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Influence of mechanical failure on the magnetoelectric response of magnetostrictive/piezoelectric multiferroic composite

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Abstract Based on modified multi-field coupling equations, the magnetoelectric (ME) response of a new kind of $Ni_{47.4}Mn_{32.1}Ga_{20.5}$/PZT multiferroic composites are calculated by considering the mechanical failure of the brittle PZT substance. It is theoretically revealed that the $Ni_{47.4}Mn_{32.1}Ga_{20.5}$/PZT bilayer composites could produce an ideal giant ME (GME) response up to 120 V/cm Oe, much larger than the best reports up to now. However, the real ME response will be strongly limited by the mechanical strength of the brittle PZT. Reducing the PZT layer and using a mechanically stronger PZT material have been suggested to enhance the ME response.

Multiferroic materials [1–3] have drawn increasing interest due to their multi-functionality, which provides significant potentials for using as next-generation multi-functional devices. The characteristic of these multiferroic materials is the coupling interaction between the multiferroic orders to produce some new effects, such as magnetoelectric (ME) effect [4, 5]. The ME response is an appearance of an electric polarization upon applying a magnetic field and hence the electric polarization of ME materials will be variant with external magnetic field [6, 7]. Multiferroic composites, made by combining ferromagnetic (magnetostrictive) and ferroelectric (piezoelectric) substances together, such as ferrite/titanate [8, 9] and ferrite/lead–zirconate–titanate (PZT) [10, 11], have been found to exhibit an extrinsic ME response, resulting from an elastic coupling interaction between the two substances. These multiferroic composites could have ME effect much larger (orders of magnitude higher) than the monophase ME materials and could be used in room temperature, which makes them have more possible applications such as in sensor, actuators, and transducers [12–14].

In order to improve the elastic coupling interaction between the two constituent substances and thus enhance the ME response in the magnetostrictive/piezoelectric multiferroic composites, much attention has been paid on the progressive usage of the magnetostrictive phase that has a superior magnetoelastic response. Giant magnetostrictive rare-earth-iron alloy $Tb_{1-x}Y_xFe_2$ (Terfenol-D), the best known and widely used magnetostrictive alloy having magnetostriction of up to 1,000 ppm [15], has been recently used [16–19] to combine with piezoelectric materials (such as PZT) to form multiferroic composites, producing a GME response (ME coefficient of several V/cm Oe). Most recently, iron-based Metglas (such as FeBSiC and FeSiCo) have been used [20–22] in the magnetostrictive/piezoelectric multiferroic composites to improve the ME coefficient up to $\sim$20 V/cm Oe. This significant improvement is attributed to the high magnetic permeability (>40,000) of the Metglas, which results in an effective piezomagnetic coefficient ($\sim 4 \times 10^{-6}$/Oe) much larger than that of the Terfnol-D counterpart ($\sim 1.2 \times 10^{-6}$/Oe [23]). However, the magnetostriction of the Metaglas is only $\sim$45 ppm, which will limit the extensive practical applications of this kind of multiferroic composites.

Alternatively, NiMnGa alloys exhibit both giant magnetostriction (up to 60,000 ppm [24, 25]) and large

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effective piezomagnetic coefficient (>10 × 10^{-6}/Oe [26]), which makes them most favorable for serving as the magnetostrictive phase in the laminated multiferroic composites. It seems that the multiferroic composites composing the constituent phases of NiMnGa and piezoelectric material should exhibit a super-GME response much larger than all the present reports. This, however, should be strongly affected by the mechanical failure of the brittle piezoelectric material. As well known, the elastic coupling interaction in the multiferroic composites is based on the transfer of magnetostriction to the piezoelectric materials. The piezoelectric materials can usually bear a tensile strain of only ~0.2% [27] and the application of more external strain should cause the mechanical failure, which means that the magnetostriction (up to 6%) induced in the NiMnGa alloy could not be fully transfer to the piezoelectric materials because of the limitation of mechanical failure. This issue, i.e., the influence of mechanical failure on the multiferroic coupling effect or on the ME response, is especially important because more and more giant magnetostrictive ferromagnetic materials are being used for multiferroic composites. Some fundamental understandings on this issue are thus in urgent need.

In this letter, we employ a finite-element method (FEM) to calculate the GME response in the laminated $Ni_{47.4}Mn_{32.1}Ga_{20.5}$/PZT composites, with aim to present the quantitative dependence of the GME response on the mechanical failure of brittle PZT. Our simulation results show that the maximum GME effect that could be achieved in the laminated multiferroic composites is controlled by the strength of the piezoelectric materials, which could be used to assist the design of multiferroic composites with superior ME effect. This calculation method is also applicable to other laminated magnetostricitive/piezoelectric systems.

The $Ni_{47.4}Mn_{32.1}Ga_{20.5}$ alloy considered here is a single crystal that has a magnetic-field-induced strain up to 6 × 10^4 ppm even at room temperature [24]. This giant strain is resulted from the twin boundary motion driven mainly by the Zeeman energy difference across the twin boundary [28], where an initial single-variant state was pre-formed by application of a compressive stress of the order of 1 MPa. As shown in Fig. 1a, the rectangular $Ni_{47.4}Mn_{32.1}Ga_{20.5}$ sample (6 × 6 × 20 mm) is oriented along the X_1 axis, which is the same direction of the pre-applied compressive stress. When a transverse magnetic field $\boldsymbol{H}_2$ is applied, twins magnetized vertical to the field will appear and grow and the field-induced twin boundary motion makes the sample elastically elongate along the length direction or the X_1 direction. The saturated magnetic-field-induced strain is strongly dependent on the pre-applied opposing stress. As revealed from Fig. 1b, the saturated strain could be achieved up to 6% at pre-stress levels below about 1.1 MPa even at room temperature. However, when

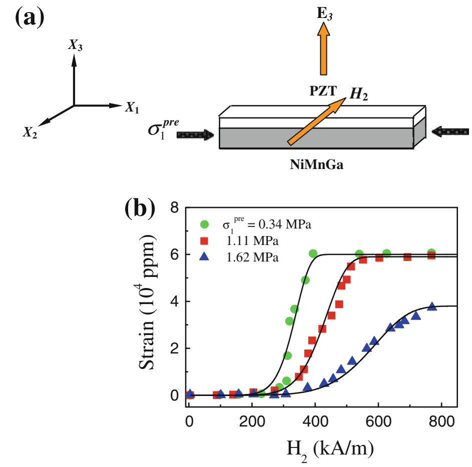

Fig. 1 **a** Triplanar sketch and definition of the coordination axes for the block-shaped laminate composite of $Ni_{47.4}Mn_{32.1}Ga_{20.5}$ and PZT and illustration on the application of magnetic field (along X_2 axis) and pre-stress (opposing along X_1 axis) on $Ni_{47.4}Mn_{32.1}Ga_{20.5}$ alloy. **b** Dependence of magnetic-field-induced strain of the $Ni_{47.4}Mn_{32.1}Ga_{20.5}$ alloy on the magnetic field ($\boldsymbol{H}$) as a function of the pre-applied compressive stress, σ_1^{pre}. *Dots* are experimental results [24] and curves are fitting ones

the pre-stress levels exceed 1.1 MPa, the strain saturates at reduced values. Monotonically, the saturated strain decreases with increasing the pre-applied compressive stress.

The response of $Ni_{47.4}Mn_{32.1}Ga_{20.5}$/PZT bilayer composite (Fig. 1a) involving the magneto-electro-elastic coupling effect can be described by the modified general multi-field coupling equations [29–32]:

$$\begin{aligned} \sigma_{kl} &= \boldsymbol{c}_{klij}^{\mathrm{H,E}}\varepsilon_{ij} - \boldsymbol{e}_{ikl}^{\mathrm{H}}E_i - \boldsymbol{c}_{klij}^{\mathrm{H,E}}\varepsilon_{ij}^{\mathrm{M}} - \sigma_{kl}^{\mathrm{pre}}, \\ \boldsymbol{D}_k &= \boldsymbol{e}_{kij}^{\mathrm{H}}\varepsilon_{ij} + \kappa_{ki}^{\mathrm{H},\varepsilon}\boldsymbol{E}_i + \alpha_{ki}\boldsymbol{H}_i, \qquad (1) \\ \boldsymbol{B}_j &= \mu_{ji}(\varepsilon,\boldsymbol{E},\boldsymbol{H})\boldsymbol{H}_i, \end{aligned}$$

where σ is the stress, ε the strain, $\boldsymbol{D}$ the electric displacement, $\boldsymbol{E}$ the electric field tensor, $\boldsymbol{B}$ the magnetic induction, and $\boldsymbol{H}$ the magnetic field; $\boldsymbol{c}$, κ, μ are, respectively, the stiffness at constant field, dielectric constant tensor at constant strain, and permeability constant tensor at constant strain; $\boldsymbol{e}$ is the piezoelectric coefficient tensor, α the ME coefficient, and ε^{M} the magnetostrictive strain. Of special interest to note is that Eq. 1 includes the item of the pre-applied stress, σ^{pre}, which is a modification on the traditional multi-field coupling equations [31, 32]. Based on Eq. 1 and the $\boldsymbol{H} - \varepsilon_1^{\mathrm{M}}$ curves (Fig. 1b), FEM calculations were performed[1] following our previous treatments [29, 30, 33] and the ME coefficient was estimated as $\alpha_{E32} = -\boldsymbol{E}_3/\boldsymbol{H}_2$. As to the mechanical failure

[1] The values of relevant parameters used in calculations (in SI units) are from Ref. [29, 30, 33]: (c_{11}, c_{12}, c_{33}, c_{44}) = (157, 120, 128, 107) Gpa for the NiMnGa alloy and (c_{11}, c_{12}, c_{33}, c_{44}) = (121, 75.4, 111, 21.1) Gpa, (ε_{11}, ε_{33}) = (916, 830), and (e_{31}, e_{33}, e_{15}) = (−5.4, 15.8, 12.3) C/m^2 for PZT.

of the brittle PZT material, the failure criterion was defined as $|\sigma_1| \geq \sigma_1^c$, where σ_1 is the principal stress along X_1 direction in PZT and σ_1^c is the critical strength generally of several tens MPa (the range from 40 to 80 MPa will be assumed in the following calculations).

For comparison reason, no mechanical failure is firstly considered. Taking $\boldsymbol{t}_m/\boldsymbol{t} = 0.8$ ($\boldsymbol{t}_m$ is the thickness of $Ni_{47.4}Mn_{32.1}Ga_{20.5}$ layer and $\boldsymbol{t}$ is the total thickness of the laminate composite), the ME coefficient was calculated under different pre-applied stress, shown as dash lines in Fig. 2a. It is surprisingly shown that a super-GME effect with α_{E32} up to 120 V/Oe cm $\left(\sigma_1^{pre} = 0.34\,\mathrm{MPa}\right)$ could be achieved in the $Ni_{47.4}Mn_{32.1}Ga_{20.5}$/PZT multiferroic composite. However, simulations reveal that, accompanying with the giant strain, the stress induced in PZT could raise up to several hundreds MPa, much larger than the critical strength (several tens MPa) of PZT (see, e.g., Ref. [27]). This stress, in excess of the critical strength, will actually cause mechanical failure. The obtained super-GME effect above is thus deemed to be an ideal case. Further consideration of the mechanical failure is needed to understand the practical performance of laminated multiferroic composites under large magnetic-field-induced strain.

The calculations of the ME effect with considering the mechanical failure with $\sigma_1^c = 60$ MPa are depicted in Fig. 2a as solid curves. With the applied magnetic field, the increase in magnetic-field-induced strain (see Fig. 1b) causes a gradual increase not only in the ME effect but also in the stress in PZT. When the stress achieves σ_1^c, mechanical failure is defined and no further operation is assumed for the laminated multiferroic composites. In Fig. 2a, the critical point marked by arrow in the curves means the occurrence of mechanical failure, after where the ME effect will be no longer produced. From Fig. 2a, one can clearly find that the influence of mechanical failure of brittle PZT on the ME effect is significant in the laminated multiferroic composites. Taking the case of $\sigma_1^{pre} = 0.34$ MPa for example, the ideal maximum ME effect without consideration of mechanical failure is high up to bout 120 V/Oe cm. However, the limit of $\sigma_1^c = 60$ MPa reduces the ME effect down to 50 V/Oe cm, over twice less than the former ideal prediction. Similar reduction is also observed in the other two cases of $\sigma_1^{pre} = 1.11$ and 1.62 MPa.

The calculated dependence of ME effect on σ_1^c is summarized in Fig. 2b. A larger σ_1^c resulting in a higher ME effect is generally found for all the three cases under different pre-applied stress. From Fig. 2b, it is of special interest to note that the ME effect in the laminated $Ni_{47.4}Mn_{32.1}Ga_{20.5}$/PZT multiferroic composite could go easily to several tens V/Oe cm even at room temperature (for example, α_{E32} larger than 80 V/cm Oe at $\sigma_i^{pre} =$ 0.34 MPa and $\sigma_1^c = 80$ MPa), much larger than the best-

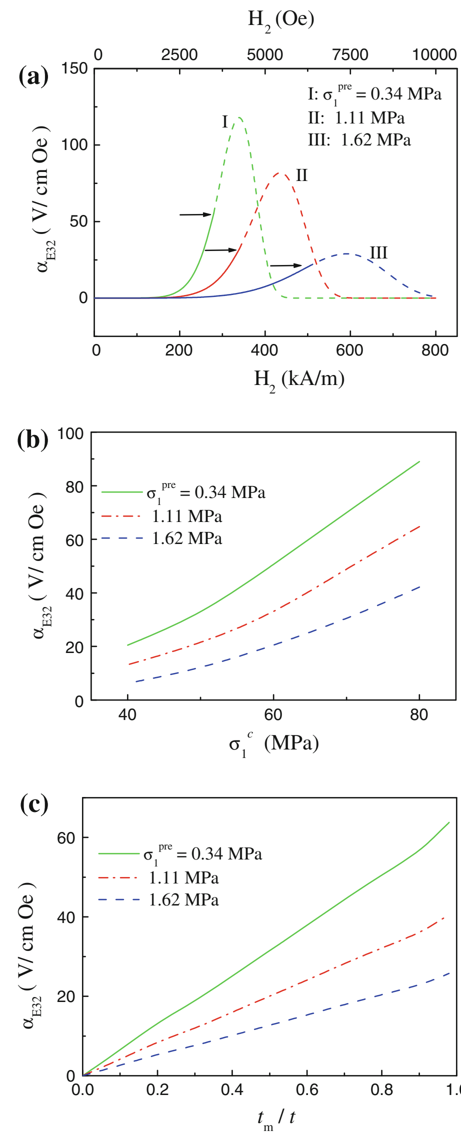

Fig. 2 **a** Dependence of the ME coefficient (α_{E32}) of the bilayer $Ni_{47.4}Mn_{32.1}Ga_{20.5}$/PZT composites on $\boldsymbol{H}$ as a function of σ_1^{pre} with and without considering the failure of the brittle PZT phase $\left(\sigma_1^c = 60\ \mathrm{MPa}\right)$, shown as solid curves and dash curves, respectively. The critical point marked with *arrow* in the curves means the occurrence of mechanical failure of PZT. **b** Dependence of α_{E32} of the bilayer $Ni_{47.4}Mn_{32.1}Ga_{20.5}$/PZT composite on critical strength of PZT (σ_1^c) as a function of σ_1^{pre}. **c** Dependence of α_{E32} on the relative thickness of $Ni_{47.4}Mn_{32.1}Ga_{20.5}$ layer ($\boldsymbol{t}_m/\boldsymbol{t}$) as a function of σ_1^{pre}

reported ME effects up to now. It is thus reasonable to expect that the $Ni_{47.4}Mn_{32.1}Ga_{20.5}$ alloys should be a kind of potential candidate for magnetostrictive phase in laminated multiferroic composites.

Figure 2c further shows the dependence of the ME coefficient on the relative thickness of the $Ni_{47.4}Mn_{32.1}Ga_{20.5}$ layer, $\boldsymbol{t}_m/\boldsymbol{t}$, under different pre-applied stress and under $\sigma_1^c = 60$ MPa. One can find that the ME coefficient increase monotonically with $\boldsymbol{t}_m/\boldsymbol{t}$, which is because thicker magnetostrictive layer could pass the strain to the thinner piezoelectric layer more effectively. This conclusion also indicates that the elastic coupling effect should be stronger at higher t_m/t. Previous calculations based on both Green's function technique [32] and simple constitutive equations [31] have yielded similar results on the relationship between ME coefficient and t_m/t.

Revealed from both Fig. 2b and c, the optimization of the laminated $Ni_{47.4}Mn_{32.1}Ga_{20.5}$/PZT composites is to thin the PZT layer down (thicken the $Ni_{47.4}Mn_{32.1}Ga_{20.5}$ layer) as well as to employ a stronger PZT material. This method is also applicable to all the other magnetostrictive/piezoelectric multiferroic composites, especially to those involving giant strain.

In summary, we have calculated the ME response of a new kind of $Ni_{47.4}Mn_{32.1}Ga_{20.5}$/PZT multiferroic composites based on a modified multi-field coupling equations, and investigated the influence of mechanical failure on ME response. It is theoretical revealed that the $Ni_{47.4}Mn_{32.1}Ga_{20.5}$/PZT bilayer composites could produce an ideal GME effect up to 120 V/cm Oe, which is much larger than the best reports up to now. However, the real ME response is strongly limited by the mechanical strength of the brittle PZT. Reducing the PZT layer and using a mechanically stronger PZT material have been suggested to enhance the ME response.

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