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RESEARCH ARTICLE

Nonlinear magnetoelectric effect in a ring composite heterostructure

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Abstract
Objectives. The relevance of the study of magnetoelectric (ME) effect in ring ferromagnetic–piezoelectric heterostructures is due to the possibility of creating various ME devices having improved characteristics. A detailed investigation of the nonlinear ME effect in a ring composite heterostructure based on lead zirconate titanate (PZT) piezoceramics and Metglas® amorphous ferromagnetic (FM) alloy under circular magnetization is presented.
Methods. The ME effect was measured by the low-frequency magnetic field modulation method. Excitation alternating- and constant magnetic bias fields were created using toroidal coils wound on a ring heterostructure for circular magnetization of the FM layer.
Results. When excited with circular magnetic fields in a non-resonant mode, the ME ring heterostructure generates a nonlinear ME voltage of higher harmonics. The field and amplitude dependencies of the first three ME voltage harmonics were investigated. ME coefficients were obtained for the linear ME effect $\alpha^{(1)}$ = 5.2 mV/(Oe·cm), the nonlinear ME effect $\alpha^{(2)}$ = 6 mV/(Oe^2·cm), and $\alpha^{(3)}$ = 0.15 mV/(Oe^3·cm) at an excitation magnetic field frequency *f* = 1 kHz. The maximum amplitudes of the 1st and 3rd harmonics were observed at a constant bias magnetic field *H* ~ 7 Oe, which is almost two times smaller than in planar PZT–Metglas® heterostructures.
Conclusions. A nonlinear ME effect was observed and investigated in a ring heterostructure based on PZT piezoceramics and Metglas® amorphous FM alloy. Due to the absence of demagnetization during circular magnetization of the closed FM layer, nonlinear ME effects are detected at significantly lower amplitudes of the exciting alternating and constant bias magnetic fields as compared to planar heterostructures. The investigated ring heterostructures are of potential use in the creation of frequency multipliers.

Keywords: nonlinear magnetoelectric effect, composite heterostructure, magnetostriction, ferromagnet, piezoelectric effect

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НАУЧНАЯ СТАТЬЯ

Нелинейный магнитоэлектрический эффект в кольцевой композитной гетероструктуре

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Резюме
Цели. Актуальность исследования магнитоэлектрических (МЭ) характеристик кольцевых гетероструктур «ферромагнетик-пьезоэлектрик» обусловлена созданием на их основе МЭ-устройств с улучшенными характеристиками. Целью настоящей работы является детальное исследование нелинейного МЭ-эффекта в кольцевой композитной гетероструктуре на основе пьезокерамики цирконата-титаната свинца (ЦТС) и аморфного ферромагнитного (ФМ) сплава Metglas® при ее циркулярном намагничивании.
Методы. МЭ-эффект исследован методом низкочастотной модуляции магнитного поля. Возбуждающее переменное и постоянное магнитные поля смещения были созданы при помощи тороидальной катушки, намотанной на гетероструктуру, для циркулярного намагничивания ферромагнитного слоя.
Результаты. Обнаружен нелинейный МЭ-эффект, заключающийся в генерации высших гармоник МЭ-напряжения при возбуждении структуры циркулярными магнитными полями в нерезонансном режиме. Исследованы полевые и амплитудные зависимости первых трех гармоник МЭ-напряжения. Получены МЭ-коэффициенты для линейного МЭ-эффекта $\alpha^{(1)}$ = 5.2 мВ/(Э·см) и для нелинейного МЭ-эффекта $\alpha^{(2)}$ = 6 мВ/(Э^2·см) и $\alpha^{(3)}$ = 0.15 мВ/(Э^3·см) при частоте переменного магнитного поля f = 1 кГц. Максимумы амплитуд 1-й и 3-й гармоник наблюдались при постоянном магнитном поле H ~ 7 Э, что почти в два раза меньше, чем в планарных гетероструктурах ЦТС–Metglas®.
Выводы. Обнаружен и исследован нелинейный МЭ-эффект в кольцевой структуре на основе пьезокерамики ЦТС и аморфного ФМ-сплава Metglas®. Вследствие отсутствия размагничивания при циркулярном намагничивании замкнутого ФМ-слоя нелинейные МЭ-эффекты проявляются при значительно меньших амплитудах возбуждающего переменного и управляющего постоянного магнитных полей по сравнению с планарными гетероструктурами. Исследуемые кольцевые структуры могут быть использованы для создания на их основе умножителей частоты.

Ключевые слова: нелинейный магнитоэлектрический эффект, композитная гетероструктура, магнитострикция, ферромагнетик, пьезоэлектрический эффект

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INTRODUCTION

Magnetoelectric (ME) effects in composite multiferroic heterostructures containing mechanically coupled piezoelectric (PE) and ferromagnetic (FM) layers manifest themselves in the appearance of sample polarization when it is placed in an external magnetic field due to a combination of magnetostriction of the FM layer and the piezoelectric effect of the PE layer [1]. Such structures are of great interest due to their practical application for the creation of highly sensitive sensors of alternating and permanent magnetic fields, actuators, radio signal processing devices, transformers, etc. [2–5].

There is currently increasing interest in the study of ring composite heterostructures consisting of PE and FM layers. The characteristics of a linear ME effect observed in such heterostructures based on lead zirconate titanate (PZT) piezoceramics and Ni-based FM layers and amorphous Metglas® FM alloy when magnetized by an external field in the plane or along the ring axis have been studied [6–8]. Tunable transformers [9] and inductors [10], in which the inductance is tunable by 1000% under the action of constant magnetic and electric fields, were fabricated based on the ME ring composite heterostructures. A number of current sensor designs based on ring-ME structures have been proposed [11].

There is a wide interest in the study of nonlinear effects in ME heterostructures, such as the generation of harmonics and combinational frequencies, bistability, hysteresis suppression [12], forming the basis the design of devices for frequency multiplication and alternating magnetic field spectrum analysis. Due to the significant influence of demagnetization effects in the FM layer, the study of such effects in planar heterostructures requires constant and alternating magnetic fields of the order of units to tens of Oe.

At the same time, circular magnetization of ring heterostructures seems a promising avenue of enquiry [13]. By such means, it may be possible to achieve a significant decrease in demagnetization effects at the same time as increasing the efficiency of ME conversion at reduced bias magnetic fields due to the closed magnetic flux in the ring FM layer. In connection with the above, the study of nonlinear ME effects in ring heterostructures during circular magnetization is of great interest. To the best of the present authors' knowledge, the present work represents the first study into nonlinear ME effects in ring heterostructures excited by circular magnetic fields. Here, the nonlinear ME effect of voltage harmonics generation in a two-layer ring heterostructure consisting of PZT-19 piezoceramics and Metglas® tape of amorphous magnetic alloy under circular magnetization by alternating and constant magnetic field was observed and studied.

SAMPLES AND MEASURING METHODOLOGY

A schematic representation and an appearance of the investigated two-layer heterostructure is depicted in Fig. 1. The investigated heterostructure comprises a two-layer ring with an outer layer made of PZT-19 piezoceramic (NII ELPA, Russia), bounded with an inner FM layer made of Metglas® 2605SA1 amorphous magnetic alloy (Metglas® Inc., USA). The ends of the FM layer have been overlapped. The layers were joined using a cyanoacrylate adhesive at a thickness of ~3 μm (Weiss, CA-500.200, Germany). The overlap of the opposite ends of the amorphous tape was less than 1 mm. The FM layer had a length $L_m = 50.2$ mm, thickness $a_m \approx 27$ μm, width $w_m = 5$ mm, saturation magnetostriction $\lambda_S \approx 25 \cdot 10^{-6}$, and magnetic permeability $\mu \approx 10^4$. Radially polarized piezoceramic ring of $Pb(Zr_xTi_{1-x})O_3$ (PZT) composition had an inner diameter of 16 mm, thickness $a_p = 1$ mm and width $w_p = 5$ mm. Ag-electrodes were deposited to the outer and inner surfaces of the PZT ring. PZT-19 piezoceramics is characterized by piezoelectric modulus values $d_{31} = 175$ pC/N and relative dielectric permeability $\varepsilon = 1750$.

Two toroidal coils of copper wire of 0.2 mm diameter having a number of evenly distributed windings $N = 90$ are wound on the ring. One coil generates a circular bias constant magnetic field $H = 0$–115 Oe by passing through it a current of $I_{dc} = 0$–5 A from an AKTAKOM APS-7305 supply source (AKTAKOM, Russia). A circular alternating magnetic field $h\cos(2\pi ft)$ having an amplitude up to $h = 3.45$ Oe and frequency $f = 0$–100 kHz was created by the second coil connected to an Agilent 33210A waveform generator (Agilent Technologies Inc., USA). The amplitude of the magnetic field was determined analytically using the formula $H = \dfrac{NI}{2\pi r}$, where r is the middle line of the toroidal coil,

N is the number of windings, and I is the current passing through the coil. Frequency spectra of the ME voltage were recorded using a Tektronix TDS3032B digital oscilloscope (Tektronix Inc., USA).

During the course of the study, voltage (u) dependencies and Fourier spectra were obtained at different excitation amplitudes (h) and constant magnetic fields (H), from which the field and amplitude dependencies of the 1st, 2nd, and 3rd harmonics of output ME voltage were obtained.

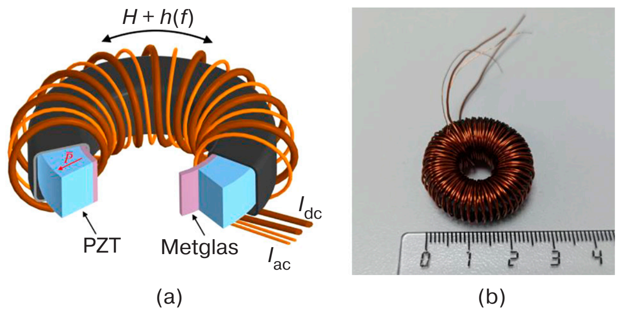


Fig. 1. Schematic representation (a) and appearance (b) of a PZT–Metglas® ring heterostructure with toroidal coils

RESULTS

At excitation of the structure by an alternating magnetic field of amplitude h = 0–3.45 Oe with a frequency f_1 = 1 kHz in a circular constant field H = 0–115 Oe, a nonlinear effect of ME voltage harmonics excitation was detected. The measurements were carried out away from the resonant frequency of the structure $f_0 \approx 54.2$ kHz. In the spectrum measured at $H = 0.45$ Oe and $h = 3.45$ Oe (Fig. 2), sixteen peaks are observed corresponding to the ME voltage harmonics with frequencies $f_n = f_1 \cdot n$, where n is an integer. The amplitudes of the first three peaks were $u^{(1)}$ = 1.8 mV, $u^{(2)}$ = 7.15 mV, and $u^{(3)}$ = 0.6 mV, respectively.

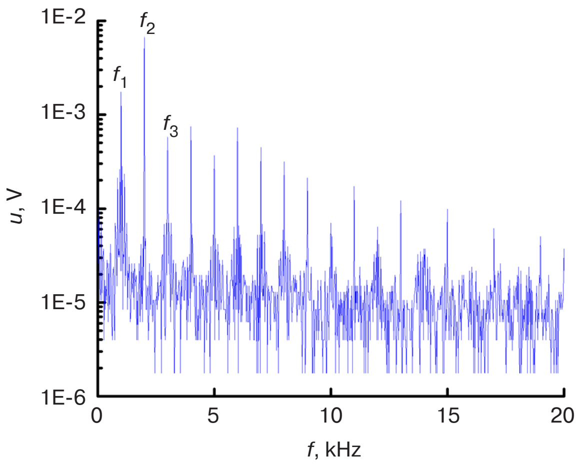


Fig. 2. Fourier spectrum of ring heterostructure ME voltage at h = 3.45 Oe with a frequency f_1 = 1 kHz and H = 0.45 Oe

Values of ME coefficients of the 1st, 2nd, and 3rd harmonics can be estimated as $\alpha^{(n)} = u^{(n)}/(h^n a_p)$. Hence, $\alpha^{(1)}$ = 5.2 mV/(Oe·cm), $\alpha^{(2)}$ = 6 mV/(Oe^2·cm), and $\alpha^{(3)}$ = 0.15 mV/(Oe^3·cm), respectively. In the absence of a constant magnetic field, only even harmonics were observed. These harmonics can be attributed to the nonlinear dependence of the magnetostriction $\lambda(H)$ of the FM layer on the constant magnetic field H [13].

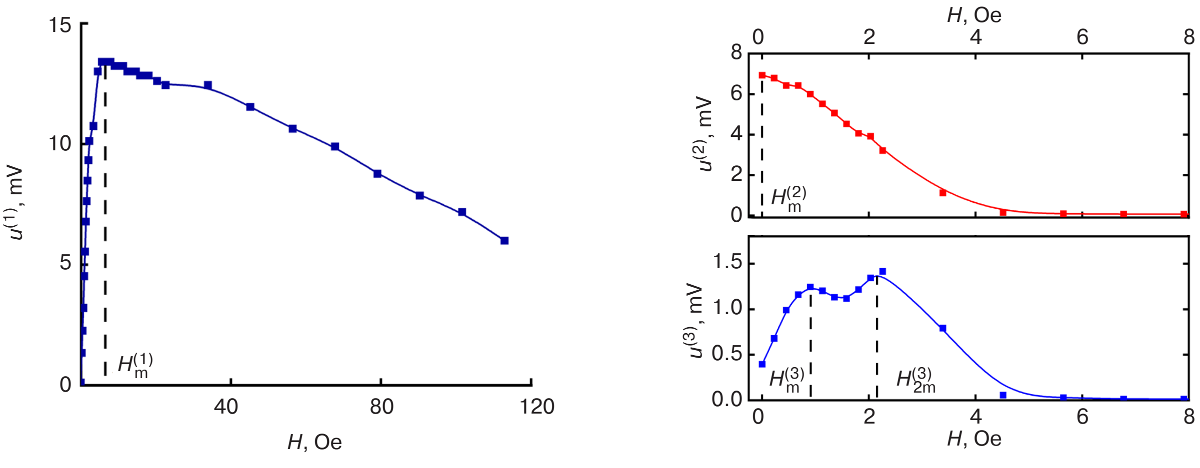
Figure 3 shows the dependencies of the amplitudes of the 1st, 2nd, and 3rd harmonics on the constant magnetic field H at h = 3.45 Oe based on the obtained Fourier spectra of the ME voltage. The shape of curve $u^{(1)}(H)$ for the 1st harmonic (Fig. 3a) is typical for the linear ME effect. The amplitude maximum $u^{(1)}_{\max} = 13.4$ mV achieved in the field $H^{(1)}_{\mathrm{m}} \approx 6.8$ Oe followed by a monotonic decrease as the constant magnetic field increases up to $H = 115$ Oe. The maximum ME voltage corresponds to the highest value of the piezomagnetic coefficient of the FM layer $\lambda^{(1)}(H) = \partial\lambda / \partial H|_H$, where $\lambda(H)$ is the field dependence of the magnetostriction of the FM layer.

Figure 3b shows the dependencies of the amplitudes of the 2nd $u^{(2)}(H)$ and 3rd $u^{(3)}(H)$ harmonics on the constant magnetic field H. The amplitude of the second harmonic $u^{(2)}_{\max} = 7.15$ mV is highest at $H^{(2)}_{\mathrm{m}} = 0$, and then monotonically decreases with increasing field to reach a minimum $u^{(2)}_{\min} = 0.1$ mV at $H = H^{(2)}_{\mathrm{m}} \approx 5.6$ Oe. Note that the graph $u^{(2)}(H)$ does not show a local minimum or subsequent local maximum typical for the field dependence of the 2nd harmonic in planar heterostructures [14, 15].

The amplitude of the 3rd harmonic at $H = 0$ has a value $u^{(3)} \approx 0.4$ mV. When the magnetic field increases, two local maxima $u^{(3)}_{1\max} = 1.2$ mV and $u^{(3)}_{2\max} = 1.4$ mV are observed in the fields $H^{(3)}_{1\mathrm{m}} \approx 0.9$ Oe and $H^{(3)}_{2\mathrm{m}} \approx 1.4$ Oe, respectively. Subsequently, the amplitude of the 3rd harmonic monotonically decreases to zero at $H \sim 5.6$ Oe.

Figure 4 shows the dependencies of the ME voltage amplitudes of the 1st, 2nd, and 3rd harmonics on the amplitude of the excitation alternating magnetic field h. Measurements were performed in the optimal magnetic fields H_{m} obtained from the curves shown in Fig. 3: $H^{(1)}_{\mathrm{m}} \approx 6.8$ Oe, $H^{(2)}_{\mathrm{m}} = 0$ Oe, and $H^{(3)}_{\mathrm{m}} = 1.4$ Oe, respectively.

The amplitude of the 1st harmonic can be seen to linearly depend on the value of h throughout the range of amplitudes of alternating magnetic fields h = 0–3.45 Oe. The amplitude of the 2nd harmonic increased in proportion to h^2 across the entire range while the amplitude of the 3rd harmonic is proportional to h^3. This type of dependence corresponds to theoretical calculations of the ME voltage $u^{(n)} \sim h^n$ [14], where n is the sequence number of the harmonic; h is the amplitude of the alternating magnetic field.



(a) (b)

Fig. 3. Dependencies of ME voltage amplitudes of the 1st (a), 2nd, and 3rd (b) harmonics of the ring heterostructure ME voltage on the constant magnetic field *H*

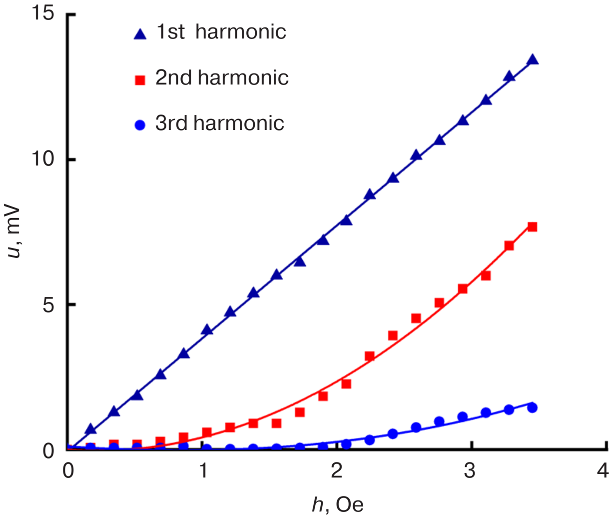


Fig. 4. Dependencies of the ME voltage amplitudes of the 1st, 2nd, and 3rd harmonics of the ME voltage on the field amplitude *h* at $H_m^{(1)} = 6.8$ Oe, $H_m^{(2)} = 0$ Oe, and $H_m^{(3)} = 1.4$ Oe, respectively. The solid lines are the power-law approximation

DISCUSSION

The appearance of the ME voltage harmonics is due to the nonlinear dependence of the magnetostriction of the FM layer on the magnetic field. By decomposing the function describing the magnetostriction $\lambda(H)$ into a Taylor series in the vicinity of the field *H* at $H << h$, we obtain the equation:

$$\lambda(H) = \lambda(H_0) + qh + \frac{ph^2}{2} + \frac{mh^3}{4}..., \quad (1)$$

where *q*, *p*, and *m* are the 1st, 2nd, and 3rd derivatives of magnetostriction by magnetic field, respectively.

The expression for the ME voltage can be written in the following form

$$u(H) = Ad\lambda(H), \quad (2)$$

where *A* is the coefficient that depends on the parameters of the PE and FM layers and the method of its constraint; *d* is the piezoelectric modulus module of the PE layer.

Substituting Eq. (1) in Eq. (2) and taking into account that the alternating magnetic field is given as $h = h_0 \cos(2\pi ft)$, we obtain:

$$u(H) = u_0(H_0) + u_1 \cos(2\pi ft) + \\ + u_2 \cos(4\pi ft) + u_3 \cos(6\pi ft).... \quad (3)$$

The first term in (3) denotes the constant component of the ME voltage, the second term denotes the generated ME voltage at the excitation field frequency, and the third and fourth terms describe the generation of the 2nd and 3rd harmonics of the ME voltage, respectively [16].

To the authors' knowledge, there are currently no published works in which multiple harmonics of higher orders have been observed in a planar heterostructure of similar composition. Although ~100 harmonics were previously observed in the langatate–Metglas® heterostructure [17], these occurred at a larger pumping amplitude $h = 20–25$ Oe of the alternating field.

Some peculiarities can be noted in the results of the study of the nonlinear MPE effect in the FM–PE ring structure. In contrast to ring structures, the magnetic field inside the FM layer H_{in} in planar structures is connected with the external field *H* by the relation $H_{in} = \frac{H}{1 + N\mu}$, where *N* is the demagnetization factor and μ is the magnetic permeability of the FM layer [18].

Hence, it follows that demagnetization reduces both the constant and the excitation alternating field inside the FM layer in a planar heterostructure. In a ring heterostructure with circular magnetization, the magnetic flux in the FM layer is closed ($N \approx 0$). Therefore, inside the FM layer $H_{in} \approx H$ and $h_{in} \approx h$, which leads to a change in the shape of the magnetostriction dependence on the external field $\lambda(H)$, in particular, to a decrease in the magnetostriction saturation field.

In the studied structure (Fig. 3), the maximum of the 1st harmonic was observed in the field $H_m^{(1)} \approx 6.8$ Oe. Amplitude $u^{(1)}$ reached 90% of the maximum in the field ~4 Oe, which is several times less than in planar PZT–Metglas® heterostructures [19, 20].

Due to the absence of demagnetization in the ring heterostructure, the nonlinear ME effects appear at lower amplitudes of the external excitation field h. Apparently, this is associated with a change in the shape of the $\lambda(H)$ dependence in the ring structure. In the dependence plot of the amplitude of the 2nd harmonic, the local maximum has disappeared, while in the dependence plot of the 3rd harmonic, it remains [17].

Values of ME coefficients of the 2nd and 3rd harmonics can be estimated as $\alpha^{(2)} = u^{(2)}/(h^2 a_p) = 6$ mV/(Oe^2·cm) and $\alpha^{(3)} = u^{(3)}/(h^3 a_p) = 0.34$ mV/(Oe^3·cm), respectively. The obtained ME coefficient values are comparable to the ME coefficients in the PZT–Metglas® planar structure of similar composition $\alpha^{(2)} = 9.6$ mV/(Oe^2·cm) and $\alpha^{(3)} = 0.4$ mV/(Oe^3·cm) measured at the same excitation field frequency [16].

CONCLUSIONS

In the present work, a nonlinear ME effect of the generation of higher harmonics of the ME voltage was observed when a ring two-layered PZT–Metglas® heterostructure is excited by circular magnetic fields in non-resonant mode. In the course of the study, we observed sixteen harmonics of the ME voltage in nonlinear mode when the structure is excited by an alternating magnetic field with a frequency of 1 kHz. Nonlinear ME effects in the ring heterostructure were detected in smaller magnetic fields compared to planar structures; this is associated with almost zero demagnetization of the FM layer. In particular, the maximum amplitudes of the 1st and 3rd harmonics were observed at $H \sim 7$ Oe, which is almost half as much as in planar PZT–Metglas® heterostructures.

The obtained results based on the investigated ring ME heterostructures demonstrate the possibility of creating efficient solid-state frequency multipliers excited by alternating magnetic fields of the order of oersted units.

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Authors' contributions

V.I. Musatov—preparing test samples and experimental setup, conducting research, data curation, preparing graphic information, and writing the text of the article.

F.A. Fedulov—preparing test samples and experimental setup, data checking, and writing the text of the article.

D.V. Savelev—conducting research, data curation, description of the research methodology.

E.V. Bolotina—conducting research, preparing graphic information.

L.Y. Fetisov—data checking and writing the text of the article.

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