Enhancement of the nonlinear magnetoelectric effect in a ferromagnet-piezoelectric heterostructure due to nonlinearity of magnetization

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We describe theoretically and experimentally a previously unobserved mechanism for the induction of nonlinear magnetoelectric response in ferromagnet-ferroelectric multiferroic composites. We show that contributions to the nonlinear magnetoelectric effects come not only from the nonlinearity of the magnetostriction coefficient in respect with the dc magnetic field, but also from the nonlinear hysteretic dependence of the magnetization of the magnetic phase within the composite. The nonlinearity of the magnetization leads to the self-generation of an additional ac magnetic field oscillating at twice the frequency of the excitation field. In turn, this leads to the strain mediated activation of the piezo component, generating a voltage output response and doubling of its frequency relative to that of the excitation field. For the $PbZrTiO_3$ / FeBSiC test sample examined in this study, we determined that this mechanism is responsible for an additional contribution of ~14% to the nonlinear magnetoelectric effect.

Strain mediated magnetoelectric (ME) effects in planar multiferroic heterostructures containing mechanically coupled ferromagnetic (FM) and ferroelectric (FE) layers arise as a result of a combination of the magnetostriction of the FM layer and the piezoelectric effect in the FE layer [1]. When an alternating magnetic field is applied to the structure, the FM layer deforms due to magnetostriction. This deformation is then transferred to the coupled FE layer and, due to the piezoelectric effect, an electric voltage is generated (direct ME effect). Conversely, when an alternating electric field is applied to the FE layer, a deformation is again induced due to the inverse piezoelectric effect or electrostriction, which is then transferred to the FM layer resulting in magnetization changes due to the elastomagnetic effects (converse ME effect).

The ferroic order parameters of the composite are nonlinearly dependent on the applied control fields, which should lead to the occurrence of nonlinear dynamic ME effects, as the amplitudes of the excitation fields increase. Indeed, in the case of the direct ME effect, the generation of the second harmonic [2] and combination frequencies [3,4] in multiferroic structures were observed upon excitation with two magnetic fields. In the case of the converse ME effect, the generation of double frequency and combination frequencies were observed upon excitation of the structure with two electric fields [5]. Mixing of the magnetic and electric fields' frequencies was also observed experimentally [6].

Until now, it was believed that nonlinearity of the direct ME effect arises from the nonlinear dependence of the magnetostriction of the FM layer on the dc magnetic field $\lambda(H)$. Expanding the magnetostriction coefficient in a Taylor series near the working around the magnetic field, and keeping the second-order terms, a theory has been developed showing the relationship between the second harmonic amplitudes and combination frequencies and the excitation fields [7].

In this work, we show that a significant contribution to the nonlinear ME effects in composite structures is induced by the nonlinear field dependence of the magnetization of the FM layer, which was not previously considered. In the case of the direct ME effect, this dependence leads to the generation of an additional magnetic field oscillating at double the frequency of the excitation magnetic field, which contributes to the resulting amplitude of the nonlinear ME effect already present in the first order term of the magnetostriction expansion over the field.

In this study, we use the theory of nonlinear ME effects in composite structures developed in [7]. Let us consider the FM-FE structure, containing mechanically bonded FM and FE layers, in the "1-2" plane (see insert of Fig. 1). We assume an external dc magnetic field H and an excitation alternating field h_1, both applied in the plane of the structure along the "1" axis. The FE layer is poled along the "3" axis. For this experimental geometry, the amplitude of the voltage generated by the structure can be written as [8]:

$$u = Ad_{31}\lambda(H) \tag{1}$$

where A is a coefficient depending on the materials and dimensions of the constituent layers, and the orientation of the dc field H, d_{31} is the piezoelectric coefficient of the FE layer of the structure, $\lambda(H) = \lambda_{11}^{(1)}(H) + \lambda_{12}^{(1)}(H)$, $\lambda_{11}^{(1)}(H) = \partial\lambda_{11} / \partial H\big|_H$ and $\lambda_{12}^{(1)}(H) = \partial\lambda_{12} / \partial H\big|_H$ are the linear piezomagnetic coefficients of the FM layer. The magnetic field acting on the structure has the form $H = H_0 + h_1\cos(2\pi ft)$, where $h_1 << H_0$. Expanding $u(H)$ in a Taylor series up to the second order terms and substituting the field, we obtain:

$$u = Ad_{31}\lambda(H_0) + Ad_{31}\lambda^{(1)}(H_0)h_1\cos(2\pi ft) + \frac{1}{2}Ad_{31}\lambda^{(2)}(H_0)h_1^2\cos^2(2\pi ft) + .. \tag{2}$$

After rearranging the terms, we have:

$$u = Ad_{31}[\lambda(H_0) + \frac{1}{4}\lambda^{(2)}h_1^2] + Ad_{31}\lambda^{(1)}(H_0)h_1\cos(2\pi ft) + \frac{1}{4}Ad_{31}\lambda^{(2)}(H_0)h_1^2\cos(4\pi ft) + .. \tag{3}$$

where $\lambda^{(2)}(H) = \partial^2\lambda / \partial H^2\,|_H$ is the nonlinear piezomagnetic coefficient of the FM layer. The first term in (3) gives a constant voltage, which rapidly decreases due to leakage currents in the FE layer and will not be considered further. The second term describes the linear direct ME effect, i.e. generation of voltage with the frequency of the excitation magnetic field. The last term describes the nonlinear voltage frequency doubling. It can be seen that the amplitude of the second harmonic u_2 is proportional to the square of the excitation field amplitude h_1 and the field dependence of the harmonic amplitude is determined by the field dependence of the nonlinear piezomagnetic coefficient $\lambda^{(2)}(H)$.

We now take into account the nonlinear dependence of the magnetic induction of the FM layer on the magnetic field $B(H) = \mu_0(H + M) = \mu_0\mu H$, where $\mu_0 = 4\pi 10^{-7}$ H/m is the magnetic permeability of vacuum, and μ is the magnetic permeability of the FM layer. Expanding the induction in a Taylor series, keeping up to the second order terms, and substituting the excitation magnetic field, we obtain:

$$B = B(H_0) + \frac{1}{4}\mu_0\mu^{(1)}h_1^2 + \mu_0\mu h_1\cos(2\pi ft) + \frac{1}{2}\mu_0\mu^{(1)}h_1^2\cos(4\pi ft) + ... \tag{4}$$

where $\mu^{(1)}(H) = \partial\mu / \partial H\,|_H$ is the derivative of the magnetic permeability with respect to the field. From (4) we observe that the nonlinearity of the magnetic induction leads to the generation of an additional alternating magnetic field of double frequency in the FM layer. The amplitude of the second harmonic of the magnetic field is:

$$h_2 = \frac{1}{2}\frac{\mu^{(1)}}{\mu}h_1^2. \tag{5}$$

Thus, due to the nonlinearity of the magnetization of the FM layer, the excitation of the structure actually occurs via a dual frequency alternating field $h = h_1\cos(2\pi ft) + h_2\cos(4\pi ft)$. Substituting this field in (3) and discarding negligible terms, we obtain the amplitude of the second harmonic of the ME voltage generated by the structure:

$$u_2 = \frac{1}{4} A d_{31} \left[\lambda^{(2)} + \frac{2\mu^{(1)}}{\mu} \lambda^{(1)} \right] h_1^2 . \quad (6)$$

Therefore, considering the nonlinearity of the magnetization led to a renormalization of the nonlinear piezomagnetic coefficient:

$$\gamma = \lambda^{(2)} + \frac{2\mu^{(1)}}{\mu} \lambda^{(1)} , \quad (7)$$

which determines the magnitude of the nonlinear ME effect. Hence, taking into account the nonlinearity of the FM layer of the structure can change both the amplitude of the second harmonic and the field dependence of the amplitude, since the magnetic permeability μ and its derivative $\mu^{(1)}$ depend on the field H.

The ferromagnet-ferroelectric heterostructure studied here is shown schematically in Fig. 1. It contains a piezoelectric / ferroelectric PbZrTiO3 (PZT) plate, 20 mm × 5 mm in dimensions and 0.5 mm thick, with Ag-electrodes on the surfaces, poled perpendicular to the plane. The PZT's piezoelectric coefficient is $d_{31} \approx 175$ pC/N, and the dielectric constant $\varepsilon \approx 1750$. A layer of amorphous ferromagnet FeBSiC (Metglas 2605SA1, Metglas Co, USA), 20 mm x 5 mm lateral size and 25 µm thick, was glued to the plate's surface on one side using epoxy. The FeBSiC has saturation magnetization $M_S \approx 15$ kG and saturation magnetostriction $\lambda_S \approx 35\times10^{-6}$, corresponding to a field of $H_S \approx 100$ Oe.

The multiferroic composite structure is placed for testing in an electromagnetic coil, 6 mm in diameter and 60 mm long, containing 107 copper wire turns. An electric current was passed through the coil, creating an alternating magnetic field with an amplitude of up to 3 Oe and a frequency of f = 10 Hz - 100 kHz. A dc field H = 0 - 200 Oe was applied parallel to the long axis of the structure using a dc electromagnet. The voltage generated between the electrodes of the structure due to the ME effect was recorded. A resistor r = 8 Ω was connected in series with the coil to monitor the current. The measurements were carried out in an off-resonant mode at the magnetic field excitation frequency of f = 1.25 kHz, located far from the frequencies of the acoustic resonances of the structure.

Figure 1 shows the positive branch of the magnetization curve $M(H)$ of the FM layer of the structure, measured on a vibrating sample magnetometer with field applied along the long axis, and the dependence of magnetostriction on the field $\lambda(H)$ in the same geometry, measured via the tensometric method [9].

Firstly, the characteristics of the doubled frequency magnetic field, generated by the FM layer exclusively due to the nonlinear dependence $M(H)$, were found. For this purpose, using a narrow-band filter at 2.5 kHz frequency, the field dependence of the voltage $V(H)$ at the additional resistance was measured and the dependence $h_2(H)$ was calculated. The measured field dependence for the excitation field amplitude h_1 = 2.6 Oe is shown in Fig. 2. The data indicate that h_2 first grows almost linearly with increasing H, it reaches a maximum of $h_2 \approx 0.22$ Oe at a characteristic field $H_1 \approx 7$ Oe, and then tends to zero as the FM layer becomes saturated. The maximum amplitude of the second harmonic of the field in our case reached ~ 8% of the amplitude of the excitation field. It was found that the amplitude of the second harmonic increases quadratically with the excitation field, $h_2 \sim h_1^2$, in accordance with (5).

Figure 3 shows the field dependences of the magnetic permeability $\mu(H)$, its first derivative $\mu^{(1)}(H)$, the piezomagnetic coefficient $\lambda^{(1)}(H)$, and the nonlinear piezomagnetic coefficient $\lambda^{(2)}(H)$, calculated by direct numerical differentiation of the measured curves $M(H)$ and $\lambda(H)$, respectively. The dependences have a typical shape corresponding to an amorphous alloy. The magnetic permeability in zero field reaches $\mu \sim 10^3$, and its derivative has a minimum at the characteristic field $H_1 \approx 7$ Oe. It is important to notice that it is at this characteristic field H_1 where the maximum amplitude of the second harmonic of the field is observed (see Fig. 2).

The linear piezomagnetic coefficient $\lambda^{(1)}$ reaches a maximum at $H_m \approx 14$ Oe. The nonlinear piezomagnetic coefficient increases with increasing H and reaches a maximum at the field $H_1 \approx 7$ Oe, then vanishes and after that passes through the second maximum at around 20 Oe.

Figure 4a shows the measured field dependence of the second harmonic amplitude of the ME voltage $u_2(H)$, corresponding to the amplitude of the excitation magnetic field $h_1 = 1$ Oe and frequency $f = 1.25$ kHz. A closer inspection of the data reveals that: a) u_2 has a finite amplitude at $H = 0$; b) it grows with increasing the field; c) it reaches a maximum of 0.65 mV at the characteristic dc field $H_1 \approx 7$ Oe; d) it vanishes at $H_m \approx 14$ Oe and then passes through the second local maximum; e) finally, it tends to zero as the FM layer of the structure is magnetically saturated. The largest value of the nonlinear ME coefficient for the described structure was $\alpha_E^{(2)} = u_2/(a_p h^2) \approx 13$ mV/(cm Oe^2), which is consistent with other studies [10].

For comparison, Fig. 4b also shows the field dependence of the renormalized coefficient $\gamma(H)$, calculated using the data from Fig. 3, which determines the magnitude of the nonlinear ME effect. It is clearly seen that the shape of the $\gamma(H)$ curve closely traces the shape of the $u_2(H)$ dependence. It follows from the data presented here that the nonlinearity of magnetization leads to an increase in the nonlinear ME effect in the region of magnetic fields, where the FM layer of the composite structure generates a magnetic field with doubled frequency. For confirmation, let us pay attention to the ratio of the heights of the first and second maxima in the presented field dependences. For the nonlinear piezomagnetic coefficient (Fig. 3b), this ratio is 2.25, for the renormalized coefficient γ (Fig. 4b) the ratio is 3.0, and for the second harmonic amplitude u_2 (Fig. 4a) the ratio is 3.1. Our estimation, according to (6) and using data from Fig. 3 for a field H_1, gives a ~14% increase in γ with respect to $\lambda^{(2)}$, which is in good agreement with the experiment.

It should be added that the nonlinearity of magnetization will also contribute to the efficiency of generation of combination frequencies when the structure is excited by two magnetic fields [4]. The described mechanism should also lead to an increase in the nonlinear ME effects in particulate multiferroic composites with strong coupling between the magnetic and electric phases [11, 12]. An increase in the nonlinear ME effect due to the nonlinearity of magnetization in the resonant mode, when the frequency of the excitation field coincides with the frequency of the acoustic resonance of the composite structure, requires additional research.

Thus, the contribution of the magnetic nonlinearity of the FM layer to an increase in the nonlinear ME effect in heterostructures has been demonstrated theoretically and experimentally. It is shown that, due to the nonlinear dependence of the magnetization of the FM layer on the dc field, the FM layer of the structure self-generates a magnetic field oscillating at doubled frequency. Hence, the excitation of the structure occurs in fact via two fields. As a result, the amplitude of the second harmonic of the ME voltage increases in the region of magnetic fields, where the magnetic permeability of the FM layer has a minimum. The nonlinearity of the magnetization leads to an increase by up to 14% in the nonlinear ME effect of our structure containing an amorphous magnetic alloy.

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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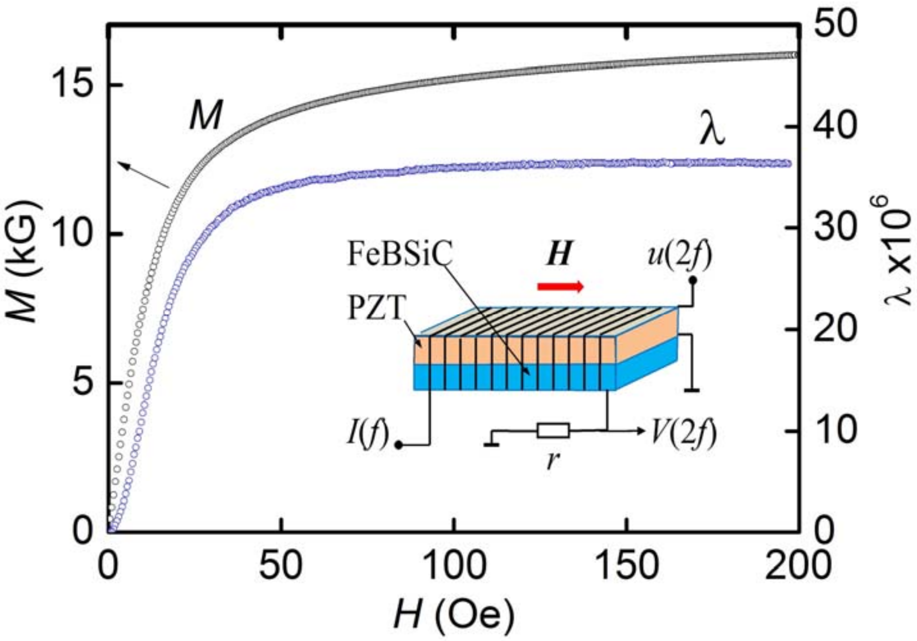

Fig.1 Dependences of the magnetization M and magnetostriction λ of the FeBSiC layer on the dc field H. The inset schematically shows the FeBSiC-PZT heterostructure.

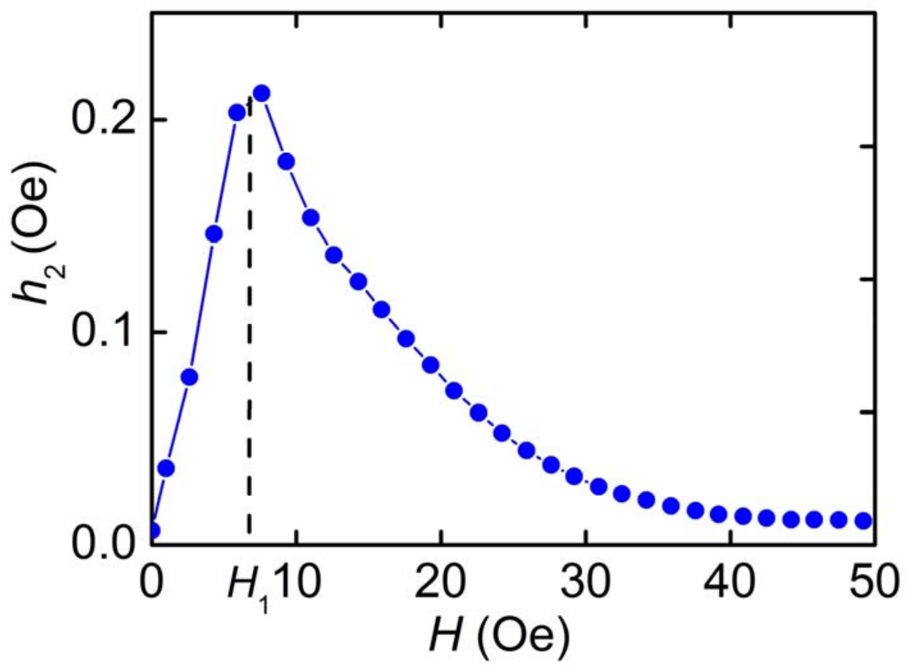

Fig. 2 Dependence of the amplitude of the field h_2 with ~~a~~ doubled frequency, generated by the FM layer due to the nonlinearity of the magnetization, on the dc magnetic field H, at h_1 = 2.6 Oe.

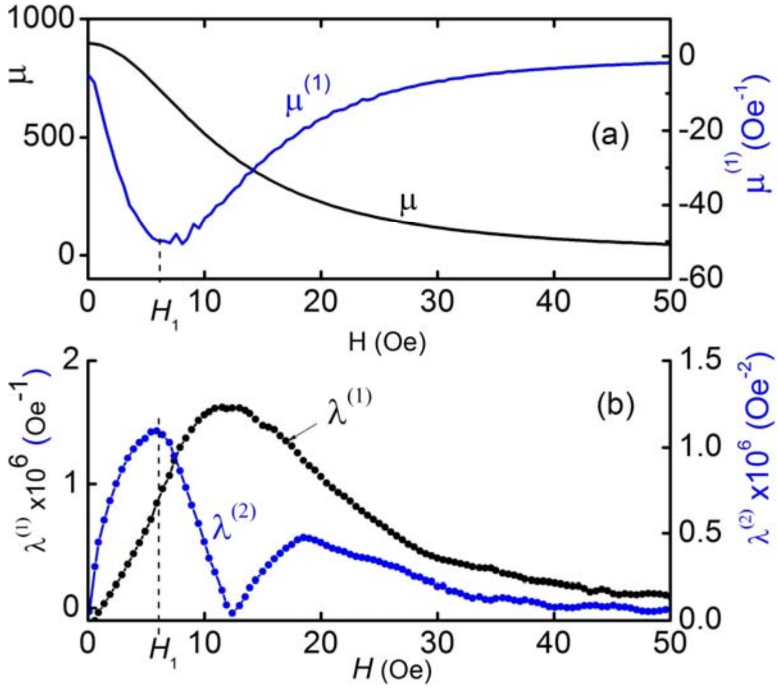

Fig. 3 (a) Dependences of magnetic permeability μ and its first derivative $\mu^{(1)}$, (b) piezomagnetic coefficient $\lambda^{(1)}$ and nonlinear piezomagnetic coefficient $\lambda^{(2)}$ versus the field *H* for the FeBSiC layer.

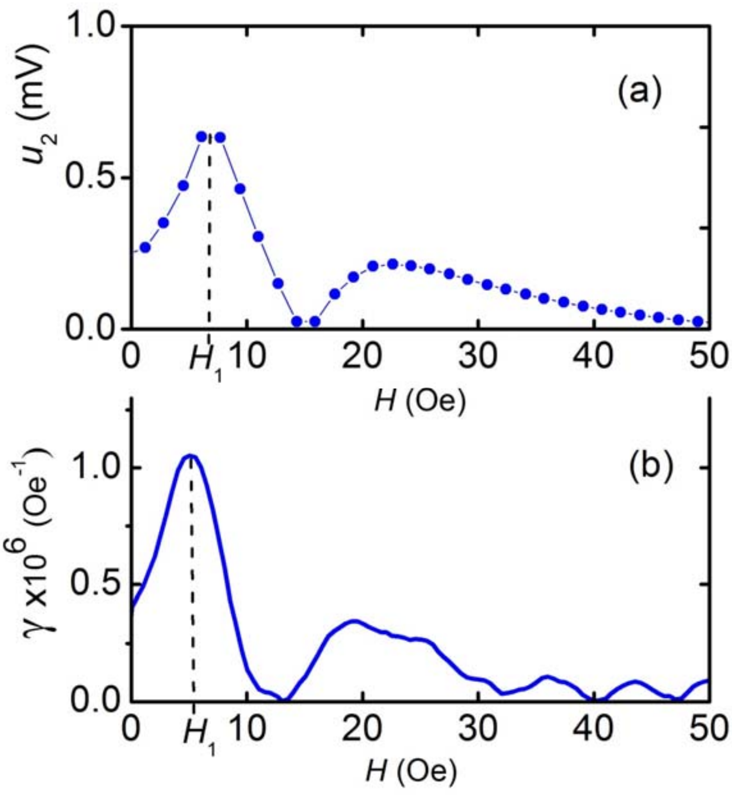

Fig. 4 (a) Dependences of the amplitude of the 2nd harmonic of the ME voltage u_2 and (b) the normalized coefficient γ on the magnetic field *H*.