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Static and high frequency magnetic properties of FeGa thin films deposited on convex flexible substrates

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Magnetostrictive FeGa thin films were deposited on the bowed flexible polyethylene terephthalate (PET) substrates, which were fixed on the convex mold. A compressive stress was induced in FeGa films when the PET substrates were shaped from convex to flat. Due to the effect of magnetostriction, FeGa films exhibit an obvious in-plane uniaxial magnetic anisotropy which could be enhanced by increasing the applied pre-strains on the substrates during growth. Consequently, the ferromagnetic resonance frequency of the films was significantly increased, but the corresponding initial permeability was decreased. Moreover, the films with pre-strains less than 0.78% exhibit a working bandwidth of microwave absorption about 2 GHz. Our investigations demonstrated a convenient method via the pre-strained substrates to tune the high frequency properties of magnetic thin films which could be applied in flexible microwave devices. © *2015 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4918964]

Magnetic thin films applied in high-frequency devices, such as recording heads, wireless inductor, and microwave noise filters, have attracted considerable attentions due to the ever-increasing demands for miniaturization in electromagnetic devices.[1–3] So far, most of previously investigated magnetic thin films for microwave applications were usually fabricated on rigid substrates such as silicon and glass.[4–6] Recently, the high-frequency electronic devices fabricated on plastics have been developed for the potential applications in Wi-Fi devices, wearable radios, and foldable phased-array antennas.[7,8] Compared with the devices grown on rigid substrates, the flexibility of high-frequency devices provides the advantages of enhanced durability, light weight, easy to bend and fold, optically transparent, and mountable to uneven surfaces.[8–12] In addition, the magnetic thin films deposited on flexible substrates often exhibit the excellent performance better than the films grown on rigid substrates. Liu *et al.* found that the FeTaN films deposited on flexible Kapton substrates possess a relatively higher ferromagnetic resonance (FMR) frequency f_r than that of the films deposited on rigid Si substrates due to the enhanced anisotropy fields caused by the formation of magnetization ripple in the films.[13]

For magnetic thin films applied in high-frequency devices, the complex permeability ($\mu = \mu' - j\mu''$) is the most important factor determining whether the magnetic thin film is suitable for this application. For the magnetic thin film applied in planar inductors, it requires the thin films have relatively high values of μ' and low values of μ'', and f_r of the magnetic thin films need to be higher than the working frequencies of the devices.[14] Hence, from an application point of view, it is desirable that the complex permeability spectra and the FMR frequency of magnetic films could be tuned to meet various requirements of application.[2,14,15] According to the Kittel equation, the FMR frequency of a magnetic film is determined by the magnetic anisotropy H_k as[15]

$$f_r = \frac{\gamma}{2\pi}\sqrt{H_k(H_k + 4\pi M_s)}, \tag{1}$$

where γ is the gyromagnetic ratio and M_s is the saturation magnetization. In order to achieve a high FMR frequency in the magnetic films, several experimental methods including oblique deposition,[16,17] annealing in magnetic field,[18] and interfacial exchange coupling,[19] have been often used to increase the magnetic anisotropy of thin films. For magnetic films deposited on flexible substrates, because of the effect of magnetostriction, the magnetic anisotropy can be substantially changed with the external strain produced by mechanical deformation.[20]

FeGa alloys, which exhibit moderate magnetostriction, relative low saturation field, and good mechanical properties, have been extensively applied in stress sensors and actuators. Recently, there were numerous interests in creating energy efficient, faster, and smaller high-frequency devices based on FeGa films for the information and communication technologies.[21] In the previous works, we have reported that the mechanical stress and the pre-strain produced by bending substrates, by means of modifying the strength and the orientation of the uniaxial magnetic anisotropy, could remarkably change the remanent magnetization of FeGa films and the

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exchange bias of FeGa/IrMn heterostructures.[20,22,23] In this paper, we provided an approach of pre-strained growth to significantly enhance the FMR frequency of flexible magnetic films. When the bowed FeGa films were flattened to a plane, they were suffered a compressive strain arisen from the shaped substrates, which induces a controllable magnetic anisotropy. Consequently, the FMR frequency of FeGa films was enhanced, but the corresponding permeability was reduced. Compared to the field annealing and the oblique deposition, the method via pre-strain of flexible substrates has the great advantage in increasing both the magnetic anisotropy and the FMR frequency.

$Fe_{81}Ga_{19}$ (FeGa) films with 100 nm in thickness were fabricated on flexible 150-μm-thick polyethylene terephthalate (PET) plastics at room temperature using a magnetron sputtering system with a base pressure better than 1×10^{-7} Torr. Before introducing PET plastics into the sputtering chamber, they were cleaned in acetone and ethyl alcohol using ultrasonic agitation for 15 min, blow-dried with nitrogen gas, then bowed and fixed onto the convex surface of aluminum alloy molds with curvature radii of 30, 15, and 10 mm, as shown in Fig. 1(a). Moreover, a reference FeGa film was grown on the flat PET plastics as well. During deposition, the Ar pressure was set at 1.5×10^{-3} Torr and the argon flow was kept at 15 sccm. The deposition rate of FeGa films was 3 nm/min. Prior to be taken out of the chamber, a 3-nm-thick Ta capping layer was deposited on FeGa films to avoid oxidation. The static hysteresis loops were measured by using vibrating sample magnetometer (VSM, Lakeshore 7410) at various magnetic field orientations of θ with respect to the direction perpendicular to the bending strain. The permeability spectra over the frequency range from 0.1 to 8 GHz were obtained in zero bias field by using a vector network analyzer (Agilent E8363B) with a shorted microstrip transmission-line perturbation method. All the measurements were conducted at room temperature.

As shown in Fig. 1(a), the as-grown FeGa films deposited on the bowed PET substrates are not stressed. After they are released from the convex molds and flattened to a plane, the films are subjected to a compressive strain due to the clamping of substrates. The strain s can be evaluated by using the relation of $s = d/2\rho$, where d is the total thickness of both the substrate and the film and ρ is the curvature radii of the convex surface of the molds. The curvature radii of 30, 15, and 10 mm correspond to the compressive strains of 0.26%, 0.52%, and 0.78% applied on the flatten films, respectively. The corresponding stress σ can be evaluated by using the relation $\sigma = sE_f/(1-\nu^2)$, where E_f and ν denote the Young's module and Poisson ratio of FeGa films, respectively. Due to the positive magnetostriction of FeGa, the compressive strain gives rise to a uniaxial magnetic anisotropy with the easy axis along $\theta = 0°$ and the hard axis along $\theta = 90°$. Figures 1(b) and 1(c) show the hysteresis loops measured with the magnetic field applied along the easy and hard axes, respectively. The unstrained reference FeGa film exhibits a relatively square loop at $\theta = 0°$ and a sheared loop at $\theta = 90°$. The obvious feature of uniaxial magnetic anisotropy is likely produced by the residual stress caused by the slightly inevitable deformation of PET substrates.[20,24] With the increase of the compressive strain from 0% to 0.78%, the remanence ratio M_r/M_s of hysteresis loops measured along the easy axis inconspicuously increases from 0.89 to 0.97, indicating the transverse alignment of the magnetic moments under the compressive strain. Due to the enhanced uniaxial anisotropy, the coercive field H_c measured along the easy axis correspondingly increases from 65 to 159 Oe. For the hysteresis loops measured along the hard axis, with increasing the compressive strain from 0% to 0.78% the value of M_r/M_s dramatically decreases from 0.78 to 0.21 and H_c increases from 36 to 87 Oe. The strain induced uniaxial magnetic anisotropy K_u of the FeGa films can be experimentally determined from the difference of the area enclosed between the hysteresis loops measured along the easy and hard axes.[25] As shown in Fig. 1(d), with the increase of compressive strain from 0% to 0.78%, K_u of the FeGa films increases from 1.01×10^5 to 1.86×10^5 erg/cm^3. In addition, according to the relation[22] $\Delta K_u = 3/2\lambda_s \Delta\sigma$, the magnetostriction coefficient λ_s of the FeGa films can be obtained by the linear fitting for the stress dependence of ΔK_u. Using $E_f = 60$ GPa for FeGa and the typical value of $\nu = 0.3$ for metals,[26,27] one can

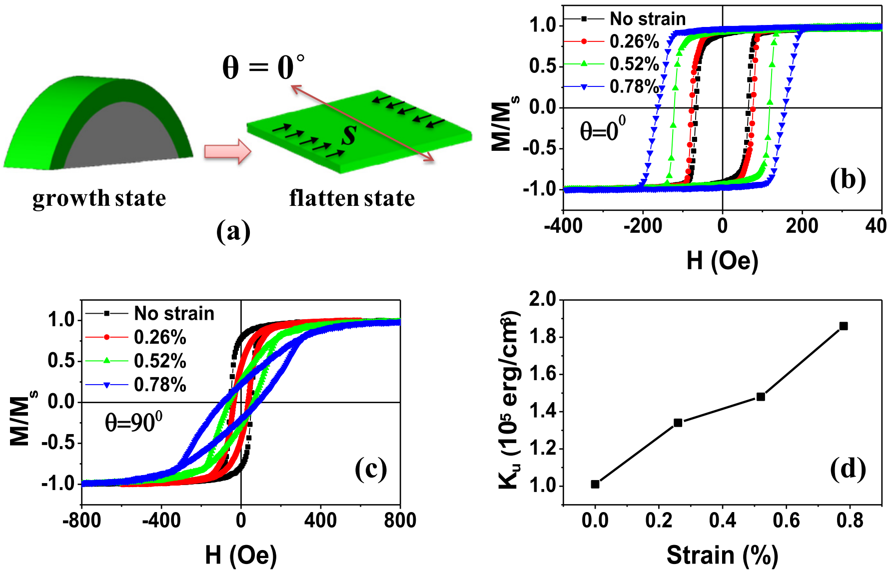

FIG. 1. (a) Schematic show of the bowed-substrates during the growth of FeGa films. After flatten to a plane, a compressive strain is induced in the films. The direction perpendicular to the compressive strain is defined as $\theta = 0°$. Hysteresis loops measured along (b) the easy axis and (c) the hard axis for FeGa films grown under different pre-strains of substrates. (d) The compressive strain dependence of the uniaxial magnetic anisotropy K_u of the FeGa films.

obtain $\lambda_s = 20$ ppm. This value is far below the magnetostriction coefficient of the bulk counterpart, but agrees well with our previously reported value for the FeGa polycrystalline films.[22] The significantly reduced magnetostriction constant of FeGa films can be ascribed to the dominative interface contribution to the effective magnetostriction.[28]

Figures 2(a) and 2(b) present the experimental measured real (μ') and imaginary (μ'') permeability spectra in zero bias field for the samples grown with different pre-strains in the frequency range from 0.5 to 8 GHz. With the increase of compressive strain, the initial permeability μ_i of the FeGa films inconspicuously decreases, while the FMR frequency corresponding to the resonance peak of the imaginary spectra shifts toward the higher values.

In order to quantitatively understanding the effect of strain onto the dynamic properties of the FeGa thin films, the complex permeability can be described by the Landau-Lifshitz-Gilbert (LLG) equation. By means of solving the LLG equation, one can readily obtain the equations for the complex permeability as[29]

$$\mu' = 1 + \chi_0 \frac{1 + (\alpha^2 - 1)\left(\frac{f}{f_r}\right)^2}{\left[1 - (1+\alpha^2)\left(\frac{f}{f_r}\right)^2\right]^2 + \left(2\alpha\frac{f}{f_r}\right)^2}, \tag{2}$$

$$\mu'' = \chi_0 \frac{\alpha\left(\frac{f}{f_r}\right)\left[1 + (1+\alpha^2)\left(\frac{f}{f_r}\right)^2\right]}{\left[1 - (1+\alpha^2)\left(\frac{f}{f_r}\right)^2\right]^2 + \left(2\alpha\frac{f}{f_r}\right)^2}, \tag{3}$$

where $\chi_0 = \mu_i - 1$ is the initial susceptibility of the FeGa films, α is the damping parameter and f is the operation frequency. Taking α, χ_0, and f_r as the fitting parameters, the experimentally obtained real and imaginary permeability spectra can be, respectively, fitted by using Eqs. (2) and (3), which yields the damping parameter α about 0.1. This value is slightly higher than the soft magnetic thin films applied in high frequency area but close to the value in the references.30,31 With the compressive strain increasing from 0% to 0.78%, μ_i decreases from 69 to 17, whilst f_r increases from 4.6 to 5.3 GHz, as shown in Fig. 2(c). The opposite features between the strain dependent χ_0 and f_r can be explained by using the Snoek-Archer's limit:[32] $(\mu_i - 1)f_r^{\,2} = \left(\frac{\gamma}{2\pi}4\pi M_s\right)^2$. For a magnetic thin film with a certain M_s, the increase of f_r will lead to the decrease of μ_i. Therefore, in order to obtain a thin film with both the high μ_i and f_r, the magnetic materials with a high M_s need be chosen. In our experiment, M_s of FeGa films is measured to be 880 emu/cm^3, which is less than that of the bulk counterpart but suitable for the application in high-frequency devices.[33,34] f_r of the FeGa films with different compressive strains can also be theoretically calculated by using Eq. (1). The calculated f_r are shown in Fig. 2(c), where the parameters of $\gamma/2\pi = 2.8 \times 10^6$ Hz/Oe (Ref. 30) and the values of H_k obtained from the "area method" are used. It is noticed that the calculated f_r is slightly lower than the fitted f_r. This deviation can be interpreted in term of the Hoffmann's ripple theory. In case of dynamic magnetic measurement, there is an additional effective isotropic field that contributes to the anisotropy field. This additional effective field depends on a so-called ripple constant which may originate from the local random anisotropies. However, in the static magnetic measurement, the additional effective field is not included.[30,35] Using the relation $\Delta f = \gamma\alpha(4\pi M_s + 2H_k)/2\pi$,[30] the strain dependence of the frequency linewidth Δf can be calculated, as shown in Fig. 2(d).

The reflection loss (RL) for a magnetic film, which reveals the loss of electromagnetic wave that propagates in a magnetic film and reflect by the film, can be calculated by using the equation:[15] $\mathrm{RL} = 20\log\left|\frac{Z_{in}-1}{Z_{in}+1}\right|$, where $\mathrm{Z_{in}}$ is the normalized input impedance and given by $Z_{in} = \sqrt{\frac{\mu}{\varepsilon}}\tanh\left(\frac{j2\pi fd}{c}\sqrt{\varepsilon\mu}\right)$. Here, $\varepsilon = \varepsilon' - j\varepsilon''$ is the relative complex permittivity of the thin films and c is the velocity of electromagnetic

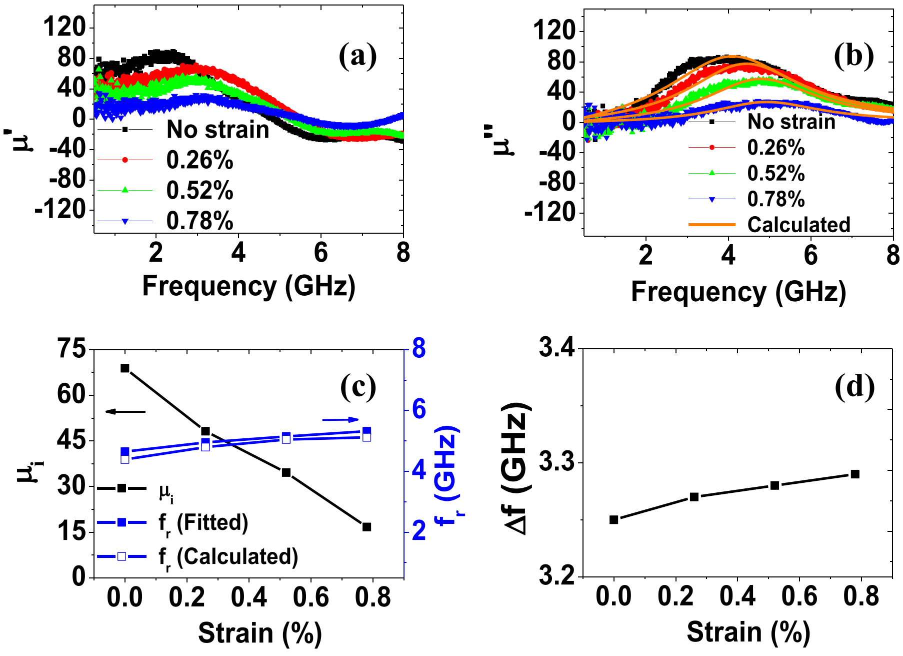

FIG. 2. (a) Real and (b) imaginary permeability spectra (the lines are the fitting curves based on LLG equation) obtained in zero bias field for FeGa films grown under various pre-strains. (c) Strain dependences of the initial permeability, the fitted FMR frequency (solid symbols), and the calculated FMR frequency based on the static magnetic measurements (open symbols). (d) Strain dependence of frequency linewidths.

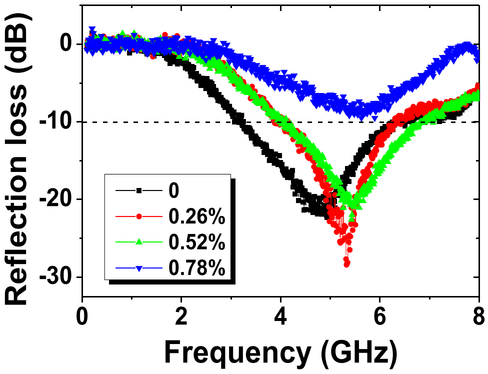

FIG. 3. Summary of reflection loss spectra of FeGa films under different compressive strains.

waves in free space. Generally, the complex permittivity of thin flexible organic substrates is closed to each other, here, we use $\varepsilon' = 2.5$ and $\varepsilon'' = 0.06$.[15] Based on the experimentally measured permeability spectra, one can calculate the RL as a function of the microwave frequency, as plotted in Fig. 3. The maximum attenuation of RL = 28 dB is achieved for the sample with a compressive strain of 0.26% at 5.3 GHz. The working bandwidths, i.e., the absorption width where the RL is less than 10 dB,[36] are from 3.2 to 6.4 GHz, 4 to 6.3 GHz, and 4 to 6.9 GHz for the films with pre-strains of 0%, 0.26%, and 0.52%, respectively. For the sample grown with pre-strain of 0.78%, the attenuation is very poor due to the small values of the μ''.

In summary, magnetostrictive FeGa thin films were deposited by using magnetron sputtering onto flexible PET substrates, which were fixed on the convex molds. Due to the effect of magnetostriction, the uniaxial magnetic anisotropy of the FeGa films can be enhanced by increasing the pre-strain of the flexible substrates during growth. As a result, the FMR frequency is significantly increased, but the corresponding initial permeability is decreased due to the Snoek-Archer's limit. The reflection loss of the films exhibits a remarkable dependence on the pre-strain. For the films with pre-strains less than 0.78%, the working bandwidths of FeGa thin films reach 2 GHz. Our investigation implies that the method of pre-strained substrates is a convenient and effective way to tune the FMR frequency of magnetic thin films deposited on flexible substrates for specific high frequency applications.

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