Ferromagnetic resonance frequency shift model of laminated magnetoelectric structure tuned by electric field*

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Based on Smith–Beljers theory and classical laminate theory, an explicit model is proposed for the ferromagnetic resonance (FMR) frequency shift of a stress-mediumed laminated magnetoelectric structure tuned by an electric field. This model can effectively predict the experimental phenomenon that the FMR frequency increases under a parallel magnetic field and decreases under a perpendicular magnetic field when the electric field ranges from −10 kV/m to 10 kV/m. Besides, this theory further shows that the FMR frequency increases monotonically as the angle between the direction of the external magnetic field and the outside normal direction of the laminated structure increases, and the frequency will increase as great as 7 GHz. In addition, when the angle reaches a certain critical value, the external electric field fails to tune the FMR frequency. When the angle is above the critical value, the increase of the electric field induces the FMR frequency to increase, and the opposite scenario happens when it is below the critical value. When the angle is 90° (parallel magnetic field), the FMR frequency is the most sensitive to the change of the electric field.

Keywords: ferromagnetic resonance (FMR), piezoelectric/ferrite laminates, converse magnetoelectric effect, microwave devices

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1. Introduction

Due to the large magnetoelectric (ME) coupling effect, ME compsite materials have great potential applications in high-tech equipments and integrated applications,[1] such as biological sensors, magnetic sensors, ME oscillators, ME conversion cells, and other ME devices[2–6] under low frequency,[7–13] and especially electric and magnetic dual-tunable devices, owing to the characteristics of the electric field tuning ferromagnetic resonance (FMR) frequency shift under high frequency.[14–16] Tunable microwave devices can meet the requirements of modern radar and communications technology to switch quickly from one operating frequency to another. Therefore, they have become a great concern in academic, industrial, and especially military fields.

Bichurin *et al.*[17] took the lead in carrying out theoretical studies of the microwave ME effect in laminated ME materials as early as in 2001. Assuming piezoelectric-ferrite layers to be a homogeneous medium, they proposed a theory of the microwave ME effect, and predicted that the FMR frequency can be tuned by an electric field. In 2004, Shastry *et al.*[18] observed the phenomenon of an electric tuning FMR frequency shift in ME composites in experiments for the first time, and they expanded the application range of Bichurin's theory to single-crystal bilayer structures. Since then, experimental researches of tunable microwave devices made of laminated ME materials have made great progress, such as the appearance of tunable ME resonators,[19–22] ME filters,[23,24] phase shifters, and antennas.[25,26] To solve the problem that the shift of FMR frequency caused by an electric field is relatively small, more and more researchers begin to focus on the exploration for larger electric tenability.[15,27–31] Lou *et al.*[29] found that the shift of FMR frequency can reach 900 MHz by using FeGaBa/PMN-PT structure. Liu *et al.*[31] discovered that the equivalent FMR field can reach up to 860 Oe (1 Oe = 79.5775 A/m) in Fe_3O_4/PZN-PT layers. In fact, the ME coupling effect is related to a number of factors, such as the type of piezoelectric phase, the direction of the external magnetic field, volume ratio of two phases, boundary conditions of laminated structure, microwave frequency, etc.[31–34] Recently, Bichurin *et al.*[35] have given a theoretical analysis of the high-frequency ME effect in ferrite/piezoelectric layers. Their theoretical predictions show that there exists a giant ME effect in a magnetoacoustic resonance area.

Although many studies on the microwave ME effect have been conducted, there is little theoretical explanation to the experimental phenomenon that the FMR frequency increases under a parallel magnetic field while decreases under a perpendicular magnetic field when the electric field changes.[20] What is more, the problem why the angle between the external magnetic field and the film surface influences the electric tunability still needs to be resolved. In this paper, we deduce a concise expression from free energy density, describing the

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influence of stress on the shift of FMR frequency according to Smith and Beljers theory. Furthermore, taking into account the fact that the in-plane dimension of the laminated structure is far larger than the transverse dimension, we adopt plane stress theory and free boundary conditions, and establish a stress expression of lamination under an external electric field. Finally, combining these two formulas, we establish a concise theoretical model of the electric field-tuning FMR frequency.

2. FMR frequency shift model

The structure of the laminated composite consisting of piezoelectric and ferrite materials is shown in Fig. 1. Taking into account the isotropic characteristics of ferrite thin films, we define the ferrite surface as the x–y plane and the z axis as being perpendicular to the film surface, and we use spherical coordinates. Let β be the polar angle between external magnetic field H and the z axis, $\beta_{\rm H}$ the azimuth angle between H and the x axis, θ the polar angle between saturation magnetization M and the z axis, φ the azimuth angle between M and the x axis, α the polar angle between the stress and the z axis, and assume the stress to be in the x–z plane, then the stress energy density will be given below

$$E_\lambda = -\frac{3}{2}\lambda\sigma\cos^2\phi, \tag{1}$$

where λ is the magnetostriction coefficient, and ϕ is the angle between the stress and the direction of magnetization. Clearly, we have $\cos\phi = \cos\theta\cos\alpha + \sin\theta\sin\varphi\sin\alpha$.

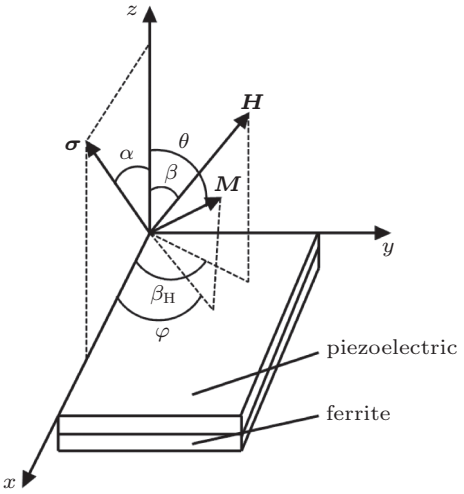


Fig. 1. Schematic diagram of ME laminated structure.

Next, the free energy density can be written as

$$\begin{aligned} E = & -\frac{3}{2}\lambda\sigma(\cos^2\theta\cos^2\alpha + \sin^2\theta\sin^2\varphi\sin^2\alpha \\ & + \frac{1}{2}\sin 2\theta\cos\varphi\sin 2\alpha) \\ & - 2\pi M^2\sin^2\theta - MH(\cos\theta\cos\beta \\ & + \sin\theta\sin\beta\cos(\varphi-\beta_{\rm H})), \end{aligned} \tag{2}$$

where the first term on the right-hand side represents the stress energy, the second term the demagnetization energy, the third term the Zeeman energy of the ferrite layer under the external magnetic field, and M denotes the saturation magnetization.

In the experiment, the magnetic field is usually along the x axis or z-axis direction. For simplicity, we set the direction of the external magnetic field to be in the x–z plane. Surely, the discussion is similar if the direction of the external magnetic field is in the y–z plane. In the case where the external magnetic field is oriented along the x–z plane, we can obtain $\beta_{\rm H} = 0$ in Eq. (2). The minimum energy principle in an equilibrium state is expressed as follows:

$$\frac{\partial E}{\partial\theta} = 0, \quad \frac{\partial E}{\partial\varphi} = 0. \tag{3}$$

Substituting Eq. (2) into Eq. (3), we can obtain $\varphi = 0$, which is the solution of the equilibrium equation. In that case, $\boldsymbol{M}$ also will be in the x–z plane. The angle θ between $\boldsymbol{M}$ and the z axis will be determined by

$$\frac{3\lambda\sigma}{2M}\sin 2(\theta-\alpha) - 2\pi M\sin 2\theta + MH\sin(\theta-\beta) = 0. \tag{4}$$

According to Smith and Beljers theory, the standard conditions of FMR can be represented by

$$\left(\frac{\omega}{\gamma}\right)^2 = \frac{1}{M^2\sin^2\theta}\left[\frac{\partial^2 E}{\partial\theta^2}\frac{\partial^2 E}{\partial\varphi^2} - \left(\frac{\partial^2 E}{\partial\theta\partial\varphi}\right)^2\right], \tag{5}$$

where ω is the FMR frequency. Substituting Eq. (2) into Eq. (5) and combing what we obtain with $\phi = 0$, we have

$$\begin{aligned} \left(\frac{\omega}{\gamma}\right)^2 = & \left[3\frac{\lambda\sigma}{M}\cos 2(\theta-\alpha) - 4\pi M\cos 2\theta + H\cos(\theta-\beta)\right] \\ & \times\left[3\frac{\lambda\sigma}{M}\cos^2(\theta-\alpha) - 4\pi M\cos^2\theta \right. \\ & \left. + H\cos(\theta-\beta)\right]. \end{aligned} \tag{6}$$

After obtaining the expression of the effect of the stress on the FMR frequency, we discuss the stress of the laminated structure, caused by applying an electric field to the piezoelectric layer. Typically, the linear constitutive of the piezoelectric phase can be denoted by the following expressions:

$$^{\rm p}\varepsilon_{ij} = {}^{\rm p}\boldsymbol{S}_{ijkl}{}^{\rm p}\boldsymbol{\sigma}_{kl} + {}^{\rm p}\boldsymbol{d}_{kij}\boldsymbol{E}_k, \tag{7}$$

$$\boldsymbol{D}_i = {}^{\rm p}\boldsymbol{d}_{ijk}{}^{\rm p}\boldsymbol{\sigma}_{jk} + {}^{\rm p}\boldsymbol{k}_{ij}\boldsymbol{E}_j, \tag{8}$$

where superscript p denotes the piezoelectric phase; $^{\rm p}\varepsilon_{ij}$ and $^{\rm p}\boldsymbol{\sigma}_{kl}$ are the strain tensor of the piezoelectric phase and the stress tensor of the piezoelectric phase respectively; $\boldsymbol{E}_k$ is the electric field; $\boldsymbol{D}_i$ is the electric displacement vector; $^{\rm p}\boldsymbol{S}_{ijkl}$, $^{\rm p}\boldsymbol{d}_{kij}$, and $^{\rm p}\boldsymbol{k}_{ij}$ are the compliance tensor, piezoelectric coefficient, dielectric tensor.

The constitutive of the ferrite phase can be written as follows:

$$^{\mathrm{m}}\varepsilon_{ij} = {}^{\mathrm{m}}\boldsymbol{S}_{ijkl}{}^{\mathrm{m}}\boldsymbol{\sigma}_{kl} + {}^{\mathrm{m}}\boldsymbol{q}_{kij}\boldsymbol{H}_k, \tag{9}$$

$$\boldsymbol{B}_i = {}^{\mathrm{m}}\boldsymbol{q}_{kij}{}^{\mathrm{m}}\boldsymbol{\sigma}_{kl} + \boldsymbol{\mu}_{ij}\boldsymbol{H}_j, \tag{10}$$

where superscript m denotes the ferrite phase; $^{\mathrm{m}}\varepsilon_{ij}$ and $^{\mathrm{m}}\boldsymbol{\sigma}_{kl}$ represent the strain and stress tensor respectively; $\boldsymbol{H}_k$ is the external magnetic field; $\boldsymbol{B}_i$ is the intensity of magnetization; $^{\mathrm{m}}\boldsymbol{S}_{ijkl}$, $^{\mathrm{m}}\boldsymbol{q}_{kij}$, and $\boldsymbol{\mu}_{ij}$ are the compliance tensor, piezomagnetic tensor, and permeability tensor respectively.

When the length and width of the piezoelectric/ferrite layers are much larger than the thickness and the direction of the electric field is along the z axis, this issue can be seen as a plane stress problem in the x–y plane, and the stress–strain relation of the piezoelectric phase can be simplified into

$$^{\mathrm{p}}\varepsilon_{11} = (^{\mathrm{p}}S_{11} + {}^{\mathrm{p}}S_{12})^{\mathrm{p}}\sigma_{11} + {}^{\mathrm{p}}d_{13}E_3, \tag{11}$$

$$^{\mathrm{p}}\varepsilon_{22} = (^{\mathrm{p}}S_{12} + {}^{\mathrm{p}}S_{11})^{\mathrm{p}}\sigma_{11} + {}^{\mathrm{p}}d_{13}E_3. \tag{12}$$

Ignoring the external magnetic field, we obtain the stress–strain relation of the ferrite phase as follows:

$$^{\mathrm{m}}\varepsilon_{11} = (^{\mathrm{m}}S_{11} + {}^{\mathrm{m}}S_{12})^{\mathrm{m}}\sigma_{11}, \tag{13}$$

$$^{\mathrm{m}}\varepsilon_{22} = (^{\mathrm{m}}S_{12} + {}^{\mathrm{m}}S_{11})^{\mathrm{m}}\sigma_{11}. \tag{14}$$

Considering the fact that the four boundaries of the laminated structure are all free, the resultant stress on the boundaries is zero. Meanwhile, taking into account the fact that the laminated structure is bonded, the transmission of strain between two layers can be described by coupling coefficient k, and the boundary conditions can be written as follows:

$$\begin{aligned} &^{\mathrm{m}}\sigma_{11}t_{\mathrm{m}} + {}^{\mathrm{p}}\sigma_{11}t_{\mathrm{p}} = 0, \quad {}^{\mathrm{m}}\sigma_{22}t_{\mathrm{m}} + {}^{\mathrm{p}}\sigma_{22}t_{\mathrm{p}} = 0, \\ &^{\mathrm{p}}\varepsilon_{11} = {}^{\mathrm{p}}\varepsilon_{22} = k^{\mathrm{m}}\varepsilon_{11} = k^{\mathrm{m}}\varepsilon_{22}. \end{aligned} \tag{15}$$

Combining Eq. (11) with Eq. (15), we can obtain the expression of the stress inside ferrite, induced by the applied external electric field along the z axis, as follows:

$$\begin{aligned} ^{\mathrm{m}}\sigma_{11} &= {}^{\mathrm{m}}\sigma_{22} = \frac{^{\mathrm{p}}d_{31}t_{\mathrm{p}}E_3}{t_{\mathrm{m}}(^{\mathrm{p}}S_{11} + {}^{\mathrm{p}}S_{12}) + kt_{\mathrm{p}}(^{\mathrm{m}}S_{11} + {}^{\mathrm{m}}S_{12})} \\ &= e_{\mathrm{m}}E_3. \end{aligned} \tag{16}$$

It can be obviously seen from Eq. (16) that the stress inside ferrite, caused by the external electric field, is the in-plane stress. Therefore, considering the fact that the stress is along the x axis in Eq. (6), that is, $\alpha = 90^\circ$, and the directions of magnetization and magnetic field are the same, we obtain

$$\begin{aligned} \left(\frac{\omega}{\gamma}\right)^2 &= \left[-3\frac{\lambda\sigma}{M}\cos 2\beta - 4\pi M\cos 2\beta + H\right] \\ &\times \left[3\frac{\lambda\sigma}{M}\sin^2\beta - 4\pi M\cos^2\beta + H\right]. \end{aligned} \tag{17}$$

If the direction of the magnetic field is along the x axis, that is, $\beta = 90^\circ$, the magnetization and magnetostriction are both along the x direction. The influence of the stress along the x axis on the FMR frequency shift is dominant, in contrast, the stress along the y axis can be ignored. So equation (17) can be deduced as follows:

$$\left(\frac{\omega}{\gamma}\right)^2 = \left(H + 4\pi M + \frac{3\lambda e_{\mathrm{m}}E_3}{M}\right)\left(H + \frac{3\lambda e_{\mathrm{m}}E_3}{M}\right). \tag{18}$$

If the direction of the magnetic field is along the z axis, that is, $\beta = 0^\circ$, the effect of the stress along the x axis on the FMR frequency shift should be exactly the same as that along the y axis. So equation (17) can be deduced as follows:

$$\left(\frac{\omega}{\gamma}\right)^2 = \left(H - 4\pi M - \frac{6\lambda e_{\mathrm{m}}E_3}{M}\right)(H - 4\pi M). \tag{19}$$

If there is no external electric field, the in-plane stress is zero. Therefore, when the direction of the magnetic field is along the x axis or the z axis, equations (18) and (19) can be respectively simplified into

$$\begin{cases} \omega = \gamma[H(H + 4\pi M)]^{1/2}, \\ \omega = \gamma(H - 4\pi M). \end{cases} \tag{20}$$

The above formulas are the classical FMR frequency expressions, the same as that given in Ref. [20].

According to Eqs. (18) and (19), we can find from this model that the influence of the electric field on the FMR frequency is nonlinear. However, the electric tunability will be approximately linear when the electric field is small. In fact, according to the experimental results,[21] when the electric field ranges from -10 kV/cm to 10 kV/cm, the strain caused by it is linear. The model works well in this situation. When the electric field is beyond this range, the strain caused by the electric field becomes a butterfly-shape curve and cannot be described by the linear piezoelectric constitutive. Therefore, the model does not work in this situation.

3. Results and discussion

Next, we verify the reliability of this model through experimental results. Taking the laminated ME structure in Ref. [20] for example, YIG is chosen as the ferrite and PZT as the piezoelectric material. The material parameters are as follows: $\lambda = -1.4 \times 10^{-6}$, $t_{\mathrm{m}} = 15$ μm, $^{\mathrm{m}}S_{11} = 4.8 \times 10^{-12}$ m^2/N, $^{\mathrm{m}}S_{12} = 1.4 \times 10^{-12}$ m^2/N, $^{\mathrm{p}}d_{31} = -243 \times 10^{-12}$ m/V, $^{\mathrm{p}}S_{11} = 17 \times 10^{-12}$ m^2/N, $^{\mathrm{p}}S_{12} = -5.8 \times 10^{-12}$ m^2/N, $t_{\mathrm{p}} = 0.5$ mm, $\gamma = 2.8$ MHz/Oe, $4\pi M = 1750$ Oe, parallel magnetic field (magnetic field along the x axis) $H_{\parallel} = 1070$ kOe, and perpendicular magnetic field $H_{\perp} = 3530$ kOe. The shifts of the FMR frequency tuned by an electric field are shown in Fig. 2. It can be seen from this figure that the

results predicted by this model are in quantitative agreement with experimental results. Meanwhile, it can qualitatively describe the phenomenon that the FMR frequency increases as the electric field increases from −10 kV/m to 10 kV/m under a parallel magnetic field while it decreases under a perpendicular magnetic field.

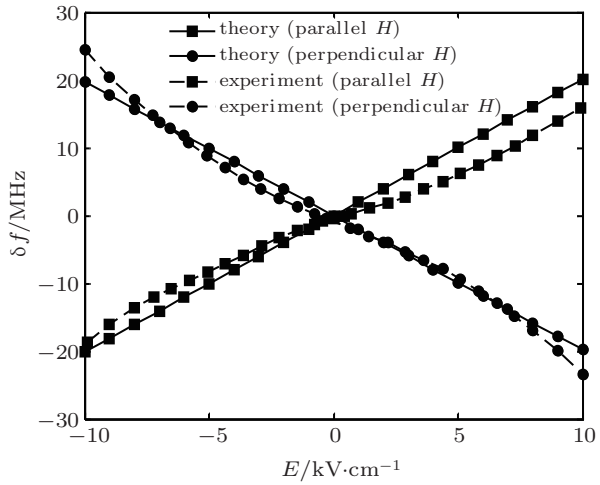


Fig. 2. Variations of FMR frequency shift with electric field of the YIG/PZT layers.

According to the YIG/PMN-PT laminated structure in Ref. [22], the parameters of PMN-PT are as follows: ${}^{\mathrm{p}}d_{31} = -600\times10^{-12}$ m/V, ${}^{\mathrm{p}}S_{11} = 23\times10^{-12}$ m^2/N, ${}^{\mathrm{p}}S_{12} = -8.3\times10^{-12}$ m^2/N, and $t_{\mathrm{p}} = 0.5$ mm. Electric tuning FMR frequency shifts predicted by this model are shown in Fig. 3. A comparison between the experimental and the predicted results shows clearly that this model can also predict the FMR frequency shift.

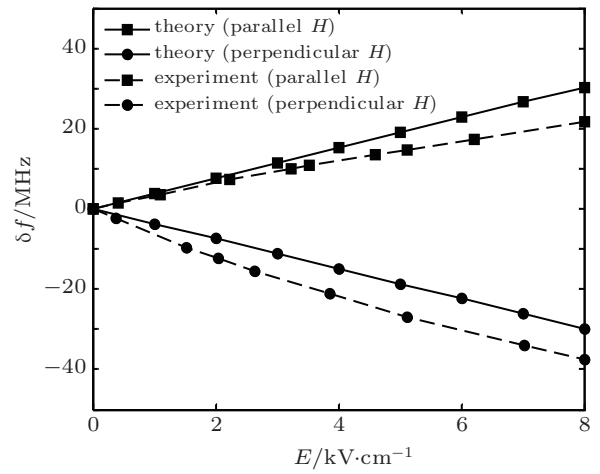


Fig. 3. Variations of FMR frequency shift with electric field of the YIG/PMN-PT layers.

When the electric field is 0, the stress produced by the piezoelectric layer is obviously 0. Therefore, equation (17) can be simplified into

$$\omega = \gamma\sqrt{(H-4\pi M\cos 2\beta)(H-4\pi M\cos^2\beta)}.$$

As can be seen from this expression, when $H < 4\pi M\cos 2\beta$ or $H < 4\pi M\cos^2\beta$, the value of the expression inside the radical sign will be smaller than 0. In this case, the solution of the formula is meaningless in physics. When $0 < \beta < \pi/2$, $\cos 2\beta < \cos^2\beta$ always holds. So, when $H > 4\pi M\cos^2\beta$, $(H-4\pi M\cos 2\beta)(H-4\pi M\cos^2\beta)$ is always larger than 0, and when $H \le 4\pi M\cos^2\beta$, ω will be meaningless in physics. According to this formula, under different magnetic fields, the variations of FMR frequency with the angle β in YIG/PZT layers are shown in Fig. 4. $\beta = 0^\circ$ represents the magnetic field perpendicular to the film surface, and $\beta = 90^\circ$ denotes the magnetic field parallel to the film surface. Then we can obtain that the FMR frequency increases monotonically when the direction of the magnetic field is changed from perpendicular to parallel to the film surface and $H > 4\pi M$, the maximum amplitude increases as high as 7 GHz. When $H < 4\pi M$, the expression is meaningless in physics when $\beta < \arccos(\sqrt{H/4\pi M})$, but the FMR frequency increases monotonically when $\beta > \arccos(\sqrt{H/4\pi M})$.

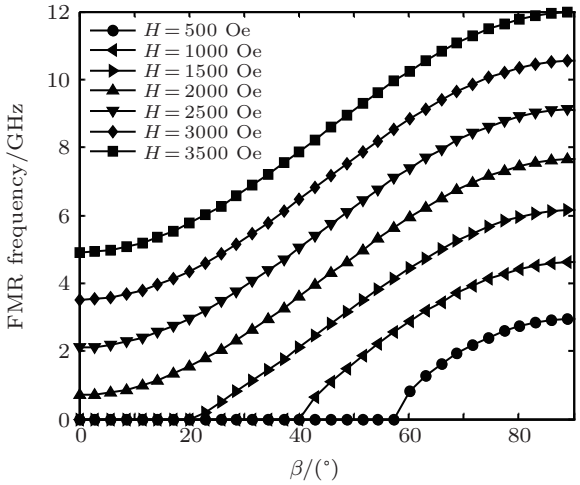


Fig. 4. Variations of FMR frequency with angle β in YIG/PZT layer under different magnetic fields.

As is well known, the increasing of the electric field induces the increase of the FMR frequency when applying a parallel magnetic field, while the FMR frequency decreases with the increasing of the electric field when applying a perpendicular magnetic field. So we justifiably believe that the electric field tunability will not hold any more when the angle between the external magnetic field and the z axis reaches a certain critical value. Next, we set the magnetic field to be $H = 3000$ Oe and keep it unchanged. We take the FMR frequency, when $E = 0$, as the reference value. The variations of FMR frequency shift with the angle β under different electric fields are shown in Fig. 5. As can be seen from the figure, the variation of the electric field does not have any effect on a FMR frequency shift when $\beta \approx 33.5^\circ$. As the electric field increases, the FMR frequency shift will decrease when $\beta < 33.5^\circ$ and increase when $\beta > 33.5^\circ$. That is, $\beta \approx 33.5^\circ$ is a critical angle

for an electric field to tune an FMR frequency shift under this condition. Meanwhile, we can reach the conclusion that electric tunability will become largest when applying a magnetic field along the x axis direction ($\beta = 90°$).

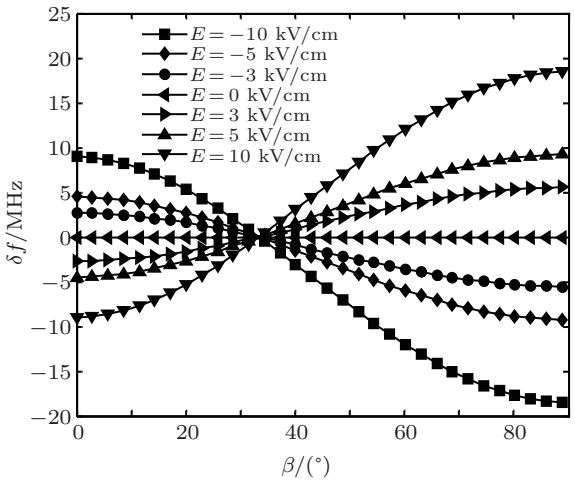


Fig. 5. Variations of FMR frequency shift with angle β in YIG/PZT layer under different electric fields.

4. Conclusions

In this paper, we focus on the phenomenon of electric tuning FMR frequency shift in a laminated ME structure, and propose a theoretical model for FMR frequency shift under the stress according to Smith and Beljerss theory. Afterwards, we obtain an expression of the stress when applying an electric field to the piezoelectric phase. Finally, combining these two formulas, we establish a theoretical model of the electric tuning FMR frequency shift. From this model, we can find that the angle between the external magnetic field and the film surface has a great influence on both the FMR frequency shift and electric tunability. Meanwhile, we find that there exists a critical angle, whose value is determined by the external magnetic field and the ferrite material property. The electric field tunability does not hold any more when the angle reaches a critical angle and becomes maximum when a parallel magnetic field is applied. These results are very important for future high-frequency signal processing devices.

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