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Resonant Magnetoelectric Effect with Strongly Nonlinear Magneto-Elastic Coupling in Magnetoelectric Laminate Composites

Ying Xiao[1], **Hao-Miao Zhou**[1,2,3], **Xiao-Wei Ou**[1] **and Chao Li**[1]

Abstract: Considering the complex strongly nonlinear coupling characteristic of the magnetostrictive strain and magnetization under the excitation of the bias magnetic field and the pre-stress in the giant magnetostrictive material, this paper adopts the nonlinear magnetostrictive constitutive model and the equivalent circuit method to establish a strongly nonlinear resonant magnetoelectric (ME) effect theoretical model for the ME laminate composites compounding by the giant magnetostrictive material and the piezoelectric material. For the L-T mode magnetostrictive/piezoelectric/magnetostrictive (MPM) ME laminate, the predicted results coincide well with the experiment results of the resonant frequency and the resonant ME field coefficient varying with the external magnetic field when the pre-stress degenerates to zero in our model. The agreement indicates the proposed theoretical model validity. On the basis, we use the theoretical model to forecast the varying characteristic of the resonant ME field coefficient and the resonant frequency effect under the influence of the different bias magnetic field and the pre-stress in ME laminate composites. And we also predict that the resonant ME coefficient and the resonant frequency appear “reversal” with the pre-stress increasing. After that, the influence of the different volume ratio on the ME effect and resonant frequency is analyzed. Particularly, a resonant frequency value not influenced by the volume ratio with increasing bias magnetic or pre-stress occurs. This research can provide theory basis for improving the resonant ME conversion performance and for controlling the resonant frequency under the excitation of the bias conditions (i.e. the bias magnetic field and the pre-stress) for the ME devices (i.e. sensor, transducer, microwave device and so on).

Keywords: Equivalent circuit method, Nonlinear constitutive model, Magneto-

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[1] College of Information Engineering, China Jiliang University, Hangzhou, 310018, P. R. China

[2] Institute of Applied Mechanics, School of Aeronautics and Astronautics, Zhejiang University, Hangzhou 310027, P. R. China

[3] Corresponding author: Tel: 086-0571-87676215; email: zhouhm@cjlu.edu.cn

electric laminate composites, Magneto-elastic coupling effect, Resonant magneto-electric effect

1 Introduction

Because of ferromagnetism and ferroelectricity, magnetoelectric (ME) materials have a comprising prospect of application in many areas including the fabrication of novel information storage devices, sensors and tunable microwave devices, and so on. Therefore, the research of ME materials has caused wide public concern [Spaldin and Fiebig (2005); Nan et al. (2008); Eerenstein et al. (2006)]. Compared with the single-phase ME materials and the granulated ME composites, the laminate composites consisting of the giant magnetostrictive and piezoelectric materials show better ME coupling effect [Spaldin and Fiebig (2005); Nan et al. (2008)]. Meanwhile, the experimental results indicate that the ME coefficient of the ME laminate in resonant status could be one to two orders of magnitude greater than the static or low frequency ones[Eerenstein et al. (2006); Dong et al. (2006a, 2003); Wan et al. (2005); Yu et al. (2005); Bichurin et al. (2003)], so the research about the ME laminate composites becomes a hot research topic [Bichurin et al. (2003); Wang et al. (2008); Popov et al. (2008); Bian et al. (2009b,c); Dong and Guo (2009); Zheng and Liu (2005)]. However, large numbers of studies [Moffet et al. (1991); Zhou et al. (2009, 2008); Zhou and Zhou (2007); Zhou et al. (2006, 2007)] found that there is very complex nonlinear magneto-elastic coupling effect in the giant magnetostrictive materials. And the magnetostrictive strain and the magnetization curves show not strongly nonlinear coupling characteristics varying with the external magnetic field (e.g. initial nonlinear, saturation and so on), but also show obvious difference (the phenomenon of “reversal”) with the different pre-stress levels. So, for the ME laminate composites based on the giant magnetostrictive materials, the magneto-electric coupling effect taken the pre-stress as a medium will equally have the complex nonlinear relationship with the external bias magnetic field and the pre-stress. And the explicit research of the complex magneto-elastic-electric coupling effect will be helpful to the optimal design of the ME material, popularization and application of devices.

Currently, the research about the ME laminate composites is focused on the experimental measurements and theoretical analysis of ME effect at static and low frequency [Nan et al. (2008)]. However, because the ME coupling effect in resonant state will be of much more significant application value compared with that at static and low frequency, the research about the resonant ME coupling effect has caused great concerns in recent years [Dong et al. (2003); Wan et al. (2005); Bichurin et al. (2003); Wang et al. (2008); Popov et al. (2008); Bian et al. (2009b,c); Dong and Guo (2009); Israel et al. (2009); Srinivasan et al. (2005);

Dong and Zhai (2008); Dong et al. (2006b); Bian et al. (2009a); Yang et al. (2008a); Zhou et al. (2011a); Yang et al. (2008b); Zhou et al. (2011b); Yang et al. (2006); Xing et al. (2006)]. For example, Wan et al. (2005) attained the first bending resonance mode's maximum bending resonant ME field coefficient of $14.6V/cmOe$. Popov *et al* (2008) reported the resonant ME field coefficient could reach $90V/cmOe$ under both the effects of DC and AC magnetic fields. Bian et al. (2009b) reported the resonant ME field coefficient of the L-T mode (i.e. the magnetostrictive layer is magnetized along the longitudinal direction and the piezoelectric layer is polarized in its thickness direction) based on high quality factor materials could reach $381.875V/cmOe$. The theoretical research of the resonant ME effect is mainly based on the equivalent circuit method proposed by Dong et al. (2006a). They first reported a high ME voltage coefficient phenomenon of the magnetostrictive/piezoelectric laminate composites at the resonance frequency in 2003 [Dong et al. (2003)]. Next, they conducted many studies on resonant ME coefficients of various ME laminate composites structures [Dong et al. (2006a, 2003); Dong and Guo (2009); Dong and Zhai (2008); Xing et al. (2006)]. But for the resonant ME voltage coefficient of the L-T mode MPM laminate composites, the predicted theoretical result is one order higher than that of the experimental result done by Dong [Dong and Zhai (2008)]. Bian *et al* have done some theoretical and experimental studies of the resonant ME effect on the different ME laminate composites, considering the mechanical loss, elastic substrates, high effective quality factor ferromagnetic material[Eerenstein et al. (2006); Bian et al. (2009b,c,a); Yang et al. (2008a); Zhou et al. (2011a); Yang et al. (2008b); Zhou et al. (2011b)]. And they used the equivalent circuit method to propose a theoretical expression for the resonant ME field coefficient [Bian et al. (2009c)]. But when this expression is used in the MPM laminate composites, the predicted result is almost twice that of the experimental result done by Bian and Yang et al. [Bian et al. (2009a); Yang et al. (2008b)]. For the L-T mode MPM laminate composites, Zhou et al.[Zhou et al. (2011a)] analyzed the influence of the bias magnetic field on the magneto-elastic-electric coupling effect. Even though the predicted result and experimental result had a good coincidence, the pre-stress was assumed directly as zero when using nonlinear magnetostrictive constitutive model. Thus, it can not be used to analyze the complex influence of the pre-stress on the ME coupling effect. A numerical model of the ME coupling analysis, which is proposed by Zhou et al.[Zhou et al. (2011b)] using the nonlinear magnetostrictive constitutive model, could predict the static ME coefficient very well. But due to using one-dimension model, when the resonant ME effect occurs, the predicted resonant frequency is about one third of the experimental result. Based on the above analysis, it is necessary to adopt the equivalent circuit method to study the characteristics of resonant ME effect of the ME laminate composites under the influence of strongly nonlinear magneto-elastic coupling.

In allusion to the L-T mode magnetostrictive/piezoelectric/magnetostrictive (MPM) symmetrical ME laminate, this paper adopts the nonlinear magnetostrictive constitutive model, which can describe the complex magneto-elastic coupling effect completely, to obtain the expressions of dynamic materials constant varying with the pre-stress and the bias magnetic field. Combined with the equivalent circuit method, we establish a resonant ME coupling effect theoretical model of the ME laminate composites under the complex magneto-elastic coupling conditions (i.e. the external magnetic field and the pre-stress). When the pre-stress degenerates to zero in this model, the predicted results of resonant ME coefficient and resonant frequency coincide well with the experiment results. After that, the established model is used to predict the dependence characteristic of resonant frequency and resonant ME field coefficient with the external magnetic field under different pre-stress, and it is also used to predict the influence of the volume ratio of piezoelectric and piezomagnetic materials on the resonant ME field coefficient and resonant frequency.

2 Model Establishment

L-T mode MPM laminate composites is the sandwich structure consisting of the piezoelectric layer and two magnetostrictive layers bonded by epoxy resins, and the piezoelectric layer is clipped between two magnetostrictive layers. When the external magnetic field H_3 (Which consists of alternative magnetic field H_{ac} and bias magnetic field H_{bias}) is applied along the longitudinal direction, the magnetostrictive layers will be magnetized along the longitudinal direction (L mode), and the piezoelectric layer will be polarized in thickness direction (T mode). The magnetostrictive layers and piezoelectric layer of laminate composites are both rectangle; the cross-sectional area are A_1 and A_2; the thickness are t_m and t_p; the length and width are expressed as l and w respectively.

2.1 Magneto-elastic Coupling Characteristic of Giant Magnetostrictive Materials

The constitutive model of giant magnetostrictive material is nonlinear coupling [Zheng and Liu (2005)], the magnetization curves and magnetostrictive curves not only have saturation and slight hysteresis features, but also are remarkably affected by the axial pre-stress. If the pre-stress exerted on a giant magnetostrictive rod along the axial direction, the increase of the magnetization and the magnetostrictive strain will slower obviously in the region of the low and moderate fields with an increasing magnetic fields. But when the magnetic field is so high that the magnetization becomes magnetic saturation, the maximum magnetostrictive strain has an obvious increasing compared with no pre-stress [Zheng and Liu (2005)]. Moffet et al. (1991) measured the piezomagnetic coefficient d_{33m} and (constant field) the elastic compliance coefficient s_{33m} and the permeability μ_{33} under eight sets of different compressive pre-stresses and magnetic bias fields, it is found that there is complex function dependence on both the given bias conditions and the varying amplitude of the stress and the magnetic field. Furthermore, the Young's modulus of Terfenol-D rod is not a constant, but a complex function changing with the stress and the magnetic field amplitude, that is ΔE effect. So, this paper chooses the nonlinear constitutive model of magnetostrictive materials, which can capture complex nonlinear magneto-elastic coupling characteristic completely, and then obtains the formulas of the piezomagnetic coefficient and the elastic compliance coefficient varying with the bias conditions. After that, the equivalent circuit method is used to establish the theoretical model of resonant ME effect under nonlinear magneto-elastic coupling of the ME laminate composites.

We chose Terfenol-D as the magnetostrictive layers of laminate composites, and used the nonlinear magnetostrictive constitutive model of Terfenol-D proposed by Zheng et al.[Zheng and Liu (2005)] to attain the formulas of dynamic materials constant. The constitutive model of giant magnetostrictive material is listed as follows:

$$\varepsilon = \frac{\sigma}{E_s} + \begin{cases} \lambda_s \tanh(\frac{\sigma}{\sigma_s}) + \left[1 - \tanh(\frac{\sigma}{\sigma_s})\right] \frac{\lambda_s}{M_s^2} M^2 & \frac{\sigma}{\sigma_s} \geq 0 \\ \frac{\lambda_s}{2} \tanh(\frac{2\sigma}{\sigma_s}) \pounds \left[1 - \frac{1}{2} \tanh(\frac{2\sigma}{\sigma_s})\right] \frac{\lambda_s}{M_s^2} M^2 & \frac{\sigma}{\sigma_s} < 0 \end{cases} \tag{1}$$

$$H = \frac{1}{k} f^{-1}(\frac{M}{M_s}) - \begin{cases} 2\left\{\sigma - \sigma_s \ln\left[\cosh(\frac{\sigma}{\sigma_s})\right]\right\} \frac{\lambda_s}{\mu_0 M_s^2} M & \frac{\sigma}{\sigma_s} \geq 0 \\ 2\left\{\sigma - \frac{\sigma_s}{4} \ln\left[\cosh(\frac{2\sigma}{\sigma_s})\right]\right\} \frac{\lambda_s}{\mu_0 M_s^2} M & \frac{\sigma}{\sigma_s} < 0 \end{cases} \tag{2}$$

The function expressions in Eq.1 and Eq.2 are $\sigma_s = \lambda_s E_s E_0 / (E_s - E_0)$, $f(x) = \coth(x) - 1/x$ and $k = 3\chi_m / M_s$ respectively. Where ε is the strain; σ is the pre-stress; λ_s is the saturation magnetostrictive coefficient; M is the magnetization; M_s is the saturation magnetization; E_0 is the initial Young's modulus; E_s is the intrinsic (or saturation) Young's modulus; σ_s is the saturation pre-stress; χ_m is the linear magnetic susceptibility; $\mu_0 = 4\pi \times 10^{-7}\,\mathrm{H/m}$ is the vacuum permeability; H is the external magnetic field (Which consists of alternative magnetic field H_{ac} and bias magnetic field H_{bias}) along the longitudinal direction of Terfenol-D.

Based on the definition of the elastic compliance coefficient s_{33m} and the piezomagnetic coefficient d_{33m} proposed by Moffet et al. (1991), substituting the nonlinear constitutive model of Eq.1 and Eq.2 into, we can calculate the partial derivatives and obtain the expressions of s_{33m} and d_{33m} affected by the external excitation magnetic field and the pre-stress respectively as follows:

$$s_{33m} = \frac{\partial\varepsilon(\sigma,H)}{\partial\sigma} = \frac{1}{E_s} + \begin{cases} \frac{\lambda_s}{\sigma_s}\sec h^2(\frac{\sigma}{\sigma_s}) + 2[1-\tanh(\frac{\sigma}{\sigma_s})]\frac{\lambda_s}{M_s^2}M\frac{\partial M}{\partial\sigma} + \frac{\lambda_s M^2}{\sigma_s M_s^2}\sec h^2(\frac{\sigma}{\sigma_s}) & \frac{\sigma}{\sigma_s} > 0 \\ \frac{\lambda_s}{\sigma_s}\sec h^2(\frac{2\sigma}{\sigma_s}) + 2[1-\frac{1}{2}\tanh(\frac{2\sigma}{\sigma_s})]\frac{\lambda_s}{M_s^2}M\frac{\partial M}{\partial\sigma} + \frac{\lambda_s M^2}{\sigma_s M_s^2}\sec h^2(\frac{2\sigma}{\sigma_s}) & \frac{\sigma}{\sigma_s} < 0 \end{cases} \tag{3}$$

$$d_{33m} = \frac{\partial\varepsilon(\sigma,H)}{\partial H} = \begin{cases} \left[1-\tanh(\frac{\sigma}{\sigma_s})\right]\frac{2M\lambda_s}{M_s^2}\frac{\partial M}{\partial H} & \frac{\sigma}{\sigma_s} \geq 0 \\ \left[1-\frac{1}{2}\tanh(\frac{2\sigma}{\sigma_s})\right]\frac{2M\lambda_s}{M_s^2}\frac{\partial M}{\partial H} & \frac{\sigma}{\sigma_s} < 0 \end{cases} \tag{4}$$

From the above formulas, it is found that there are two partial derivatives $\frac{\partial M}{\partial\sigma}$ and $\frac{\partial M}{\partial H}$ of the magnetization to the pre-stress and the magnetic field respectively in Eq.3 and Eq.4; the independent variable (magnetization M) exists in Eq.2; and there is an inverse function in Eq.2. Therefore, in order to get the expressions of s_{33m} and d_{33m} we firstly revert the implicit function expression $f^{-1}(\frac{M}{M_s})$ in Eq.2, then calculate the partial derivative of two sides of explicit function $\frac{M}{M_s} = f(\sigma,H)$ for σ and Hrespectively to get explicit expressions of $\frac{\partial M(\sigma,H)}{\partial\sigma}$ and $\frac{\partial M(\sigma,H)}{\partial H}$. Substituting $\frac{\partial M(\sigma,H)}{\partial\sigma}$ and $\frac{\partial M(\sigma,H)}{\partial H}$ into Eq.3 and Eq.4, we can obtain the elastic compliance coefficient s_{33m} and the piezomagnetic coefficient d_{33m} as follows:

$$s_{33m} = (E_{33m})^{-1} = \begin{cases} \frac{1}{E_s} + \frac{\lambda_s(1+\frac{M^2}{M_s^2})}{\sigma_s\cosh^2(\frac{\sigma}{\sigma_s})} + \frac{4k\lambda_s^2M^2[1-\tanh(\frac{\sigma}{\sigma_s})]^2}{\frac{\mu_0 M_s^3}{-\csc h^2 M_1 + M_1^{-2}} - 2k\lambda_s M_s^2\left\{\sigma-\sigma_s\ln\left[\cosh(\frac{\sigma}{\sigma_s})\right]\right\}}, & \frac{\sigma}{\sigma_s} > 0 \\ \frac{1}{E_s} + \frac{\lambda_s(1+\frac{M^2}{M_s^2})}{\sigma_s\cosh^2(\frac{2\sigma}{\sigma_s})} + \frac{4k\lambda_s^2M^2[1-\frac{1}{2}\tanh(\frac{2\sigma}{\sigma_s})]^2}{\frac{\mu_0 M_s^3}{-\csc h^2 M_2 + M_2^{-2}} - 2k\lambda_s M_s^2\left\{\sigma-\frac{\sigma_s}{4}\ln\left[\cosh(\frac{2\sigma}{\sigma_s})\right]\right\}}, & \frac{\sigma}{\sigma_s} < 0 \end{cases} \tag{5}$$

$$d_{33m} = \begin{cases} \frac{2k\lambda_s M\left[1-\tanh(\frac{\sigma}{\sigma_s})\right]}{\frac{M_s}{-\csc h^2 M_1 + M_1^{-2}} - 2k\left\{\sigma-\sigma_s\ln\left[\cosh(\frac{\sigma}{\sigma_s})\right]\right\}\frac{\lambda_s M}{\mu_0}}, & \frac{\sigma}{\sigma_s} \geq 0 \\ \frac{2k\lambda_s M\left[1-\frac{1}{2}\tanh(\frac{2\sigma}{\sigma_s})\right]}{\frac{M_s}{-\csc h^2 M_2 + M_2^{-2}} - 2k\left\{\sigma-\frac{\sigma_s}{4}\ln\left[\cosh(\frac{2\sigma}{\sigma_s})\right]\right\}\frac{\lambda_s M}{\mu_0}}, & \frac{\sigma}{\sigma_s} < 0 \end{cases} \tag{6}$$

M_1 and M_2 in Eq.5 and Eq.6 are respectively as follows:

$$\begin{cases} M_1 = kH + 2k\left\{\sigma - \sigma_s\ln\left[\cosh(\frac{\sigma}{\sigma_s})\right]\right\}\frac{\lambda_s M}{\mu_0 M_s^2}, & \frac{\sigma}{\sigma_s} \geq 0 \\ M_2 = kH + 2k\left\{\sigma - \frac{\sigma_s}{4}\ln\left[\cosh(\frac{2\sigma}{\sigma_s})\right]\right\}\frac{\lambda_s M}{\mu_0 M_s^2}, & \frac{\sigma}{\sigma_s} < 0 \end{cases} \tag{7}$$

Eq.5 and Eq.6 are theoretical expressions of the elastic compliance coefficient s_{33m} and the piezomagnetic coefficient d_{33m}, which are established based on the existing nonlinear constitutive model and can be used to reflect the influence of the bias magnetic field and the pre-stress.

2.2 Equivalent Circuit Model of Magnetoelectric Laminate Composites

Based on the magnetostrictive and piezoelectric constitutive equations, we used the equivalent circuit method to process the coupling effect of magneto-elastic-electric of the MPM laminate composites. The cross-sectional areas of the magnetostrictive layers and piezoelectric layer are A_1 and A_2; the thicknesses are t_m and t_p; the length and width are expressed as l and w; the densities are ρ_m and ρ_p respectively. $A = 2A_1 + A_2 = (2t_m + t_p)w = tw$ is the cross-sectional area of the laminate; $n = 2A_1/A = 2t_m/t$ is the thickness ratio of the magnetostrictive layers; $\overline{\rho} = (2\rho_m A_1 + \rho_p A_2)/A$ is the average density of the laminate;$\dot{u}_1$ and $\dot{u}_2$ are the displacement speeds at $z = 0$ and $z = l$ in laminate composites respectively. According to the equivalent circuit method, the magneto-elastic-electric equivalent circuit for the L-T mode MPM laminate composites can be obtained by Yang et al.[Yang et al. (2006)], shown in Fig. 1.

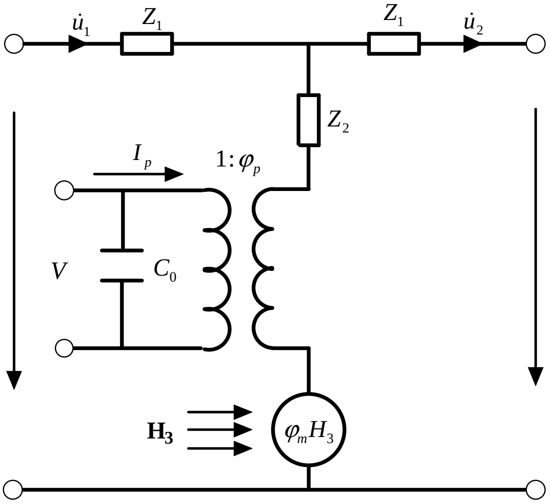

Figure 1: Magneto-elastic-electric equivalent circuit

In Fig. 1, V is the induced voltage; F_1, F_2 are the face forces on the two ends ($z = 0$ and $z = l$) of the laminate; $Z_1 = j\overline{\rho}vA\tan\frac{kl}{2}$, $Z_2 = \frac{\overline{\rho}vA}{j\sin kl}$ are the mechanical impedances of the laminate; $C_0 = \frac{lw\overline{\varepsilon}_{33}}{t_p}$ is the static clamped capacitance of the piezoelectric layer; $\phi_m = \frac{2A_1 d_{33m}}{s_{33m}}$, $\phi_p = \frac{wd_{31p}}{s_{11}^E}$ are the magneto-elastic of the magnetostrictive layer and elasto-electric coupling factors of the piezoelectric layer respectively. Here $k = \omega/v$,$v^2 = (\frac{n}{s_{33\mathrm{m}}} + \frac{1-n}{s_{11}^E})/\bar{\rho}$, $\overline{\varepsilon_{33}} = \varepsilon_{33}[1-(k_{31}^p)^2]$, $k_{33}^m = \frac{d_{33m}}{\sqrt{s_{33m}\mu_{33}}}$, $k_{31}^p = \frac{d_{31p}}{\sqrt{s_{11}^E\varepsilon_{33}}}$. ω is the angle frequency; v is the mean sound velocity; s_{33m}, d_{33m}, μ_{33} are the elastic compliance coefficient of the magnetostrictive material under the constant magnetic field H, the longitudinal piezomagnetic coefficient and the magnetic permeability under the constant stress, respectively; s_{11}^E,d_{31p}, ε_{33} are the piezoelectric elastic compliance of the piezoelectric material under the constant electric field E, the transverse piezoelectric coefficient, and the permittivity under the constant stress, respectively; k_{33}^m is the magneto-mechanical coupling coefficient of the magnetostrictive material; k_{31}^p is the electro-mechanical coupling coefficient of the piezoelectric material.

When the ME laminate composites vibrate freely ($F_1 = F_2 = 0$), the circuit in Fig.1 can be simplified to get the equivalent mechanical impedance of the ME laminate Z and $Z = Z_1//Z_1 + Z_2 = -\frac{1}{2}j\bar{\rho}vA\tan(\frac{kl}{2})$. When the external excitation magnetic field works around the laminate composites' natural frequency, mechanical resonance will happen to the laminate. Taking dissipation into consideration in the ME conversion and then processing the mechanical impedance Z in resonant status, the equivalent circuit can be further simplified to a series *RLC* circuit given by Dong et al. [Dong et al. (2006a)] shown in Fig. 2.

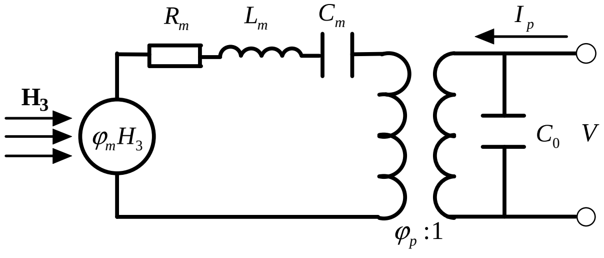

Figure 2: Resonant ME equivalent circuit

Here R_m, L_m and C_m are the motional mechanical resistance, inductance and capacitance of the laminate respectively.

$$L_m = \frac{\pi Z_0}{8\omega_r}, \quad C_m = \frac{8}{\pi Z_0 \omega_r}, \quad R_m = \frac{\omega_r L_m}{Q_{mech}} = \frac{\pi Z_0}{8Q_{mech}}, \quad \omega_r^2 = \frac{1}{L_m C_m}, \quad Z_0 = \bar{\rho} vA.$$

When there is no AC magnetic field input, using the impedance conversion method, the ME equivalent circuit can be simplified to the standard equivalent circuit of a piezoelectric vibrator. We analyzed the simplified equivalent circuit based on the resonance principle of the piezoelectric vibrator and impedance conversion method, and then we could obtain the relational expressions among R_m, L_m, C_m, the series resonance frequency f_s, parallel resonance frequency f_p of the ME laminate composites, the effective electro-mechanical coupling factor k_{eff} and the effective mechanical Q-factor at the series resonance frequency Q_{mech} [IEEE (1987, 1966)] as follows:

$$k_{eff}^2 = \frac{f_p^2 - f_s^2}{f_p^2} = \frac{\varphi_p^2}{\varphi_p^2 + C_0/C_m} \tag{8}$$

$$Q_{mech} = \frac{\omega_r L_m}{R_m} = \frac{1}{\omega_r C_m R_m} \tag{9}$$

As the effective electro-mechanical coupling factor k_{eff} and the effective mechanical Q-factor Q_{mech} influence the Magneto-elastic-electric coupling effect in the ME laminate composites and the magnetoelastic damping in the magnetostrictive layers directly. Both of them influence the resonant ME effect obviously.

2.3 Resonant ME Effect in ME Laminate Composites

Analyzed the equivalent mechanical impedance Z shown in Fig.1, when the imaginary part of the equivalent mechanical impedance is assumed as zero (that is $Im(Z) = 0$), combined with $Z = Z_1//Z_1 + Z_2 = -\frac{1}{2}j\overline{\rho}vA\tan(\frac{kl}{2})$, the resonant frequency equation of the ME laminate $\overline{\rho}vA\tan(\frac{kl}{2}) = 0$ can be obtained. Simplified this equation, here $k = \omega/v$, $\omega_r = 2\pi f_s$, we can get the one-dimensional vertical resonant frequency f_{s1} shown in Eq.10.

$$f_{s1} = \frac{1}{2l}\sqrt{\frac{\frac{n}{s_{33m}(H,\sigma)} + \frac{1-n}{s_{11}^E}}{\overline{\rho}}} \tag{10}$$

From Eq.10, we know that the unique factor of the elastic compliance coefficient s_{33m} is affected by the external magnetic field H and the pre-stress σ. The formula of Eq.10 indicates that the bias magnetic field H_{bias} and the pre-stress affect the one-dimensional vertical resonant frequency of ME laminate composites by influencing the elastic compliance coefficient s_{33m} of the giant magnetostrictive materials.

Combined with Fig.2 and analyzed the resonant ME effect of the ME laminate composites based on the resonance principle, Ohm's law and impedance conversion method, we can get the resonant ME voltage coefficient expression considering the mechanical loss as follows:

$$\alpha_V^r = \left|\frac{dV}{dH_{ac}}\right| = \left|\frac{\frac{\phi_m\phi_p}{j\omega_r C_0}}{R_m + \frac{\phi_p^2}{j\omega_r C_0}}\right| = \left|\frac{\phi_m\phi_p}{j\omega_r C_0 R_m + \phi_p^2}\right| \tag{11}$$

Substituting Eq.8 and Eq.9 into Eq.11, we get the following equation:

$$\alpha_V^r = \left| \phi_m \phi_p Q_{mech} \frac{C_m}{C_0 + \phi_p^2 Q_{mech} C_m} \right| = \frac{\phi_m Q_{mech} k_{eff}^2}{\phi_p \left(1 - k_{eff}^2 + Q_{mech} k_{eff}^2\right)} \tag{12}$$

In Eq.12, k_{eff} is the effective electro-mechanical coupling factor of ME laminate composites, calculated by Bian et al. (2009a) as follows:

$$k_{eff}^2 = \frac{f_p^2 - f_s^2}{f_p^2} = \frac{8}{\pi^2} \frac{(1-n)s_{33m}}{(1-n)s_{33m} + ns_{11}^E} \frac{(k_{31}^p)^2}{1-(k_{31}^p)^2} \tag{13}$$

Substituting the effective electro-mechanical coupling factor (Eq.13) into Eq.12, we can obtain the resonant ME voltage coefficient as follows:

$$\alpha_V^r = \frac{8Q_{mech} n(1-n) t d_{33m}(H,\sigma) d_{31p}}{\varepsilon_{33}\{n\pi^2[1-(k_{31}^p)^2] s_{11}^E + (1-n)[\pi^2 + (8Q_{mech} - 8 - \pi^2)(k_{31}^p)^2] s_{33m}(H,\sigma)\}} \tag{14}$$

Because the relation between the resonant ME field coefficient and the resonant ME voltage coefficient is $\alpha_E^r = \alpha_V^r / t_p$, the resonant ME field coefficient is given as:

$$\alpha_E^r = \frac{8Q_{mech} n d_{33m}(H,\sigma) d_{31p}}{\varepsilon_{33}\{n\pi^2[1-(k_{31}^p)^2] s_{11}^E + (1-n)[\pi^2 + (8Q_{mech} - 8 - \pi^2)(k_{31}^p)^2] s_{33m}(H,\sigma)\}} \tag{15}$$

From Eq.14 and Eq.15, we know that the elastic compliance coefficient s_{33m} and the piezomagnetic coefficient d_{33m} of the giant magnetostrictive materials appear complex relation varying with the external magnetic field and the pre-stress. It is for this reason that the complex magneto-elastic coupling results in the complex strong nonlinear characteristic for the ME voltage coefficient and the ME field coefficient.

3 Validity of the Model

We first analyzed the validity of the expressions of materials constant derived by the nonlinear magnetostrictive constitutive model, and then compared the theoretical result of Young's modulus predicted by Zheng et al.[Zheng and Liu (2005)], keeping the chosen basic parameters of the materials same as Zheng's research [Zheng and Liu (2005)]. Because of Eq.5 being the expression of the elastic compliance coefficient, its inverse of one dimension case is just the Young's modulus. Under

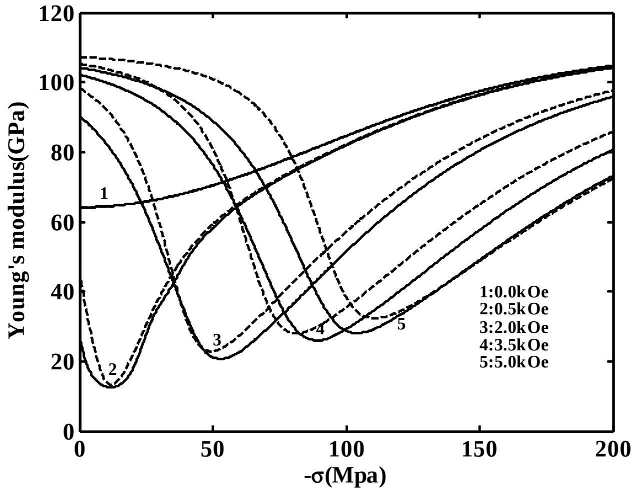

Figure 3: Young's modulus vs compressive stress under various magnetic bias fields (the real lines are results in this paper, the dashed lines are results done by Zheng [Zheng and Liu (2005)])

the different bias magnetic field, the comparison curves of the Young's modulus calculated by Eq.5 and by Zheng [Zheng and Liu (2005)] varying with the pre-stress are shown in Fig.3. In Fig.3, the dashed lines show the results calculated by Zheng[Zheng and Liu (2005)], and the real lines show the results in this paper. From the figure, we can find that the dashed line and real line almost coincide with each other completely when the bias magnetic field H_{bias} is zero, and that the curves show a monotonic increasing tendency with the pre-stress increasing. When the bias magnetic field H_{bias} is not zero, both of them show a good coincidence qualitatively. The curves of Young's modulus E_{33m} decrease fast firstly and then increase slowly with the pre-stress increasing. However, the curves of Young's modulus have a little difference on quantity. Meanwhile, under the condition of the different bias magnetic field and pre-stress, we can find from the Fig.3 that the value of Young's modulus can be close to 90GPa at most, which can reflect the ΔE effect of the giant magnetostrictive material well. Based on above analysis, we can obtain that the explicit expressions of dynamic material constant derived in this paper are believable.

Next, in order to quantitatively prove the validity of the equivalent circuit theory of the resonant ME effect in this paper, we chose the experimental results done by Wang [Wang et al. (2008)] and theory results predicted by Yang [Yang et al. (2008b)] of resonant ME effect in MPM magnetoelectric laminate composites consisted of Terfenol-D and PMN-PT as reference. Because the ME effect of the ME laminate composites was measured in experiment without considering exerting the compressive stress in the magnetostritive material, the model of the resonant frequency and the ME field coefficient proposed in this paper includes the influence of the pre-stress. Thus, firstly the pre-stress was assumed as zero in the model, and then let the theoretical model degenerate and be same state as experiment. Here, the physical dimension of *Terfenol-D* and *PMN-PT* is same as one in experiment, and they are $12mm \times 6mm \times 1mm$ and $12mm \times 6mm \times 1mm$ respectively. Magnetostrictive layers are magnetized along the longitudinal direction, and the piezoelectric layer is polarized in its thickness direction. The material parameters are shown in the appendix. If a weak external AC magnetic field ($H_{ac} < 5Oe$ as usual) is applied in the ME laminate studied in this paper, the amplitude of H_{ac} is much smaller than that of the bias magnetic field H_{bias} obviously. Therefore, H_{ac} is ignored and the external magnetic field H is replaced by H_{bias} when discussing the influence of bias magnetic field on the resonant ME effect. The comparative figure, which is composed by the theoretical result of the formula of resonant frequency of the ME laminate composites derived in this paper (the pre-stress is zero in Eq.10) and the experimental result done by Wang[Wang et al. (2008)], is shown in Fig.4. In the figure, the comparative results show a good coincidence qualitatively. The curves decrease firstly, and then increase with the external bias magnetic field increasing. On quantity, the predicted result in this paper is smaller than the experiment result done by Wang et al. [Wang et al. (2008)]. As for the reason of the difference between the predicted result and the experimental result, it could be residual stress existed in magnetostritive phase during fabrication of the ME laminate composites in experiment. According to Eq.5, it is easy to know that the residual stress will result in changing of the elastic compliance coefficient, and also result in changing of resonant frequency. So, there is a little difference on quantity between the predicted result by the theoretical model and the experimental result done by Wang [Wang et al. (2008)]. As well, the comparative figure, composed by the result of the ME field coefficient predicted by the theoretical model (Eq.15) as the pre-stress being zero, the theory result predicted by Yang [Yang et al. (2008b)] and the experimental result done by Wang [Wang et al. (2008)], is shown in Fig.5. From the figure, we can find that the theoretical result in this paper and the experimental result show a good coincidence, while the theory result predicted by Yang [Yang et al. (2008b)] is nearly three times bigger than the result in this paper and the experiment result done by Wang [Wang et al. (2008)]. So the predicted result in this paper is more valid compared with the result predicted by Yang [Yang et al. (2008b)]. When the bias magnetic field satisfies $H_{bias} < 0.35kOe$, the predicted theoretical result of the ME field coefficient in this paper is smaller than the experimental result; while the bias magnetic field satisfies $H_{bias} > 0.35kOe$, the predicted result in this paper is a

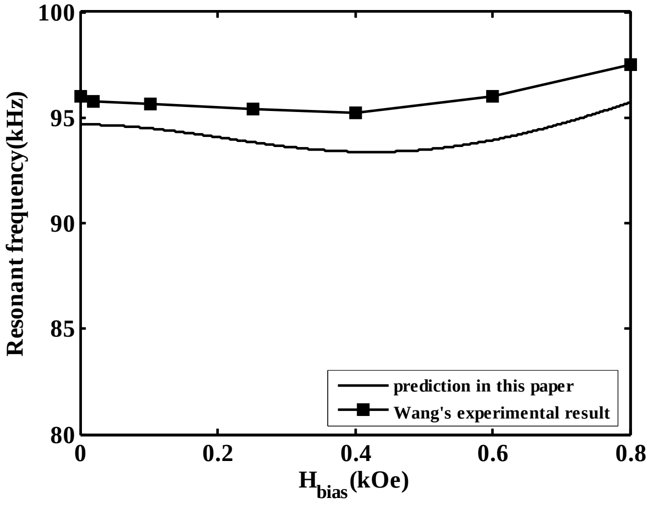

Figure 4: Comparison of one-dimensional vertical resonant frequency f_{s1}

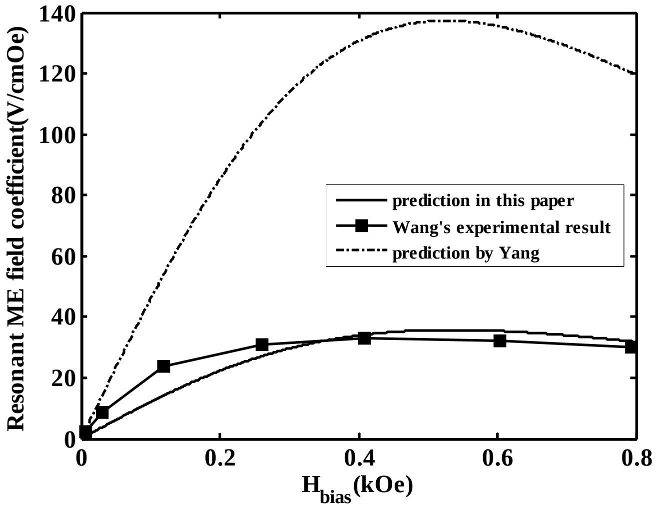

Figure 5: Comparison of one-dimensional vertical resonant ME field coefficient α_E^r

litter larger than the experimental result.

Compared with the previous model considering only the influence of the bias magnetic field, the model of resonant ME effect in this paper can take into account the ME effect in the case of complex magneto-elastic coupling. So, we can use the model to predict the characteristics of strongly nonlinear magneto-elastic-electric

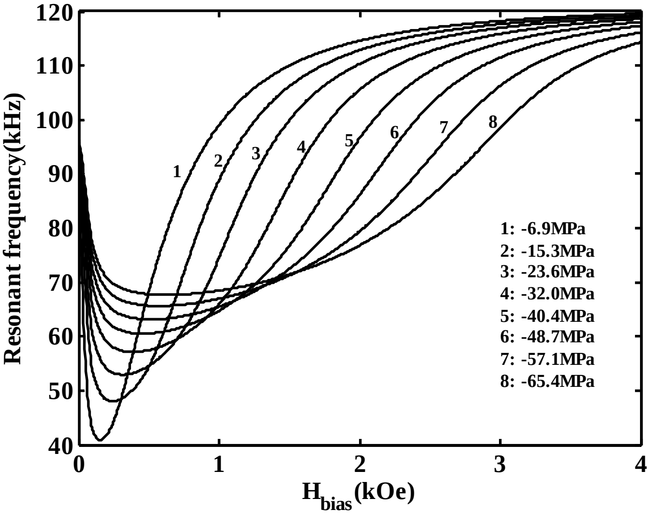

Figure 6: One-dimensional vertical resonant frequency f_{s1} changing with H_{bias}

coupling of the ME material under the influence of the external magnetic field and the pre-stress. By Eq.10, we can get the relationship between the one-dimensional vertical resonant frequency f_{s1} and the bias magnetic field under the different pre-stress (shown in Fig.6.). In the figure, with the bias magnetic field increasing, the one-dimensional vertical resonant frequency decreases fast firstly, and then increases, and finally tends to be a steady value under the different pre-stress. When the pre-stress is smaller, it is found that the resonant frequency changes dramatically varying with the bias magnetic field, that is, the resonant frequency drops to the minimum value quickly, then increases fast and tends to be a stable value. While the pre-stress is larger, the resonant frequency changes relatively gently with the bias magnetic field. At the same time we can find that there is a phenomenon of “reversal” for resonant frequency varying with the pre-stress, that is, when the bias magnetic field is smaller than about 0.35kOe, the resonant frequency increases gradually with the pre-stress increasing; when the bias magnetic field is larger than 1.5kOe, the resonant frequency decreases gradually with the pre-stress increasing. From the formula of the one-dimensional vertical resonant frequency in Eq.10, we know that only one unique factor, the elastic compliance coefficient s_{33m}, is influenced by the external bias magnetic and pre-stress simultaneously. As a result, the bias magnetic and pre-stress influence the resonant frequency of the ME laminates by influencing the elastic compliance coefficient s_{33m} of Terfenol-D.

Similarly, we can use the resonant ME field coefficient (Eq.15) to predict the re-

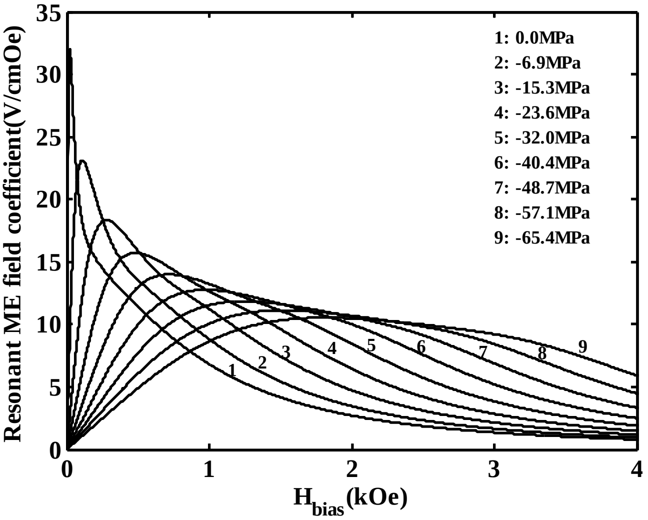

Figure 7: One-dimensional vertical resonant ME field coefficient α_E^r changing with H_{bias}

lationship of the one-dimensional vertical resonant ME field coefficient of the ME laminate composites under different pre-stress levels and the bias magnetic field $\alpha_E^r - H_{bias}$, shown in Fig.7. In Fig.7, the curves of resonant ME field coefficient increase firstly, and then decrease, and finally tend to zero varying with the bias magnetic field under different pre-stress levels. And we can also find from the figure that the phenomenon of "reversal" exists because of the influence of the pre-stress on the resonant ME field coefficient. When the external bias magnetic field is smaller, the resonant ME field coefficient decreases quickly with the increasing pre-stress. Whereas, the external bias magnetic field is larger, the resonant ME field coefficient shows increasing tendency with the pre-stress increasing. Therefore, the influence tendency of the pre-stress on the resonant ME field coefficient appears "reversal". In the formula of the ME field coefficient (Eq.15), due to the different pre-stress and bias magnetic field, the elastic compliance coefficient and the piezomagnetic coefficient of the giant magnetostrictive materials appear complex nonlinear variation, which then results in the complex change of the resonant ME field coefficient of the ME laminate composites. However, most of the recent researches considered only the influence of the bias magnetic field on the ME field coefficient and ignored the influence of the pre-stress on the ME field coefficient. It can be seen From Fig.6 and Fig.7 that the pre-stress also greatly influences the ME coefficient of the ME laminate composites. So, it is possible to realize the optimal design of the resonant frequency and the ME coefficient of the ME laminate composites by adjusting the different pre-stress levels.

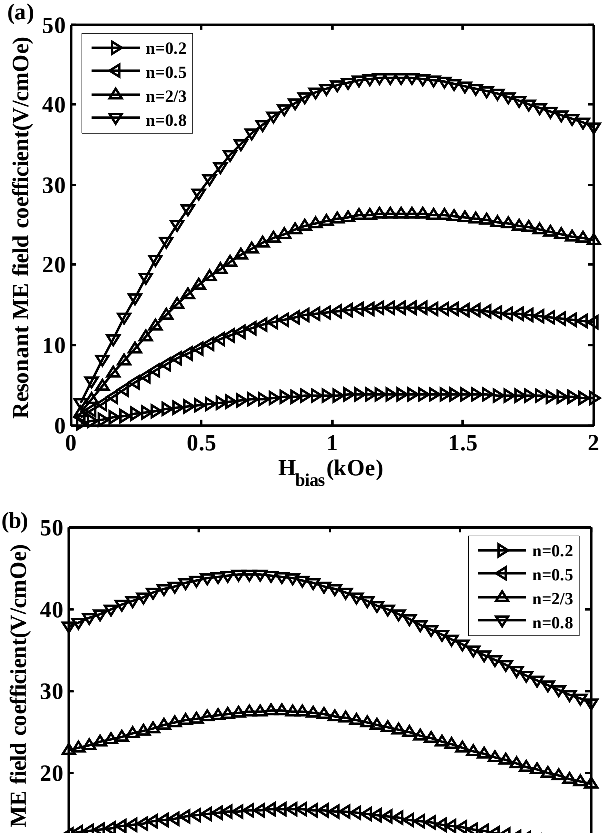

Figure 8: (a) Calculated values of resonant ME field coefficient under different Terfenol-D thickness ratio n, changing with H_{bias}; (b) Calculated values of resonant ME field coefficient under different Terfenol-D thickness ratio n, changing with σ

The thickness ratio of Terfenol-D also influences the resonant ME field coefficient. Based on the theoretical model in this paper, we can predict the influence of the thickness ratio on the resonant ME field coefficient for a given external magnetic field or the pre-stress, shown in Fig.8. It can be seen from Fig.8 (a), for a given pre-

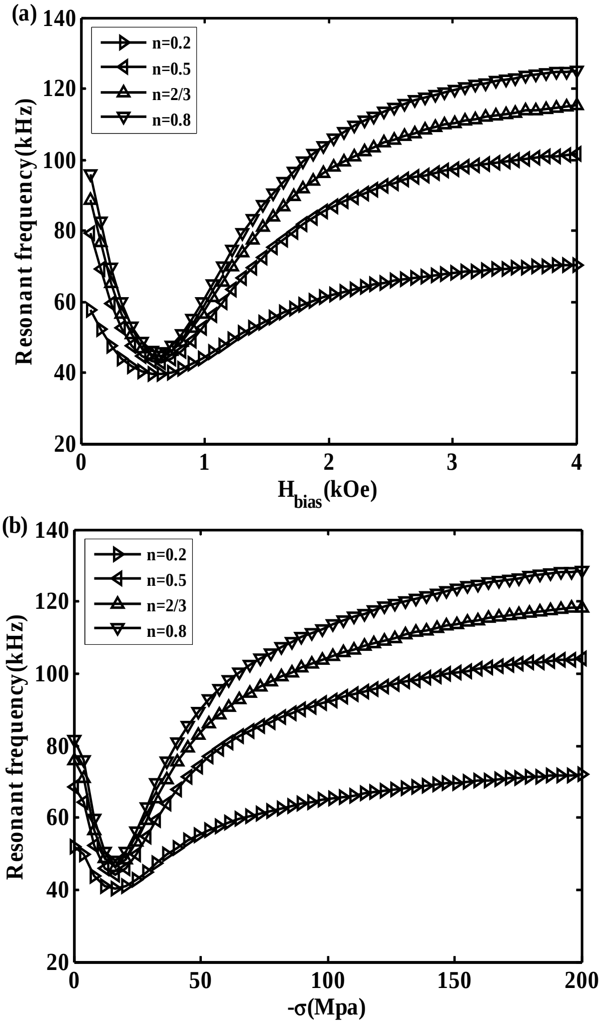

Figure 9: (a) Calculated values of resonant frequency under different Terfenol-D thickness ratio n, changing with H_{bias}; (b) Calculated values of resonant frequency under different Terfenol-D thickness ratio n, changing with σ

stress (i.e. -13Mpa), that the resonant ME field coefficient increases monotonically with the increasing thickness ratio n of Terfenol-D. For a given bias magnetic field (i.e. 1kOe), the curves of the resonant ME field coefficient varying with the pre-stress under the different thickness ratio are shown in Fig.8 (b). Similarly, it can be seen from Fig.8 (b) that the resonant ME field coefficient becomes larger with the thickness ratio increasing, and the bigger thickness ratio, the greater influence of the pre-stress on the ME field coefficient.

For a given pre-stress(13MPa), the influence of the thickness ratio of Terfenol-D on the resonant frequency is shown in Fig.9. In Fig.9, the resonant frequency increases with the thickness ratio increasing of Terfenol-D, but the minimum of the resonant frequency keeps almost unchangeable. Whereas the bias magnetic field or the pre-stress is continually changing, the resonant frequency fast drops to 40-50kHz, and then increases gradually. When the bias magnetic field continues to increase, the resonant frequency increases gently and tends to be a certain value, which can also be verified by Fig.6. In Fig.9 (a), for a given pre-stress (i.e. 13MPa), the resonant frequency decreases quickly for $H_{bias} < 0.62kOe$ and increases gently for $H_{bias} > 0.62kOe$ with the bias magnetic field increasing. Similarly, in Fig.9 (b), for a given bias magnetic field (i.e. 1kOe), the resonant frequency decreases quickly for $\sigma < 13MPa$ and increases gently for $\sigma > 13MPa$ with the pre-stress increasing continually. We can find from Fig.9 that the minimum resonant frequency about 46kHz exists which is not influenced by the thickness ratio of Terfenol-D.

4 Conclusion

Combined with the nonlinear constitutive relationship of magnetostrictive materials, this paper adopted equivalent circuit method to establish a complex resonant ME effect theoretical model of the ME laminate composites under the influence of the pre-stress and the external bias magnetic field. When the pre-stress was degenerated to zero in this model, the results showed that the predicted resonant frequency and resonant ME coefficient of the degenerated model coincided well with the experiment results on quality and quantity. After that, we used the theoretical model to predict the ME field coefficient and resonant frequency varying with the bias magnetic field under the influence of the different pre-stress. We predicted theoretically for the first time that the phenomenon of "reversal" arise from the influence of the pre-stress on the ME field coefficient and resonant frequency. Therefore, the research in this paper can provide theory basis for predicting the ME characteristics of the ME laminate composites and for preparation of the ME devices with good performance under the complex magneto-elastic coupling. On the basis, we further analyzed the influence of the thickness ratio of magnetostrictive materials on the resonant ME coefficient and resonant frequency. The thickness ratio has a great influence on the ME coefficient and resonant frequency. Thereinto, a special value of resonant frequency exists, which is not influenced by changing the thickness ratio.

Appendix

Parameters of *Terfenol-D*:

$E_0 = 6.5 \times 10^{10} Pa$; $E_s = 11 \times 10^{10} Pa$; $\lambda_s = 1 \times 10^{-3}$; $\chi_m = 40$;

$\mu_m M_s = 4T$; $\rho_m = 9230 kg/m^3$; $\mu_{33}/\mu_0 = 5$; $Q_{TD} \approx 10$;

Parameters of *PMN-PT:*

$d_{31p} = -2645 \times 10^{-12} C/N$; $s_{11}^E = 141.3 \times 10^{-12} m^2/N$; $\rho_p = 8100 kg/m^3$;

$\varepsilon_{33}/\varepsilon_0 = 6200$; $k_{31}^p = 0.95$; $Q_{PMN-PT} = 31$.

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References

Bian L X, Wen Y M, Li P, (2009): Analysis of magneto-mechano-electmronic coupling factors in magneto strictive-piezoelectric laminated composite. *Acta Phys. Sin*, 58:4205-5209 (in Chinese).

Bian L X, Wen Y M, Li P, Gao Q L, Zhu Y, Yu M, (2009): Magnetoelectric Performances in Composite of Piezoelectric Ceramic and Ferromagnetic Constant-Elasticity Alloy. *IEEE Sensors Journal*, 9:1620-1226.

Bian L X, Wen Y M, Li P, (2009): Composite Magnetoelectric Transducer of Terfenol-D and Pb(Zr,Ti)O_3 Plates Bonded on an Elastic Substrate. *IEEE Sensors Journal* 9:45-50.

Bichurin M I, Filippov D A, Petrov V M, Laletin V M, Srinivasan G, (2003): Resonance Magnetoelectric Effects in Layered Magnetostrictive-Piezoelectric Composites. *Phys. Rev. B*, 68:132408.

Dong S X, Li J F, Viehland D, (2006): Magnetoelectric coupling, efficiency, and voltage gain effect in piezoelectric-piezomagnetic laminate composites. *Journal of Materials Science*, 41: 97–106.

Dong S X, Zhai J Y, Li J F, Viehland D, (2006): Magnetoelectric effect in Terfenol-D/ Pb(Zr,TiO)$_3$/μ-metal laminate composites. *Appl. Phys. Lett.*, 89:122903.

Dong S X, Li J F, Cheng J R, Viehland D, (2003): Enhanced magnetoelectric effects in laminate composites of Terfenol-D/Pb(Zr,Ti)O$_3$ under resonant drive. *Appl. Phys. Lett.*, 83:4812.

Dong S X, Guo M S, (2009): A Resonance-Bending Mode Magnetoelectric Coupling Equivalent Circuit. *IEEE Transactions on Ultrasonics, Ferroelectrics, and Frequency Control*, 56:2578-2586.

Dong S X, Zhai J Y, (2008): Equivalent circuit method for static and dynamic analysis of magnetoelectric laminated composites. *Chinese Science Bulletin*, 53:2113-2123.

Eerenstein W, Mathur N D, et al, (2006): Multiferroic and magnetoelectric materials. *Nature*, 442(17):759-765.

IEEE (1987): Standard on Piezoelectricity. IEEE Std 176-1987.

IEEE (1966): Standard definitions and methods of measurement for piezoelectric vibrators. IEEE 177-1966.

Israel C, Petrov V M, Srinivasan G, Mathur N D, (2009): Magnetically tuned mechanical resonances in magnetoelectric multilayer capacitors. *Applied Physics Letters*, 95: 072505.

Moffet M B, Clark A E, Wun-Fogle M, Linberg J, Teter J P, McLaughlin E A, (1991): Characterization of Terfenol-D for magnetostrictive transducers. *J. Acoust. Soc. Am.*, 89 (3): 1448-1455.

Nan C W, Bichurin M I, Dong S X, Viehland D, Srinivasan G, (2008): Multiferroic magnetoelectric composites: Historical perspective, status, and future directions. *Journal of Applied Physics*, 103:031101.

Popov C, Chang H, Record P M, Abraham E, Whatmore R W, Huang Z, (2008): Direct and converse magnetoelectric effect at resonant frequency in laminar piezoelectric- magnetostrictive composite. *J Electroceram* 20:53–58.

Spaldin N A, Fiebig M, (2005): The Renaissance of magnetoelectric multiferroics. *Science,* 309: 391-392.

Srinivasan G, Vreugd C P De, Laletin V M, Paddubnaya N, Bichurin M I, Petrov V M, Filippov D A, (2005): Resonant Magnetoelectric Coupling in Trilayers of Ferromagnetic Alloys and Piezoelectric Lead Zirconate Titanate: The Influence of Bias Magnetic Field. *Phys. Rev. B*, 71:184423.

Wan J G, Li Z Y, Wang Y, Zeng M, Wang G H, Liu J M, (2005): Strong flexural resonant magnetoelectric effect in Terfenol-D/epoxy-Pb(Zr,Ti)O3 bilayer. *Appl. Phys. Lett.*, 86:202504.

Wang Y J, Siu Wing Or, Helen Lai Wa Chan, Zhao X Y, Luo H S, (2008): Enhanced magnetoelectric effect in longitudinal-transverse mode Terfenol-D/ $Pb(Mg_{1/3}Nb_{2/3})O_3$–$PbTiO_3$ laminate composites with optimal crystal cut. *Journal of Applied Physics*, 103:124511.

Xing Z P, Dong S X, Zhai J Y, Yan L, Li J F, Viehland D, (2006): Resonant bending mode of Terfenol-D /steel/$Pb(Zr,Ti)O_3$ magnetoelectric laminate composites. *Appl. Phys. Lett.*, 89:112911.

Yang C H, Wen Y M, Li P, Bian L X, (2008): Influence of bias magnetic field on magnetoelectric effect of magnetostrictive/elastic/piezoelectric laminated composite. *Acta. Phys. Sin*, 57:7292-7297 (in Chinese).

Yang F, Wen Y M, Li P, Bian L X, (2008): Resonant magnetoelectric response of magnetostrictive /piezoelectric laminate composite in consideration of losses. *Sensors and Actuators A*, 141:129–135.

Yang F, Wen Y M, Li P, Zheng M, (2006): Magnetoelectric Response of Magnetostrictive/Piezoelectric/Magnetostrictive Laminate Composite. IEEE International Conference on Information Acquisition, Weihai, China, 1010-1014.

Yu H, Zeng M, Wang Y, Wan J G, Liu J M, (2005): Magnetoelectric resonance-bandwidth broadening of Terfenol-D/epoxy-$Pb(Zr,Ti)O_3$ bilayers in parallel and series connections. *Appl. Phys. Lett.*, 86:032508.

Zheng X J, Liu X E, (2005): A nonlinear constitutive model for Terfenol-D rods. *Journal of Applied Physics*, 97:053901.

Zhou H M, Zhou Y H, (2007): Vibration Suppression of Laminated Composite Beams Using Actuators Of Giant Magnetostrictive Materials. *Smart Materials and Structure*, 16: 198-206.

Zhou H M, Li C, Xuan L M, Wei J, Zhao J X, (2011): Equivalent circuit method research of resonant magnetoelectric characteristic in magnetoelectric laminate composites using nonlinear magnetostrictive constitutive model. *Smart Materials and Structure*s, 20: 035001.

Zhou H M, Xuan L M, Li C, Wei J, (2011): Numerical simulation of nonlinear magnetic–mechanical–electric coupling effect in laminated magnetoelectric composites. *J. Magn. Magn. Mater.*, 232(22): 2802-2807.

Zhou H M, Zhou Y H, Zheng X J, Wei J, (2009): A general magnetoelastic coupling theory of deformable magnetized medium including magnetic forces and magnetostriction effects. *CMC: Computers, Materials, & Continua*, 12(3): 237-249.

Zhou H M, Zhou Y H, Zheng X J, (2008): A General Theoretical Model of Magnetostrictive Constitutive Relationships for Soft Ferromagnetic Material Rods. *Journal of Applied Physics*, 104: 023907.

Zhou H M, Zhou Y H, Zheng X J, (2007): Numerical Simulation of Nonlinear Dynamic Responses of Beams Laminated with Giant Magnetostrictive Actuators. *CMC: Computers, Materials, & Continua*, 6(3):201-211.

Zhou H M, Zheng X J, Zhou Y H, (2006) : Vibration Active Control of Nonlinear Giant Magnetostrictive Actuators. *Smart Materials and Structure*. 15:792-798.