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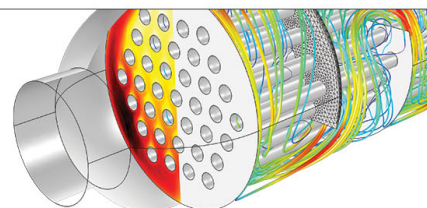P. Finkel, J. Bonini, E. Garrity, K. Bussman, J. Gao, J. F. Li, S. E. Lofland, and D. Viehland

Enhanced resonant magnetoelectric coupling in frequency-tunable composite multiferroic bimorph structures

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We report on a giant tunable enhanced resonant magnetoelectric (ME) coupling in multiferroic magnetostrictive/piezoelectric composite bimorph structures. The approach uses a magnetic/electric field assisted stress-reconfigurable resonance to produce frequency tuning of up to 100%. The studies were performed by laser Doppler spectroscopy. We also show that this principle of a continuously tuned resonance might be used to improve sensitivity for ME magnetic sensors. © *2011 American Institute of Physics*. [doi:10.1063/1.3560055]

In recent years the phenomenon of multiferroic behavior has been studied in materials that demonstrate both magnetic and electrical ordering. An exchange between subsystems results in a coupling of magnetic and electric properties.[1] To date, the highest magnetoelectric (ME) coupling has been found in the ME composites, where the magnetic and electric properties are mediated via strain across an interface between magnetostrictive and piezoelectric phases.[2–12] For example, magnetic sensors designed from Metglas/Pb(Zr,Ti)O$_3$ (PZT) ME composite structures have recently been shown sensitivity of ~ 100 pT/$\sqrt{Hz}$.[10,11] However, the limiting factor has been the ME coefficient [$\alpha_{ME} = dE/dH$ (Refs. 1–5)], that must be enhanced. It is established that the maximum of α_{ME} is achieved in a free vibration mode under electromechanical resonance (EMR) conditions.[1,2,5] At EMR α_{ME} is enhanced by the mechanical quality factor, Q, which can be 100 or greater. However, the advantage is useful only for very narrow bandwidth. This appreciably limits the performance of ME resonant mode devices in applications, where the frequency is variable. One solution is to lower Q at the expense of maximizing the ME coefficient. Another approach uses multielement ME laminates with different lengths in order to broaden the bandwidth.[9] The drawback is that its efficiency will be lower due to inadequate mechanical energy coupling of ME resonators of different frequencies. On the other hand, there have also been reports of tunable ME devices where the resonance frequency can be shifted by changing elastic properties of the ME components. For example, a resonance tuning of a longitudinal mode of TerfenolD/PZT laminates has been realized by applying dc magnetic bias and taking advantage of the pronounced magnetoelastic effect in TerfenolD.[13] This particular approach requires rather high magnetic fields, which cannot be varied quickly; and even then, only a relatively small frequency shift be realized without losing ME coupling at the saturation field.[13]

Recently, we have demonstrated tunability of the transverse resonance frequency of ME bimorph by prestressing

the double-side clamped (fixed end boundary conditions) ME structures and controlling the frequency shift by external magnetic field by forced magnetostrictive stresses.[14–16] Here, we show that it is possible to achieve stable enhancement in α_{ME} in a controllable resonant transverse fundamental mode by implementing this tunable approach.[16] It is confirmed that the bandwidth of the ME resonant response can be effectively broadened by resonance tuning via magnetic, electrical, or stress field.

In this investigation, we have studied the ME response in two different systems: Fe$_{(1-x)}$Ni$_x$(x $\sim 0.36, 0.42$)/polyvinylidene fluoride (PVDF) and asymmetric bimorph Metglas/PZT-fiber laminates, both operating in a transverse vibrational mode. The design and fabrication details of Metglas/PZT-fiber system were discussed previously.[2,11,13] The fabrication of Fe–Ni $(2 \times 50 \times 0.050$ mm$^3)$/PVDF (0.027 mm) laminate bimorphs have also been reported;[14] however, the current samples were made with the longitudinal axis normal to the rolling direction of the Fe–Ni film in order to maximize the effective magnetostriction λ.[16–18] ME samples were mounted on a nonmagnetic ceramic loading fixture [inset, Fig. 1(a)].

The key approach is the ability to create tunable prestress σ in the bimorph double-side-clamped laminated structure. Controlled tension was applied with a PZT piezoactuator installed inside the loading fixture [Fig. 1(a) inset].

Magnetic characterization of the ME sample was performed by vibrating sample magnetometry (VSM). The ac susceptibility of the ferromagnetic element was studied as a function of stress, both before and after fabrication of the structure. In order to measure the induced ME response, an ac magnetic field H_{ac} was superposed on a dc bias magnetic field H_{dc} produced by the VSM magnet and the voltage V_{ME} from the bimorph and the amplitude A of its oscillation was monitored as a function of H_{dc}. The piezoelectric components of the bimorphs were also driven by ac voltage as an excitation stimulus to vibrate the ME structure. The frequency was swept in order to determine resonance frequency f_0 and Q of the structure, and the resulting vibration was characterized via scanning laser Doppler vibrometry (LDV).
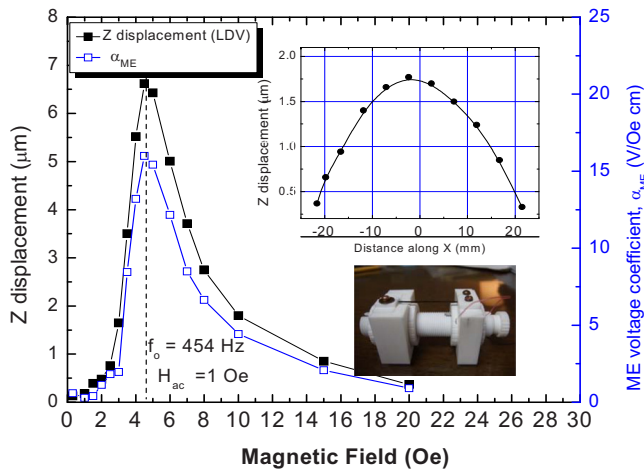
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FIG. 1. (Color online) Magnetic field bias dependence of transverse motion and ME coefficient (α_{ME}) in the Metglas/PZT-fiber sample. The top inset shows transverse vibrational mode measured as Z-displacement by LDV at the fundamental resonance $f_0 = 454$ Hz. The bottom inset show schematic diagram of the loading fixture.

The transverse vibration amplitude as a function of position [Fig. 1(a)] demonstrates that the device operated in the fundamental mode. Most of the details of the measurements have been described elsewhere previously.[14–16] Ferromagnetic resonance (FMR) studies were done at 9.2 GHz to measure the saturation magnetostriction of the Fe–Ni ribbons. Details of this technique can be found elsewhere.[19,20] The saturation magnetostriction was found to be about 30 ppm. The Metglas ribbons were not measured as the magnetostriction was reported by the manufacturer to be 32–35 ppm.

Figure 1(b) shows the field dependence of A and α_{ME} of the Metglas/PZT-fiber bimorph as a function of H_{dc}. The ac magnetic field used here to drive the ME laminate was 1 Oe. The two curves exhibit identical behavior with a maximum at ~4.6 Oe. The correspondence between the two curves confirms that the induced strain, which gives rise to V_{ME} in the piezoelectric element of the bimorph, is directly proportional to A. This allows one to use ac magnetic, electric, or elastic stimulation of the bimorph to investigate the ME coupling simply from the results of LDV [Fig. 1(b) inset].
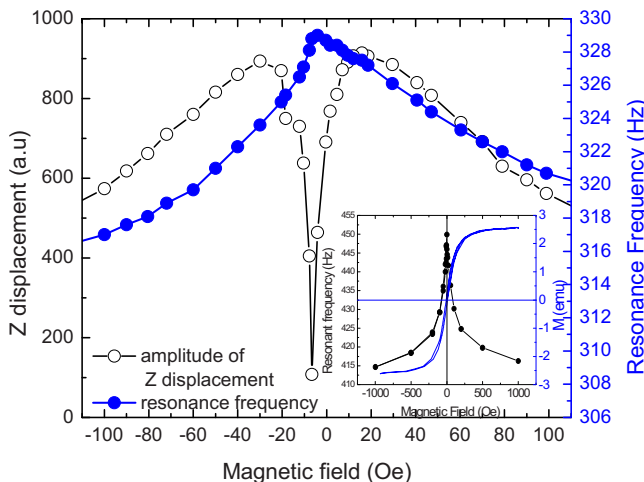
Figure 2 shows the transverse vibration and resonance

FIG. 2. (Color online) The amplitude of the transverse (z) vibration mode and frequency shift as a function of bias magnetic field (H_{dc}) for Ni–Fe/PVDF bimorph. Inset shows magnetic hysteresis loop overlapped with magnetic dependence of frequency shift in this sample.
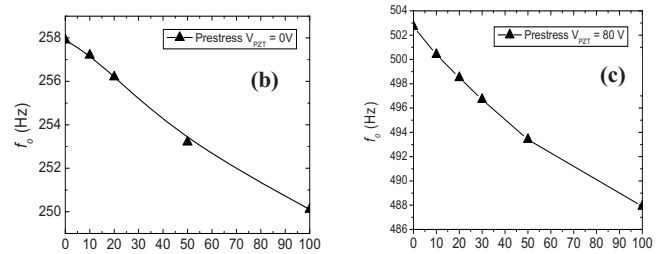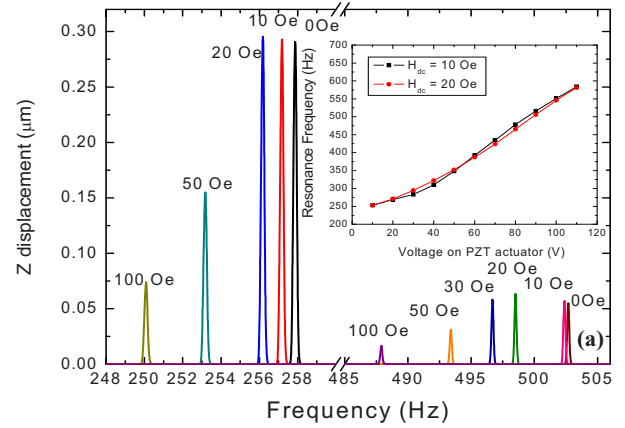

FIG. 3. (Color online) Frequency spectrum obtained for two selected levels of prestress levels created by PZT actuator also showing variation in the resonance frequency tuned by applied magnetic field H_{dc} in the Ni–Fe/PVDF bimorph (a); plots (b) and (c) illustrate magnetic field frequency shift tuned by magnetic field H_{dc} for these two different prestress levels.

frequency shift as a function of H_{dc} for the Fe–Ni/PVDF bimorph sample. Figure 3(a) illustrates how the resonance in the ME bimorph Fe–Ni ($x = 0.36$)/PVDF can be tuned over a rather large range of frequencies by controlling the tension in the structure simply by adjusting the loading of the fixture–frame with the PZT actuator. Figures 3(b) and 3(c) show the magnetic field dependence of the frequency spectrum for two different prestress levels. It can be seen the resonance frequency decreased notably with increasing dc magnetic bias.

For a clamped sample under a constant length condition with an initial axial stress σ_0, the resonance frequency f_0 is shifted by an applied magnetic field H due to a change in the net stress via the magnetostriction $\lambda(H)$.[15,16] This results in a resonant frequency change given by

$$f_0 = \frac{1}{2L}\sqrt{\frac{\sigma - \lambda(H)Y}{\rho_c}}, \tag{1}$$

where Y is the effective modulus of the bimorph structure, L the length of the laminate, and ρ the average density. Since $\sigma \gg \lambda(H)Y$, Eq. (1) suggests that Δf is linear in λ. Results shown in Fig. 3 fully support this. Note that there is a factor of 2 in the f_0 values for the two different levels of prestress, yet the low and high frequency data have the same field dependence. The value of Q was on the order of several hundred and effectively independent of H, suggesting that most of the damping is either from the piezoelectric and/or the viscous drag from the air. It has to be also noted that effective magnetostriction $\lambda(H)$ estimated from Eq. (1) at $H < 200$ Oe (i.e., in the unsaturated state) is ~20 ppm, in a reasonable agreement with the saturation value obtained from FMR.

Assuming an ideal bond between elements and that the piezoelectric has a much greater compliance than that of the
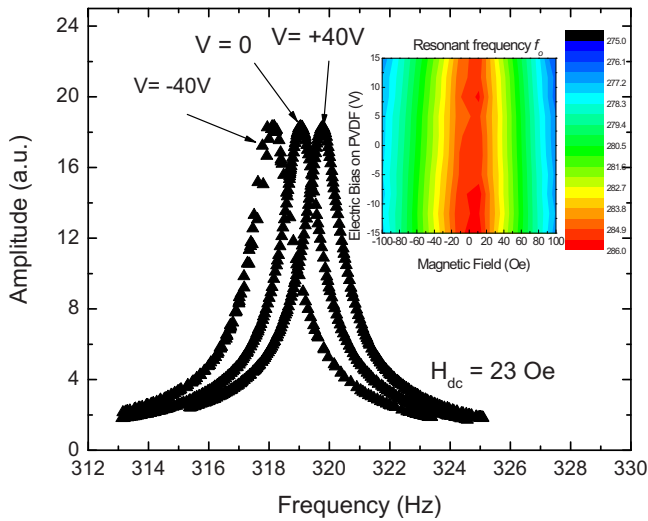
FIG. 4. (Color online) Effect of electric dc bias E_{PVDF} applied to PVDF layer on resonant frequency of transverse vibrational mode in Fe–Ni/PVDF bimorph. Inset shows the contour plot of resonance frequency f_0 as function of electric (E_{PVDF}) and magnetic (H_{dc}) fields for this sample.

magnetostrictive layer, which holds for the Ni–Fe/PVDF bimorph, the ME coupling constant $\alpha_{ME} \approx dV_{ME}/dH \sim (d\lambda/dH)d_{31}Q$ for an unconstrained longitudinal vibration where d_{31} is the piezoelectric coefficient. Since $\lambda(H) \sim M^2(H)/M_s^2$, where M_s is the saturation magnetization, $\alpha_{ME} \sim 2M(H)(dM/dH)Qd_{31}$. This is consistent with magnetic field dependent behavior for $V_{ME}(\sim \alpha_{ME})$ shown in Figs. 2 and 3. Note the maximum of A (and likewise α_{ME}) corresponds to the maximum of the dM/dH and disappears when $d\lambda/dH = 0$ (Fig. 2 inset), consistent with the previous reports.[4–8]

The reduction in A at higher frequency is related to the fact that there is already considerable elastic energy stored in the bimorph due to the stress. The relevant parameter of the clamped-clamped bimorph is the receptance, the amplitude displacement per unit force. Virgin and Plaut[21] have shown that for a beam under axial load, the receptance is effectively proportional to $1/f_0$. That is, α_{ME} is inversely proportional to f_0. This is in accord with the observed decrease in A with increasing frequency (Fig. 3). In contrast, A (and α_{ME}) of free-bending cantilevers is much more sensitive to frequency—the 3 dB bandwidth of the loaded double clamped bimorph is $2f_0$ whereas it is only f_0/Q for a free-bending one. As noted above, f_0 also depends on H as well as σ while the characteristics of the piezoelectric element are determined by the electric field E and σ. Thus, there is a very large parameter space for optimization of α_{ME} for a given frequency.

Figure 4 shows the effect of electric dc bias E_{dc} on resonant frequency shift in the transverse vibrational mode in Fe–Ni/PVDF bimorph sample when driven by E_{ac} applied to the PVDF layer. Note that a rather small bias of $\pm 6 \times 10^5$ V/m created perceptible frequency shift $\Delta f_0 > 2$ Hz. Figure 4 inset shows the contour plot of f_0 as function of E_{dc}

and H for the Fe–Ni/PVDF sample. While the shift in either f_0 or α_{ME} is not large, considering the compliance of the PVDF as compared to that of the Fe–Ni laminate, the effect is remarkable and will be noticeably enhanced in devices with stiffer piezoelectric elements.

In summary, we demonstrated here that by implementing tunable prestress in a fix-fix double clamped ME bimorph it is possible to achieve high effective enhancement of the ME coupling coefficient (almost up to two orders of magnitude in selected cases) in a controllable resonant mode in a wide frequency range. We also report that bandwidth ME bimorph structure can be notably increased via resonant frequency tuning using combinations of magnetic, electric, and mechanical conditions. These results show the multiparameter tuning of the ME device response in a rather wide band of frequencies and it is actually achievable without a sacrifice in ME coupling efficiency and sensitivity.

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