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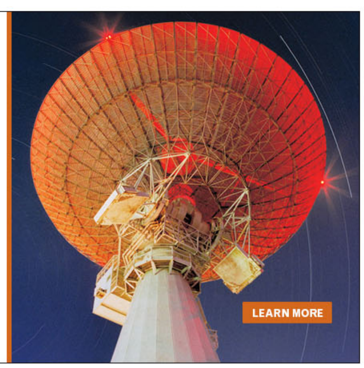

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Giant magnetoelectric torque effect and multicoupling in two phases ferromagnetic/piezoelectric system

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The physical mechanism of a non-magnetostrictive magnetoelectric (ME) effect was revealed and designated as the ME torque (MET) effect. Experimental results showed that the MET effect could be huge; a simple MET device could achieve giant ME voltage coefficients of 100 V/cm.Oe at 1 Hz and 2100 V/cm.Oe at the first order resonant frequency. These are the highest reported ME coefficients in a bulk device ever. We then proposed the multicoupling ME effect, which comes from the interaction of magnetostriction, magnetic torque, and piezoelectricity, and rewrite the ME constitutive tensor equation. The abnormal phenomenon in the (1-3) structure ME thin film that T-L mode might bring larger ME coupling than L-L mode was successfully explained from the multicoupling concept. These researches have extended the giant ME effect from the traditional magnetostrictive/piezoelectric system to a common ferromagnetic/piezoelectric system, and gave more choices to scientists/engineers for constructing the giant ME device. © *2011 American Institute of Physics*. [doi:10.1063/1.3662912]

I. BACKGROUND

The magnetoelectric (ME) effect is characterized by an induced dielectric polarization in response to an applied magnetic field (H), or by an induced magnetization in response to an applied electric field (E).[1] It can be evaluated by a parameter designated as the ME voltage coefficient $\alpha_{ME,}$ which can be written as $\alpha_{ME} = \delta E/\delta H = (\delta V_i/t)/\delta H$ (where H is the applied magnetic field, E and V_i are the electric field and voltage induced across the piezoelectric (PE) layer, and t is the thickness or more accurate the effective distance between the electrodes of the PE phase).[2]

The ME effect was proposed earlier by Curie in 1894 and discovered experimentally in Cr_2O_3 by Astrov in 1960.[3,4] After that, numerous single phase ME materials were reported, however, direct coupling between spin and dipole in atomic scale is very difficult, which makes the ME effect in single phase quite small.[5–9] Scientists then turned to the consideration of structural composites; the ME particle composite was first reported by Philips Research Lab in Holland in the mid-1970 s. This composite approach combines magnetostrictive (MS, or piezomagnetic (PM)) and PE phases, which interact via elastic force.[10] Later on, many successful researches were carried out.[11–14] However, the ME effect in a particle composite, although much larger than that in a single phase, is still much smaller than that predicted by theory due to some technical limitations.[1] Until 2001, the giant ME effect was first reported in laminated composites consisting of MS and PE layers, although both giant MS and PE materials had been found for decades.[15] Since then, the researches on MS-PE laminate composite developed quickly and successfully in theory and engineering.[16–23] However, the applications are always limited by the materials properties, price, and the forming technology. Some people then tried to find new giant MS materials, new forming technologies, and some began to seek new mechanisms.[24,25]

Although the ME effect could appear in the ferromagnetic (FM)/PE two phase system from the symmetry point of view, giant ME contribution of the two phase system are always accredited to the MS-PE interaction.[1,23,26] This might comes from the intuitive understanding of the intimate contact coupling of the MS and PE phase, however, another coupling has been overlooked for years. In the following section, we will introduce this ME coupling which comes from magnetic-torque (MT). This non-MS ME coupling will give us more consideration on the ME effect contribution; multicoupling from magnetostriction, magnetic-torque, and piezoelectricity. Giant ME devices will not be limited to the MS-PE two phase system, but to a broader system constructed from the FM-PE phase.

II. MECHANISM AND EXPERIMENTS

The MT $\boldsymbol{\tau}$ to a FM phase in an applied magnetic field **B** can be calculated from the classic electrodynamics as[27]

$$\tau = \mathbf{m} \times \mathbf{B} = (\mathbf{M} \times \mathbf{B}) \cdot V, \quad (1)$$

where **m**, **M** are the magnetic moment and magnetization of the FM phase, respectively, and V the phase's volume. This MT $\boldsymbol{\tau}$ will try to line up the magnetic moment **m** and the external field **B**. If one tried to keep **m** and **B** in different orientation by structural design or specific boundary condition, strain will occur inside the FM phase. If this strain is coupled to the PE phase, the electro-mechanical coupling inside the PE phase then induces a charge output. This is how and why the MT will contribute to the ME effect in a FM-PE two phase system. We designated the MT to the ME effect as ME

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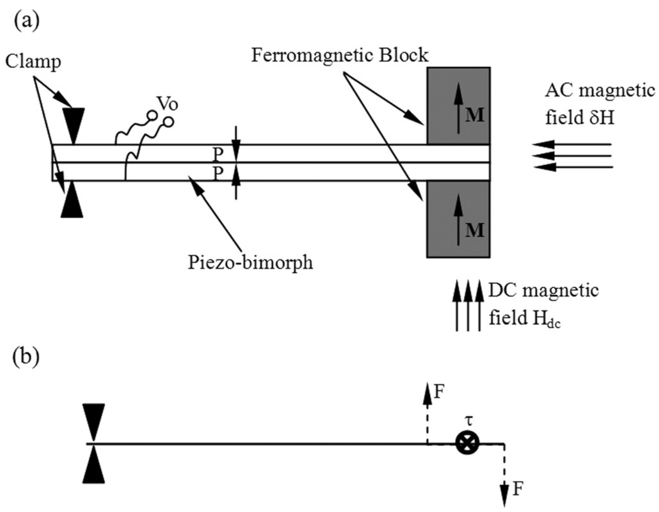

FIG. 1. (a) A recommended structure for the MT-PE coupling device; and (b) illustration of the working mechanism.

TABLE I. Magnetic unit samples.

No.	Material	Magnet shape	Magnet volume (cm^3)
F1/N1	Ferrite/NdFeB	$2\times\phi8\times3$	0.3
F2/N2	Ferrite/NdFeB	$2\times\phi8\times6$	0.6
F3/N3	Ferrite/NdFeB	$2\times\phi8\times9$	0.9
F4/N4	Ferrite/NdFeB	$2\times\phi8\times12$	1.2

torque (MET) effect, and as a comparison, we designated the traditional MS-PE coupling ME effect as MES effect. From equation (1), we could evaluate the MET effect by

$$\alpha_{MET} = \kappa \cdot |\mathbf{M}| \cdot \sin\theta \cdot V/\mu_0, \tag{2}$$

where κ is the MT-PE transfer function, θ the angular between **M** and **B**, and μ_0 the permeability of the free space.

From equation (2), one can see that the α_{MET} linearly increases with κ, M, and V. The angle θ brings a spatial effect to the α_{MET}, and a maximum value of α_{MET} could be achieved as it is equal to 90°. M, V, and θ could be easily controlled by the materials/device designers, however, κ is quite a complex parameter; it is highly structural dependence. To increase κ, one should increase the bending mode MT-PE coupling due to the torque contribution. A simple piezo-bimorph structure, as recommended in Fig. 1(a), will help much. The apply DC magnetic field H_{dc} helps to increase the magnetization M inside the ferromagnetic unit. A ferromagnetic unit with large residual flux density, such as Fe_3O_4, NdFeB, AlniCo, Ticonal, SmCo, etc., will help to save the extra mechanism for producing the H_{dc}. If one tries a ferromagnetic unit with low residual flux density, such as cobalt, nickel, silicon-steel, conetic, permalloy, mumetal, metglass, etc., the H_{dc} should be applied. The higher the saturate magnetic flux density Bs, the higher the MET effect could achieve.

To evaluate how strong the MET effect could be, we constructed MET devices with structure shown in Fig. 1(a). We used NdFeB (N35 type) or Fe_3O_4 Ferrite as the ferromagnetic units for saving the mechanism of producing the H_{dc} due to their high residual flux density (1.2 T and 0.3 T, respectively), and they all have low magnetostriction. The sizes of the ferromagnetic units used in the experiments are shown in Table I. The piezo-bimorph was constructed by two pieces of PZT-850 (each $30\times6\times0.3$ mm^3 in size), whose layers were poled in opposite directions. The ferromagnetic units were attached to the tip of the PZT bimorph by use of a high strength epoxy. In Fig. 2(a), it can be seen that, with the mass of the magnetic units increase, the resonant frequency will decrease; this is because of the tip mass loading contribution.[28] And the NdFeB has much higher

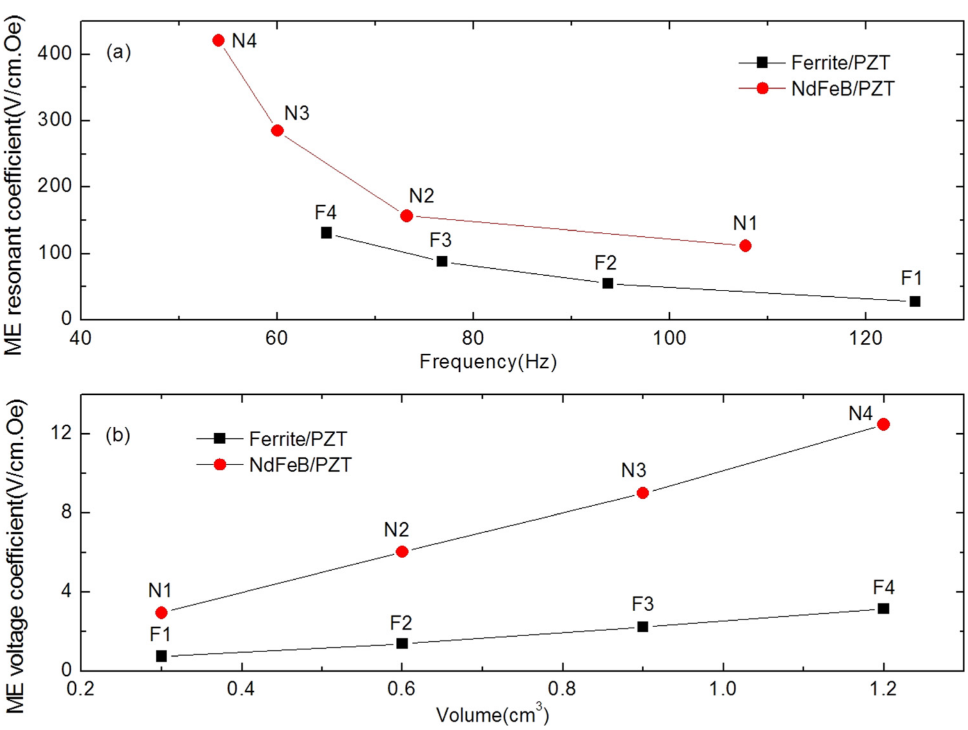

FIG. 2. (Color online) ME voltage coefficients for samples in Table I (a) at the first order bending resonant frequency; and (b) on the magnetic units volume V at 10 Hz.

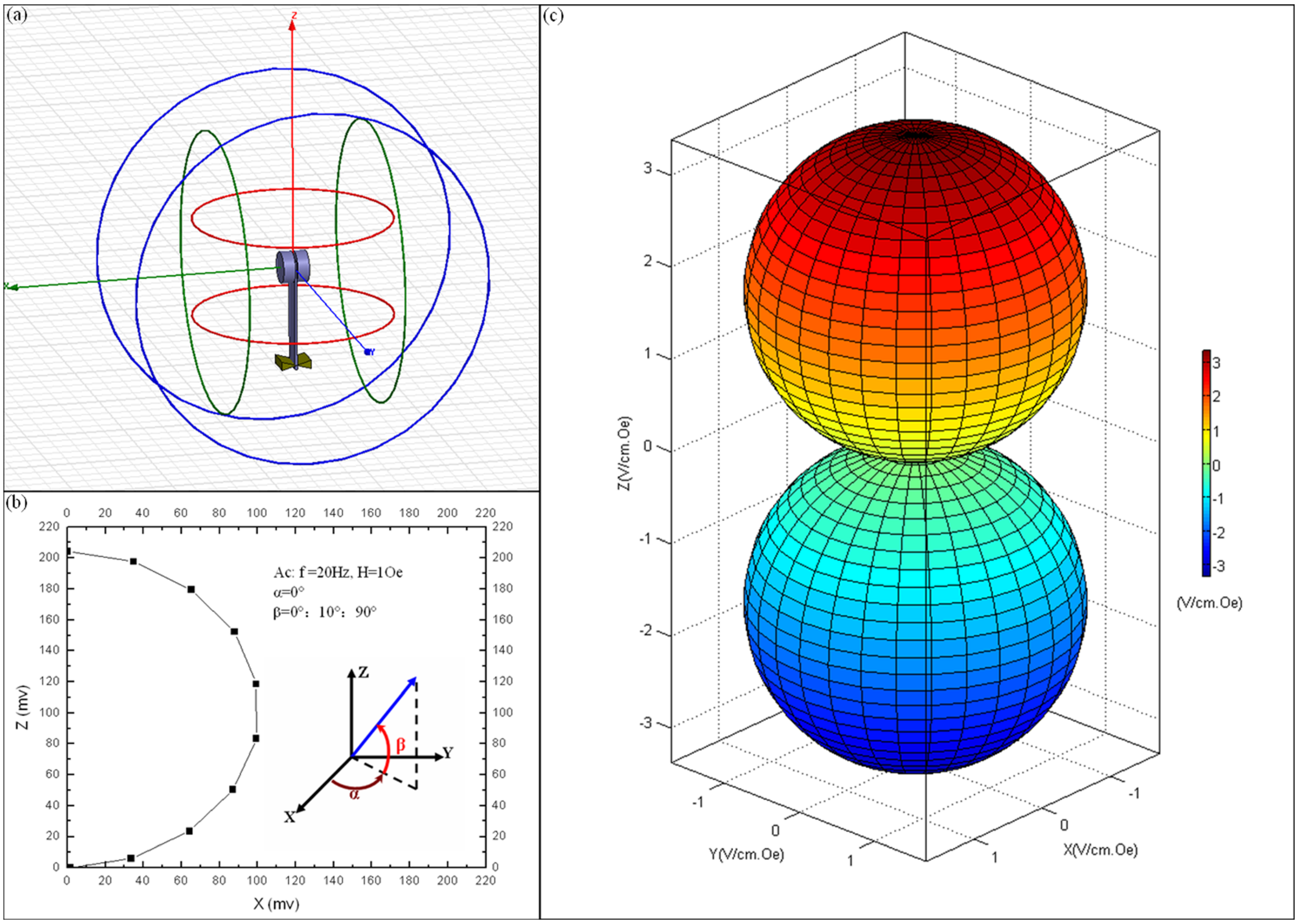

FIG. 3. (Color online) (a) Setup for spatial ME measurement; (b) ME effect in one quadrant plane; and (c) full spatial ME effect.

residual flux density than that of the Fe_3O_4 ferrite; which leads to a much higher MET effect at the resonant frequency for the same mass. Figure 2(b) supports equation (2) quite well: i) The ME voltage coefficients linearly increased with the volumes; ii) the slopes are 10.53 and 2.7 for the NdFeB/PZT samples and ferrite/PZT samples, separately, which coincided well with the ratio of their residual flux densities. The spatial MET effect could be evaluated by setting the MET device (N1) inside three Helmholtz coil pairs with each axial perpendicular to each other, as shown in Fig. 3(a). The experimental data for the ME effect were shown in Fig. 3(b) for one quadrant plane ($\alpha = 0°$, $\beta = 0$: $10°$: $90°$) and Fig. 3(c) for full space. It can be seen that the ME effect is $\sin\theta$ dependence, which also fit equation (2) as well.

We also used two pieces of NdFeB (N42 Type, Br≈1.3 T) with size of $50.8 \times 25.4 \times 3.2$ mm^3 each as the ferromagnetic units and did the same ME measurement. We acquired a figure of ME voltage coefficient as illustrated in Fig. 4. In this result, we acquired giant ME coefficients of 100 V/cm.Oe or 400 nC/cm.Oe at a low frequency (1 Hz) and 2100 V/cm.Oe or 8.4 μC/cm.Oe at the first order resonant bending frequency. These values are much larger than the highest value ever reported for a MS-PE device, which was 22 V/cm.Oe at the low frequency and 737 V/cm.Oe at the resonance.[25,29] These results show that the MET effect could be huge and we should take the MET effect into the ME coupling consideration.

III. MULTICOUPLING AND DISCUSSION

Since the FM phase possesses the MT easily ($\theta \neq 0$), the MET effect exists widely in multiferroics composite and should be taken into the ME coupling consideration. As the MET was taken into the consideration, the traditional MS-PE coupling will then be extended to a multi-coupling of

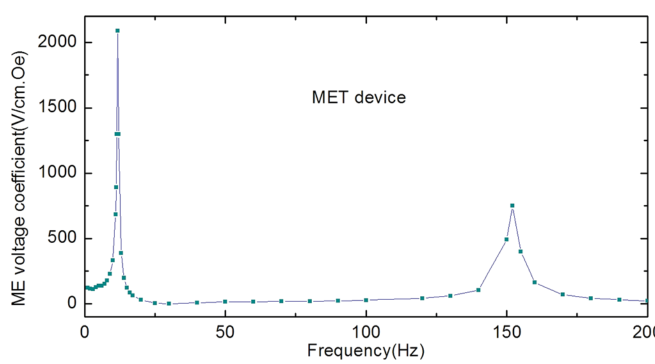

FIG. 4. (Color online) Frequency dependence of the ME voltage coefficient for the MET device.

magnetostriction, magnetic-torque, and piezoelectricity, and the ME coefficient could be evaluated as

$$\partial P/\partial H = \alpha = k_c e^m e + k_t e^T e + \rho(e^m, e^T)e, \tag{3a}$$

$$\partial P/\partial H|_{e^T=0} = \alpha_{MES} = k_c e^m e, \tag{3b}$$

$$\partial P/\partial H|_{e^m=0} = \alpha_{MET} = k_t e^T e, \tag{3c}$$

where k_c is the coupling factor of the MS and PE phase, e^m and e are piezomagnetic and piezoelectric coefficients, respectively, k_t is the coupling factor between the magnetic-torque and PE phase, e^T the magnetic-torque strain factor, $\rho(e^m,e^T)$ the correlation factor of the magnetic-torque and piezomagnetic to the strain.

For the case of $e^T=0$ (nonmagnetic torque case), equation (3a) becomes (3 b), the same as the previous report,[23] and the ME coupling will be a pure MS-PE coupling. For the case of $e^m=0$ (nonmagnetostrictive case), equation (3a) becomes (3 c), and one can still achieve a giant ME effect by making full use of e^T. For a normal case that e^T and e^m coexist, giant MS-PE coupling does not mean a giant ME coefficient due to the fact that e^T might play a contrary role, and giant MT-PE coupling does not mean giant ME effect either. Proper design should consider both MS and MT contribution. MT-PE coupling was considered mostly in Fig. 1(a); however, for multicoupling, Fig. 1(a) might not be a good choice because it might reduce the MS contribution. The design will be quite complicated as the multicoupling is taken into the consideration. As an example, let us consider a fundamental structure of clamped-free unimorph as show in Fig. 5. One can easily judge that δH_1 helps the MS-PE coupling, while δH_2 helps the MT-PE coupling. An optimized multi-coupling will require that the device is set to some angle to the detected magnetic field δH_T; and we designated this angle ϕ as optimized multi-coupling angle φ_{MOPT}. It is a complex parameter that related to the materials properties, device structure and the boundary condition (clamp status, H_{DC}, etc.). The electric field E of the device can then be calculated as

$$\begin{aligned} E &= \alpha_{ME}\cdot H = \alpha_{MET}\cdot H + \alpha_{MES}\cdot H + \rho(\alpha_{MET},\alpha_{MES})H \\ &= T\cdot\sin\theta\cdot|B'|\cdot|B|\cdot V/\mu_0 + \alpha_{MES}\cdot H + \rho(\alpha_{MET},\alpha_{MES})H. \end{aligned} \tag{4}$$

It is not that easy to determine the correlation factor ρ due to the fact that the stress, strain, electric displacement, magnetic induction would coupling together, which makes it complex. It will be much better to evaluate α_{ME} by writing the constitutive equation in tensor form for the magnetic-mechanical–electric coupling as follows:

$$\boldsymbol{\sigma} = c\mathbf{S} - e^T\mathbf{E} - (q^T + q_M^T)\mathbf{H}, \tag{5a}$$

$$\mathbf{D} = e\mathbf{S} + \varepsilon\mathbf{E} + \alpha\mathbf{H} + \mathbf{P}_S, \tag{5b}$$

$$\mathbf{B} = (q + q_M)\mathbf{S} + \alpha^T\mathbf{E} + \mu\mathbf{H} + \mathbf{M}_S, \tag{5c}$$

where $\boldsymbol{\sigma}$, **S**, **D**, **E**, **B**, and **H** are stress, strain, electric displacement, electric field, magnetic induction, and magnetic field tensor, respectively, c, ε, and μ are the stiffness, dielectric constant, and permeability tensor separately; e, q, q_M are, respectively, piezoelectric, piezomagnetic, magnetic-torque (MT) coefficient tensor; $\mathbf{P_S}$, $\mathbf{M_S}$ are the spontaneous polarization and magnetization tensor separately; and α is the ME coefficient. The superscript T denotes the transpose of the tensor. The solution for static and dynamic status could refer to Refs. 12 and 30, respectively. However, finite element method will help much as the FM-PE system become complicated.

Figure 5 is the fundamental for nano-pillar ME composites grow on a substrate. As the multi-coupling ME effect is considered, the anomaly that the ME coefficient of T-L mode were slightly higher than that for L-L mode in (001) BF-CF/STO self-assemble (1-3) structure nano composite could be easily explained.[31]Although the ME contribution from the magnetostriction for T-L mode (d_{31} prefer) is much smaller than that for the L-L mode (d_{33} prefer), the ME contribution from magnetic torque for the T-L mode (bending prefer) is much larger than that for the L-L (expanding prefer) mode, which in multi-coupling combination leads to a higher ME effect in the T-L mode than that in the L-L mode.

IV. SUMMARY

In summary, the physical mechanism of the MET effect was revealed and proved to be huge by a simple MET devices; giant ME coefficients of 100 V/cm.Oe or 400 nC/cm.Oe at a low (sub-resonant) frequency and a 2100 V/cm.Oe or 8.4 μC/cm.Oe at the first order resonant frequency was achieved. Multicoupling ME effect, which comes from the interaction of magnetostriction, magnetic-torque and piezoelectricity, is taken into the consideration of designing device with the giant ME effect. The construction of giant ME devices have been extended to a broad range of ferromagnetic-piezoelectric systems.

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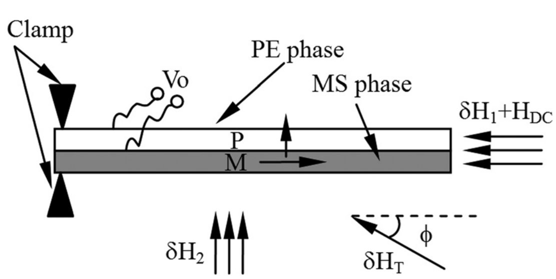

FIG. 5. Illustration of multicoupling for a unimorph cantilever.

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