\Delta H_{\perp} > \Delta H_{\parallel}$. The cyan dash

lines in Fig. 7 are shown the calculated curves of inhomogeneous linewidth, it is evident that the ΔH_{inhom} sensitively depends on the Zn substitution x . The inhomogeneous linewidth increases with increasing Zn^{2+} concentration x from 0 to 0.3, implying that the structure of the films becomes more inhomogeneous as Zn content rises. According to Luo's argument [25], ΔH_{inhom} will be influenced by the surface roughness of the thin films. In our case, the values of the surface roughness show a very strong dependence on the Zn concentration as shown in Fig. 3, which is similar to inhomogeneous linewidth broadening.

Fig. 8. Gilbert damping parameter α as a function of the Zn concentration.

The procedure used to determine the FMR linewidth was determined using the Eqs. (1)-(7). The parameters used to fit the angular dependence of the FMR linewidth are the Gilbert damping coefficient α of Eq. (4), $H_u^2 b^2 p / \pi D$ and $\langle \frac{c}{a} \rangle$ of the expression corresponding to the two-magnon contribution (Eq. (5)), as well as $\Delta 4\pi M_{\text{eff}}$ and $\Delta \theta_H$ of Eq. (7). The values of $4\pi M_{\text{eff}}$ and the anisotropy fields needed for fitting the linewidth data were determined by the angular dependence of FMR field theoretical fitting (see Table 1), with these parameters held constant, these values serve in the fit of the theory

to the experimental linewidth. By adjusting the three parts of FMR linewidth contribution, we obtain the most appropriate theoretical curve, and then the intrinsic FMR linewidth ΔH_{Gilb} . Taking into account only one of the mechanisms through to Eqs. (4), (5), or (7), one obtains the dashed curves (Fig. 7). This shows that each of them separately is not sufficient to describe the magnetization relaxation of the films. The entire angular dependence of the FMR linewidth (solid line) can be explained by adding the two-magnon scattering and local inhomogeneous effects. The Gilbert damping parameter α , calculated from the Eq. (4), shows a strong dependence on the Zn concentration, as shown in Fig. 8. With increasing Zn concentration x from 0 to 0.3, the Gilbert damping value increases from 0.095 to 0.115. It is known that the splitting factor g is related with the orbital and spin moments, $\mu_l/\mu_s = (g - 2)/2$, and also according to Pelzl et al. [26], there is a close relation between g value and α , α should be proportional to $(g - 2)^2$. In our case, the obtained α of $\text{Zn}_x\text{Fe}_{3-x}\text{O}_4$ thin films is roughly proportional to the $(g - 2)^2$, suggesting the increase in spin-orbit coupling (SOC) by Zn doping increases the magnetic damping constant of $\text{Zn}_x\text{Fe}_{3-x}\text{O}_4$ films. Although we do not calculate the SOC strength, but the same tendency of the increase in α and $(g - 2)^2$ was obtained. This result is consistent with previous reports [25,27,28], in which the increase in α were achieved by rare earth element doping in $3d$ ferromagnetic via SOC.

4. Conclusion

In summary, we have investigated the structural and magnetization dynamic

behavior of Fe_3O_4 thin films doped with various amounts of Zn. The distinct HRTEM images and corresponding FFT patterns reveal that highly (111)-textured structured films are composed of nanograins with a columnar structure grown perpendicular to the substrate. From the theoretical fitting, it is found that a texture structure related magnetic texture anisotropy energy should be taken into consideration in the expression of free energy density F . The perpendicular anisotropy is enhanced due to the Zn dopants. The calculations of the angular variations of linewidth agreed quantitatively with the experimental ones. There is no correlation between Zn dopants and two-magnon scattering. However, the contribution of the inhomogeneous linewidth broadening increases with increasing Zn concentration. The Gilbert damping parameter α increases with increasing Zn content, which indicates that the Zn impurities affect the spin-orbit coupling of Fe_3O_4 film. Our results will contribute to the understanding of anisotropy, and magnetization dynamics in Zn-doped Fe_3O_4 thin films.

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Figure Captions

Fig. 1. XRD patterns of as-prepared $\text{Zn}_x\text{Fe}_{3-x}\text{O}_4$ thin films. The inset shows the lattice constant of $\text{Zn}_x\text{Fe}_{3-x}\text{O}_4$ thin films.

Fig. 2. (a) Typical bright field HRTEM image of the Fe_3O_4 film. (b) and (c) HRTEM images and corresponding FFT patterns of marked areas A and B in Fig. 2a, respectively.

Fig. 3. Topography images with the same area of $2 \times 2 \mu\text{m}$ for $\text{Zn}_x\text{Fe}_{3-x}\text{O}_4$ thin films, (a) $x = 0$, (b) $x = 0.05$, (c) $x = 0.1$, (d) $x = 0.3$, respectively.

Fig. 4. The magnetic hysteresis loops of the samples with different doping concentrations.

Fig. 5. Room-temperature FMR spectra for the Fe_3O_4 film at $\theta_H = 0^\circ, 30^\circ, 50^\circ$, and 90° . Inset: coordinate system used for FMR measurement and analysis.

Fig. 6. Angular dependence of resonance field H_{res} on out-of-plane orientation θ_H for $\text{Zn}_x\text{Fe}_{3-x}\text{O}_4$ thin films.

Fig. 7. Angular dependence of different linewidth contributions related to the intrinsic linewidth, two-magnon scattering and the inhomogeneous broadening for $\text{Zn}_x\text{Fe}_{3-x}\text{O}_4$ thin films.

Fig. 8. Gilbert damping parameter α as a function of the Zn concentration.