Resonance magnetoelectric characteristics of Terfenol-D/Pb(Zr_{0.52}Ti_{0.48})O_3/Ni asymmetric three layered composites

Ying Liu¹,², Chao Yang¹, Peng Zhou¹,², Gopalan Srinivasan², Yajun Qi¹, Kun Liang¹, Tianjin Zhang¹ and D A Filippov ³,⁴

¹Department of Materials Science and Engineering, Hubei University, Wuhan 430062, P.R. China
²Physics Department, Oakland University, Rochester, Michigan 48309, USA
³Yaroslav-the-Wise Novgorod State University, ul. B. St. Peterburgskaya, 41 173003, Velikiy Novgorod, Russia
⁴E-mail: Dmitry.Filippov@novsu.ru

Abstract. Three resonance modes, a first-order bending resonance mode and nonlinear and first-order longitudinal resonance modes, were observed in Terfenol-D/lead zirconate titanate(PZT)/Ni asymmetric magnetoelectric(ME) laminated composites. The ME outputs and resonance modes were tuned by the ratio of the thickness of the Ni negative magnetostrictive layer to the Terfenol-D positive magnetostrictive layer that of the (t_Ni/t_T). As (t_Ni/t_T) increased from 0.4 to 2.0, the strength of the first-order longitudinal resonance mode was decreased, while the first-order bending resonance was enhanced. The asymmetric threelayer composites exhibited a large ME coefficient of 250mV/(cm Oe) under a bias magnetic field of 400 Oe at quasi-static frequency (1 kHz). This work sheds some light on the design of ME composites with tunable ME resonance modes and multifrequency operation.

1. Introduction
Magnetoelastic (ME) composites are currently attracting increasing interest and have provoked a great number of research activities, driven by their potential applications in novel multifunctional devices such as sensors and transducers [1]. The direct ME effect, which describes the appearance of an electric polarization \( P \) upon applying a magnetic field \( H \), is usually evaluated by the magnetoelectric voltage coefficient (MEVC) \( \alpha_E \) [1, 2]. It is well known that the ME effect of composites is extrinsic, strongly depending on the intrinsic piezoelectric or piezomagnetic properties of constituents and coupling interaction across magnetic/piezoelectric interfaces.

Various properties of ME laminated composites have been optimized for their use in potential applications, such as varying the magnetostrictive and piezoelectric materials; optimizing the composite structure, shape, and size; changing interfacial bonding conditions; and modulating the operation mode of ME coupling [3–7]. The symmetric threelayered composite structure formed by sandwiching a piezoelectric layer between two identical magnetostrictive layers is a classical ME composite structure [8, 9]. Compared with a symmetric threelayered ME composite, an asymmetric threelayered ME composite can operate in not only the longitudinal vibration mode, but also the bending vibration mode because of the flexural deformation caused by the asymmetric stress distribution in the ME composite [10–12]. When the ME composite operates in the bending vibration...
mode, its resonance frequency will be much lower than that of other modes, which can markedly decrease the eddy current loss for the magnetostrictive phase. This is especially relevant for rare earth alloys with large magnetostriction, such as Terfenol-D, because it can result in inefficient ME energy conversion. Meanwhile, the composite size could be kept quite small and ME coupling is strong. However, the bending vibration mode usually appears with the longitudinal vibration mode at comparable resonance intensity [10–12].

In this paper, we design a new asymmetric three-layered composite that can be operated in both bending and longitudinal modes. The resonance strength of each mode can be tuned by the thickness ratio of the positive and negative magnetostrictive layer. The results illustrate the possibility of implementing asymmetric three-layered composites in multifunctional devices with multifrequency and multimode operation.

2. Methods

Figure 1 depicts a schematic diagram of the asymmetric three-layered composite structure. The piezoelectric Pb(Zr0.52Ti0.48)O3 (PZT) layer with dimensions of 20×5×1 mm was polarized along the thickness direction. The magnetostrictive orientation of the positive and negative magnetostrictive layers composed of Tb$_{1-x}$Dy$_x$Fe$_{2-y}$ (Terfenol-D) and Ni flakes with dimensions of 15×5×0.5 mm, respectively, was along the longitudinal direction. The Terfenol-D, PZT, and Ni plates were bonded together by epoxy adhesive. The quick-drying epoxy adhesive could cure rapidly and ensure good mechanical coupling and strong bonding between layers at room temperature. Bottom Ni layers with thicknesses of 0.2, 0.5, and 1 mm were used to give composites with thickness ratios of the positive Terfenol-D to the negative Ni magnetostrictive layer $t_{Ni}/t_T$ of 0.4, 1.0, and 2.0, respectively.

![Figure 1. Schematic diagram of the asymmetric Terfenol-D/PZT/Ni three-layered composite.](image)

The ME effects of the composites were measured by applying both bias constant ($H_{dc}$) and alternating ($\delta H$) magnetic fields parallel to the length direction. A shield cover was used to apply $H_{dc}$ as well as shield external signals. Small Helmholtz coils were used to excite an ac magnetic field with amplitude of 3 Oe. The output voltage signal was detected using a lock-in amplifier (Stanford, SR830). All measurements were carried out at room temperature. The MEVC $\alpha_{E,31}$ was calculated using the formula of $\alpha_{E,31} = \delta V/(t_{PZT}\delta H)$, where $\delta V$ is the voltage variation of PZT and $t_{PZT}$ is the thickness of the PZT layer.

3. Results and discussion

Unlike a symmetric three-layered ME composite, an asymmetric three-layered ME composite can exhibit not only the longitudinal vibration mode, but also the bending vibration mode because of the flexural deformation caused by its asymmetric stress distribution. Accordingly, asymmetric three-layered laminated composites were designed with Terfenol-D with positive magnetostriction as the surface layer and Ni with negative magnetostriction as the bottom layer, as shown in figure 1. Composites with different Ni layer thicknesses of 0.2, 0.5, and 1 mm were fabricated to modulate the bending vibration, giving samples with $t_{Ni}/t_T$ of 0.4, 1.0 and 2.0 respectively.
According to resonance theory, a bending resonance frequency is expected and can be calculated by the following formula [12, 13]:

$$n_{fb1} = \frac{\pi t}{4\sqrt{3}l} \bar{\rho} \beta_{n2},$$  \hspace{1cm} (1)

where $t$ is the total thickness of ME composite materials, $l$ is the length of ME composite materials, $\bar{\rho}$ is the average density of the sample, and $\beta_n = (n + 0.5)$, where $n$ is the order of flexural vibration ($n \geq 1$). $\bar{Y}$ is the average value of Young’s modules, which can be calculated according to the following formula:

$$\bar{Y} = \left(\frac{t_1^Y Y_Y t_1 + t_2^Y Y_Y t_2 + t_3^Y Y_Y t_3}{t_1 + t_2 + t_3}\right),$$  \hspace{1cm} (2)

in which $t_1^Y$, $t_2^Y$, and $t_3^Y$ are the thickness of PZT, Terfenol-D, and Ni layers, respectively; $Y_P = 1/s_{11}^P$, $Y_T = 1/s_{11}^T$, $Y_N = 1/s_{11}^N$ are the Young’s modules, and $s_{11}^P$, $s_{11}^T$, and $s_{11}^N$ are the elastic compliances of PZT, Terfenol-D, and Ni, respectively.

For the longitudinal mode, we can use the calculations in our previous work [14]. The constitutive equations for the magnetostrictive and piezoelectric phases are:

$$^pS_i = \frac{1}{^pY} ^pT_i + ^p d_{31}E_3,$$  \hspace{1cm} (3)

$$^T S_i = \frac{1}{^T Y} ^T T_i + ^T \lambda_i(H),$$  \hspace{1cm} (4)

$$^N S_i = \frac{1}{^N Y} ^N T_i + ^N \lambda_i(H),$$  \hspace{1cm} (5)

$$^P D_3 = ^p e_{33}^P E_3 + ^p d_{31}^P T_1,$$  \hspace{1cm} (6)

where $^pS_i$, $^T S_i$, $^N S_i$ are the strain tensor components of the PZT, Terfenol-D and Ni layers, $^pE_3$, $^P D_3$ are vector components of the electric field and electric displacement; $^pT_i$, $^T T_i$, $^N T_i$ are the stress tensor components of the piezoelectric and the magnetostrictive phases; $^p d_{31}$ is piezoelectric coefficients; $^T \lambda_i(H)$, $^N \lambda_i(H)$ are magnetostriction of Terfenol-D and Ni layers, respectively; and $^p e_{33}$ is the tensor components of permittivity. The field dependent magnetostriction of Terfenol-D and Ni layers are shown in figure 2.

**Figure 2.** Dependence of the longitudinal magnetostriction $\lambda_i$ of Ni and Terfenol-D on bias magnetic field $H_{dc}$.  

Magnetostriction is nonlinear function of ac and bias magnetic fields. We can get the following function by taking into account the first and second terms of series:

\[
\lambda_i(H) = q_{11}H_1 + q_{11}^{(2)}H_1^2,
\]

where 

\[
q_{11} = \left. \frac{\partial \lambda_i}{\partial H_1} \right|_{H_1=H_{dc}}, \quad q_{11}^{(2)} = \left. \frac{\partial^2 \lambda_i}{\partial H_1^2} \right|_{H_1=H_{dc}}
\]

are linear and nonlinear piezomagnetic coefficients, \(H_1\) is ac magnetic field with angular frequency \(\omega\). The first term in Eq.(7) give the deformation of magnetic with frequency \(\omega\) and second term give the deformation of magnetic with frequency \(2\omega\) \([15-18]\). These coefficients are also nonlinear functions vs magnetic field, the dependencies between field and piezomagnetic coefficients of Terfenol-D and Ni layers are shown in Fig. 3.

**Figure 3.** Dependence of the linear \(q_{11}\) and nonlinear \(q_{11}^{(2)}\) piezomagnetic coefficients of Ni and Terfenol-D on bias magnetic field \(H_{dc}\) for thickness \(t_{\text{Ni}} = 0.5\) mm and \(t_{T} = 0.5\) mm.

In figure 3, the linear and nonlinear piezomagnetic coefficients for Terfenol-D and Ni exhibit different signs. This leads to the opposite contribution to the value of ME effect for longitudinal mode.

Using approximation given by Equation (7), the Equation (4) and Equation (5) can be rewritten as:

\[
\tau S_i = \frac{1}{\tau Y} \tau T_i + \tau q_{11}H_1 + \tau q_{11}^{(2)}H_1^2,
\]

\[
\frac{N_S}{N} = \frac{1}{N_{\text{Y}}} N_{\tau T_1} + \frac{N_{\tau q_{11}H_1}}{N_{\tau q_{11}H_1}} + \frac{N_{\tau q_{11}^{(2)}H_1^2}}{N_{\tau q_{11}^{(2)}H_1^2}}
\]

The ac magnetic field with angular frequency \(\omega\) produces the deformation with angular frequencies \(\omega\) and \(2\omega\) in Terfenol-D and Ni layers. These deformations transmitted via mechanical interaction in PZT layer and electric field induced by piezoelectric effect. Since the layers are thin enough so that we could neglect the change in the displacements of the medium along the thickness of the layer, and assume that the displacements of the medium, the magnetic and the piezoelectric layer are equal, i.e. \(u_{\text{Y}} = u_{\text{T}} = u_{\text{Ni}} = u_1\). Therefore, in the first approximation the equation of motion for the medium has following form:

\[
\rho \frac{\partial^2 u_1}{\partial t^2} = \frac{\partial T_{11}}{\partial x_1},
\]
where \( \bar{\rho} = \left( \rho^t + N_i \rho^{Ni} + \rho^P t \right) \left( \rho^t + N_i t + P t \right) \), \( \bar{T} = \left( T^t + N_i T^Ni + P T^P t \right) \left( T^t + N_i t + P T^P t \right) \) are the average values of the density and stress tensor of the sample.

Using the method of calculations, which was presented in our previous work [14], we can obtain the MEVC as following expressions:

\[
\alpha_E = \frac{p Y^d d_{31}}{e_{33} \left( \gamma^t + N_i \gamma^{Ni} + P \gamma^P t \right)} \left[ \left( \gamma^t T^t \cdot \gamma^{q_{11}} + N_i \gamma^{Ni} t \cdot \gamma^{q_{11}} \right) \frac{tg(\kappa^{(1)})}{\Delta_i^{(1)}} + \left( \gamma^t T^t \cdot \gamma^{q_{11}} + N_i \gamma^{Ni} t \cdot \gamma^{q_{11}} \right) H_i \frac{tg(\kappa^{(2)})}{\Delta_d^{(2)}} \right].
\]  

(11)

Here we introduce following denotes \( \Delta^{(1)}_i = 1 - K^2 \left( 1 - \frac{p Y^d p t}{\gamma^t + N_i \gamma^{Ni} + P \gamma^P t} \right) \frac{tg(\kappa^{(1)})}{\kappa^{(1)}} \) and \( \Delta^{(2)}_d = 1 - K^2 \left( 1 - \frac{p Y^d p t}{\gamma^t + N_i \gamma^{Ni} + P \gamma^P t} \right) \frac{tg(\kappa^{(2)})}{\kappa^{(2)}} \); \( K^2 = \frac{p Y^d p d_{31}}{e_{33}} \) is the squared coefficient of electromechanical coupling; \( \kappa^{(1)} = k^{(1)} l / 2 \) and \( \kappa^{(2)} = k^{(2)} l / 2 \) are dimensionless parameters. The dispersions relation between wave number and angular frequency are as following expression:

\[
k^{(1)} = \sqrt{\frac{\bar{\rho}}{Y}} \omega , k^{(2)} = \sqrt{\frac{\bar{\rho}}{Y}} 2 \omega .
\]  

(12)

The first term in square brackets in Equation (11) describes the contribution to MEVC from the linear effect ME, and the second term in square brackets in Equation (11) describes the contribution from the nonlinear effect.

If \( \Delta^{(1)}_i = 0 \) or \( \Delta^{(2)}_d = 0 \), the parameter MEVC \( \alpha_E \) has a resonance peak. The increase of resonance is observed near the value of dimensionless parameters \( \kappa^{(1)} \approx (2n - 1) \frac{\pi}{2} \) and \( \kappa^{(2)} \approx (2n - 1) \frac{\pi}{2} \), where \( n = 1, 2, \ldots \). We can get the resonance frequencies from linear and nonlinear ME effect by using dispersion relation (12) and the following expressions:

\[
f^{(1)}_t = \frac{1}{2l} \sqrt{\frac{\bar{\rho}}{Y}} , f^{(2)}_t = \frac{1}{4l} \sqrt{\frac{\bar{\rho}}{Y}} = f^{(1)}_t .
\]  

(13)

Thus the value of resonance frequency for nonlinear (quadratic) ME interaction is half of the linear ME effect. For the calculation, we can use following parameters of the structure: the parameters of plate \( \text{Tb}_{1-x}\text{Dy}_{x}\text{Fe}_2 \) (Terfenol-D) is 15×5×0.5 mm and thicknesses of Ni layers are 0.2, 0.5, and 1 mm; the piezoelectric PZT layer with dimensions of 20×5×1 mm; the material parameters of Terfenol-D, PZT, and Ni are: \( \rho_p = 7.5 \times 10^3 \text{ kg/m³}, \rho_{Ni} = 8.9 \times 10^3 \text{ kg/m³}, s_{11}^P = 15 \times 10^{-12} \text{ m²/N}, s_{11}^Ni = 4.5 \times 10^{-12} \text{ m²/N}, \rho_{d31} = -175 \times 10^{-12} \text{ mV}. \) The values of linear and nonlinear piezomagnetic coefficients for Terfenol-D at \( H_{bias} = 400 \text{ Oe} \) were: \( q_{11}^{(1)} = 0.35 \times 10^{-61} / \text{Oe}; q_{11}^{(2)} = -0.4 \times 10^{-91} / \text{Oe²}. \) The values of linear and nonlinear piezomagnetic coefficients for Ni are dependent on the thickness of nickel layer. The parameters we used for calculations are: \( N_i = 0.2 \text{ mm}, q_{11}^{(2)} = 0.05 \times 10^{-61} / \text{Oe}; q_{11}^{(2)} = 0.1 \times 10^{-91} / \text{Oe²}, N_i = 0.5 \text{ mm}, q_{11}^{(2)} = -0.06 \times 10^{-61} / \text{Oe}; q_{11}^{(2)} = 0.2 \times 10^{-91} / \text{Oe²}; N_i = 1.0 \text{ mm}, q_{11}^{(2)} = -0.15 \times 10^{-61} / \text{Oe}; q_{11}^{(2)} = -0.25 \times 10^{-91} / \text{Oe²}. \) Using these parameters we get the values of resonance frequencies, as shown in table 1.
Table 1. Calculated resonance frequencies of Terfenol-D/PZT/Ni composites.

| $P_t$ (mm) | $L_t$ (mm) | $N_t$ (mm) |
|------------|------------|------------|
| 1          | 0.2        | 0.5        |
| 1          | 0.5        | 1.0        |
| Length vibration, linear effect $f_{l1}$ (kHz) | 83 | 86 | 93 |
| Length vibration, nonlinear effect $f_{l2}$ (kHz) | 41.5 | 43 | 46.5 |
| Flexural vibration frequency $f_b$ (kHz) | 13.5 | 16.5 | 20 |

These theoretical predictions were confirmed by experimental measurements. First, the linear magnetostriction of each composite was measured using a standard strain gauge technique in the directions parallel to $H_{dc}$ at room temperature. The dependence of the in-plane parallel magnetostriction $\lambda_0$ on $H_{dc}$ for Terfenol-D and Ni layers both with a thickness of 0.5 mm is shown in figure 2. These two materials exhibit different magnetostrictive properties. The Ni layer exhibits negative magnetostriction, whereas the Terfenol-D layer shows positive magnetostriction. When $H_{dc} > 600$ Oe, the magnetostriction of Ni was saturated. Conversely, the magnetostriction of Terfenol-D was not saturated even when $H_{dc}$ was as high as 2.5kOe. Although the magnetostriction of the Ni flake was small, it presented a relatively large linear piezomagnetic coefficient $d\lambda/dH$ of $-0.3$ ppm/Oe at 100 Oe, which then decreased to nearly zero as $H_{dc}$ increased further, as shown in figure 3(a). The $d\lambda/dH$ value of Terfenol-D was positive, with a maximum of about 0.43 ppm/Oe at 300 Oe. With increasing $H_{dc}$, $d\lambda/dH$ decreased almost linearly. Figure 3(b) shows the field dependency of nonlinear piezomagnetic coefficient $d^2\lambda/dH^2$ at bias magnetic field near 400 Oe. As shown in figure 3, the linear and nonlinear piezomagnetic coefficients for Terfenol-D and Ni have the opposite signs.

Figure 4 shows the $\alpha_{E,31}$ values of the Terfenol-D/PZT/Ni composites as a function of frequency ($f$) at $H_{dc} = 400$ Oe. The thickness of Terfenol-D was 0.5 mm and that of the Ni layer was 0.2, 0.5, or 1 mm to give $t_N/t_T$ of 0.4, 1.0, and 2.0, respectively. As shown in figure 4, three resonance peaks located around 20, 40, and 80 kHz were observed, which correspond to the first-order bending vibration, nonlinear and first-order longitudinal vibration, respectively, according to the above calculation results. The multiple resonance modes appear in the low-frequency region because of the asymmetric stress distribution in the Ni and Terfenol-D layers.

Figure 4. Frequency dependence of MEVC at $H_{dc} = 400$ Oe for composites with different $t_N/t_T$. Scattered data denote experimental results and solid line with same color denote theoretical calculations for longitudinal modes.
The magnetostriction of Ni is negative, whereas that of Terfenol-D is positive. For the bending modes it leads to increase the bending moment, because the Terfenol-D and Ni layers are located on opposite sides of the neutral layer. The magnitude of the induced voltage depends not only on the magnitude of the bending moment, but also on the location of the neutral layer relative to the axis of the piezoelectric. If the neutral layer is located on the axis of the piezoelectric, then the resulting voltage will be zero, since the tensile and compressed layers induced an opposite directions electric field. The bending moment is also increasing with increasing the thickness ratio $t_{Ni}/t_T$. In addition, it leads to increase the asymmetry of the neutral layer location of relative to the axis of the piezoelectric. All of these lead to an increase of the induced voltage and the MEVC for bending mode with increasing the $t_{Ni}/t_T$, which is in full accordance with figure 4.

For the longitudinal mode, when the magnetic field was applied, the Ni layer is compressed and the Terfenol-D layer is tensile. The value of MEVC, according to Equation (11), is proportional the sum of $Y T \cdot q_{t1} + \frac{Ni}{Nt} Y Ni \cdot q_{t1}$. When the Ni layer was thinner than the Terfenol-D layer, and as long as there is a relationship $Y T \cdot q_{t1} > \frac{Ni}{Nt} Y Ni \cdot q_{t1}$, the tensile deformation was dominant. As the $t_{Ni}/t_T$ increased, the tensile and compression deformations began to compete and the value of MEVC is decreased with increase the thickness of Ni-layer. When the thickness of the Ni layer has increased so much that the relationship: $\frac{Ni}{Nt} Y Ni \cdot q_{t1} > Y T \cdot q_{t1}$ has been fulfilled, since then the coefficient value begins to increase with increasing nickel layer. The low-frequency operation of ME composites can lower the eddy current loss in the magnetostrictive phase, resulting in efficient ME energy conversion. Furthermore, ME composites that operate at a bending resonance mode with low frequency are also advantageous to miniaturize devices, because the normal transverse resonance frequency of layered ME composites decreases with increasing length [19].

Figure 5 shows the $\alpha_{E,31}$ values as a function of $H_{dc}$ at $f = 1$ kHz for the three Terfenol-D/PZT/Ni threelayered composites with various $t_{Ni}/t_T$. The dependence of $\alpha_{E,31}$ on $H_{dc}$ was similar for the three samples. With increasing $H_{dc}$ from zero, $\alpha_{E,31}$ increased to a maximum value at around $H_{dc} = 400$ Oe and then decreased gradually. The negative $\alpha_{E,31}$ of the threelayered composite with $t_{Ni}/t_T = 1.0$ is mainly caused by the negative magnetostriction of the Ni layer at low $H_{dc}$. At low $H_{dc}$, $d\lambda/dH$ of the Ni layer is comparable with that of the Terfenol-D layer (figure 3(a)) when these two layers are of equal thickness, resulting in negative $\alpha_{E,31}$. With increasing $H_{dc}$, the magnetostriction of Ni became saturated and the $d\lambda/dH$ values approached zero. In contrast, the Terfenol-D layer did not show saturated magnetostriction even at 2.5 kOe.

![Figure 5. ME voltage coefficient $\alpha_{E,31}$ as a function of biasmagnetic field $H_{dc}$ at $f = 1$ kHz for composites with different $t_{Ni}/t_T$.](image-url)
Moreover, the $\alpha_{E,31}$ values were proportional to $d\lambda/dH$ and strongly depended on the volume fraction of the piezoelectric phase $v$ [20, 21]. The composite with $t_{Ni}/t_T = 2$ exhibited the largest $\alpha_{E,31}$ values throughout the measured $H_{dc}$ range, whereas that with $t_{Ni}/t_T = 1$ displayed the lowest $\alpha_{E,31}$ values. This may result from the competition between the contributions of $d\lambda/dH$ of Terfenol-D and Ni layers and the volume fraction $v$.

The composite with $t_{Ni}/t_T = 2$ has a thicker Ni layer than those in the other composites, which may result in large deformation and increase $d\lambda/dH$. However, the thicker Ni layer will decrease $v$, which will in turn decrease $\alpha_{E,31}$ [20, 21]. In this composite, the larger contribution from $d\lambda/dH$ dominated its behavior. Although a thinner Ni layer can increase $v$, the effective vibrational deformation of both Terfenol-D and Ni layers results in the small $\alpha_{E,31}$ values of the composite with $t_{Ni}/t_T = 1$, as shown in figure 5. This also can be interpreted from the plots of $\alpha_{E,31}$ against $v$, which display a double maximum when the longitudinal strain and flexural strain in the asymmetric threelayered composite are both taken into consideration, as predicted in our previous work [22].

4. Conclusion
Terfenol-D/PZT/Ni asymmetric ME laminated composites were designed to generate a bending vibration mode with the aim of realizing multifrequency and multimode operation in ME devices. In these Terfenol-D/PZT/Ni asymmetric three layered composites, three resonance modes, first-order bending resonance and nonlinear and first-order longitudinal resonance modes were observed. The ME outputs and resonance mode were tuned by $t_{Ni}/t_T$ of the composites. As $t_{Ni}/t_T$ increased from 0.4 to 2.0, the strength of the first-order longitudinal resonance was suppressed considerably, whereas the first-order bending resonance and nonlinear longitudinal resonance became stronger. The asymmetric three layered composites exhibited a large ME coefficient of 250mV/cm·Oe under a magnetic field of 400 Oe at quasi-static frequency. This work provides an approach to design ME devices with tunable ME resonance modes and multi frequency operation.

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