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# Nonlinear magnetoelectric effect in a ferromagnetic-piezoelectric structure induced by rotating magnetic field

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## Abstract

The magnetoelectric (ME) effect induced by a rotating magnetic field,  $h$ , in the presence of a dc magnetic field,  $H_0$ , is investigated in a disk-shaped ferromagnetic FeBSiC - piezoelectric lead zirconate titanate bilayer structure. It is found that, due to the nonlinear field-dependence of magnetostriction  $\lambda(H)$  in the ferromagnetic layer, voltage harmonics are generated. These harmonics have a specific dependence of their amplitude and phase on  $H_0$  and  $h$ , which is different from the case of excitation with a linearly polarized field. A theory is developed that describes characteristics of the ME effect for the cases of weak  $h \ll H_0$  and strong  $h \gg H_0$  excitation fields. The effect can be employed in designing highly sensitive sensors of permanent and alternating magnetic fields.

**Keywords:** magnetoelectric effect, magnetostriction, piezoelectricity, composite structure, harmonics generation

## 1. Introduction

The magnetoelectric (ME) effect that performs mutual conversion between electric and magnetic fields in single-phase and composite multiferroic structures, has been intensively studied in recent years in connection with its use in highly sensitive sensors of magnetic field, magnetic memory elements, novel radio-signal processing devices, and autonomous power sources [1-4]. In artificially fabricated planar structures containing alternate ferromagnetic (FM) and piezoelectric (PE) layers, the ME effect arises due to the combination of magnetostriction in the FM layers and piezoelectricity in the PE layers that interact with each other through mechanical coupling between the layers [5-6]. When a variable magnetic field  $h(f)$  and a dc bias magnetic field  $H_0$  simultaneously applied to the structure, magnetostriction causes a deformation of the FM layers. This deformation is

transferred to the PE layers producing an alternating voltage  $u(f)$  there. Quantitative characteristics of the ME effects in a multilayer structure depend on the materials of the layers and their dimensions [6-8]. They can be altered by applying a dc magnetic field  $H_0$  to the structure [9] or a dc electric field  $E_0$  to the PE layer [10].

To date, mainly linear characteristics of the ME effect at small excitation fields  $h \ll H_0$  in structures of various shapes (rectangles, disks, rings, cylinders) have been investigated [6, 11-13]. Practically all studies were performed for parallel alignment of the excitation and dc bias fields, i.e.  $h \parallel H_0$ . At the same time, it was shown that orientation of the excitation field  $h$  with respect to  $H_0$  significantly affects the efficiency of the ME conversion in composite structures [14, 15]. The ME effect was observed in a planar rectangular FM-PE structure that was rotated in an external dc magnetic field [16]. The ME effect was considered theoretically in a two-

layer FM-PE cylinder excited by an annular alternating field [17].

It was also shown that an increase in the amplitude of the ac excitation field  $h$  gives rise to nonlinear effects, such as generation of voltage harmonics [18], mixing of magnetic fields frequencies [19-21], suppression of the ME effect hysteresis [22], and occurrence of bistability [23,24].

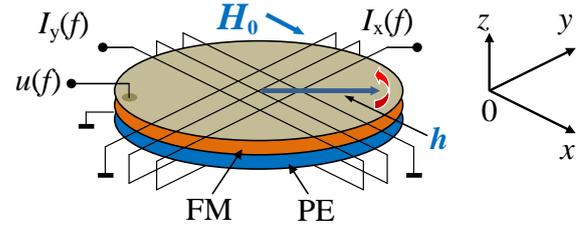
It makes interesting to study characteristic features of the ME effect for the case of a rotating excitation magnetic field. Such a field with an amplitude of tens of oersted and rotational frequency of up to tens of kHz, can be produced using two coils with orthogonal axes. Using a rotating field of constant magnitude, rather than a linearly polarized excitation field, can give certain advantages. For example, this would make it possible to investigate the anisotropy of the ME effect in the structure plane. Also, it can be used to reduce noise of ME sensors of magnetic fields, because magnetic domains will disappear when the magnitude of the rotating magnetic field is large enough to magnetically saturate the FM layer [25].

In this work, the ME effect in a planar ferromagnetic-piezoelectric disk structure induced by a rotating field  $h$  in presence of a dc tangential field  $H_0$  was investigated. The first part of the work describes the sample structure, geometry of the magnetic fields and the measurement technique. The second part presents experimental characteristics of the ME effect for various magnitudes of the excitation and dc bias magnetic fields. The third part outlines a simple theory and discusses the specific features of the ME effect when it is induced by a rotating magnetic field. The conclusion summarizes results of the research.

## 2. Sample and measurements

The structure under study is shown schematically in Fig. 1. It contains an FM disk of diameter  $R = 16$  mm and thickness  $a_m = 23$   $\mu\text{m}$ , and a PE disk of diameter 16 mm and thickness  $a_p = 200$   $\mu\text{m}$ . The FM disk was cut from a ribbon of an amorphous ferromagnetic alloy of composition FeBSiC (Metglas 2605 SA1) and had the saturation magnetostriction  $\lambda_s \approx 21 \cdot 10^{-6}$  in the saturation field  $H_s \sim 100$  Oe. To reduce the in-plane magnetic anisotropy, the FM disk was annealed at 300  $^\circ\text{C}$  and then slowly cooled in air in an alternating magnetic field. The PE disk was made of lead zirconate titanate ceramics of composition  $\text{Pb}_{0.52}\text{Zr}_{0.48}\text{TiO}_3$  (PZT) that had the piezomodule  $d_{31} = 175$  pC/N and the dielectric constant  $\epsilon = 1750$ . The PE disk was coated with 2  $\mu\text{m}$  thick Ag electrodes. The FM and PE discs were mechanically bonded to each other using a quick-drying epoxy.

The composite disk was placed inside two mutually orthogonal flat electromagnetic coils, whose axes were parallel to the sample plane. The inner coil of cross-section 1



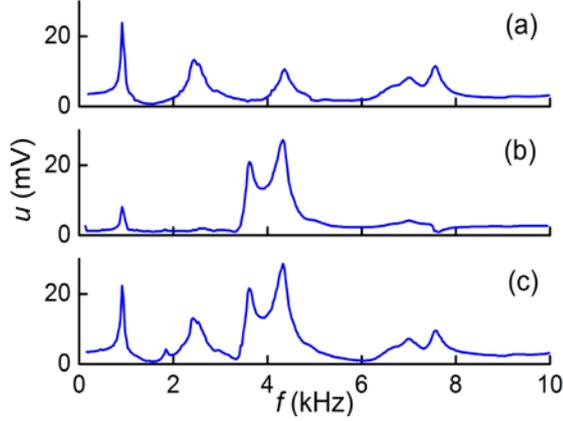
**Figure 1.** Geometry of the ferromagnetic-piezoelectric disk structure.

mm x 18 mm and 18 mm long had 200 turns of 0.2 mm thick wire, while the outer coil of cross-section 4 mm x 22 mm and length 22 mm contained 240 turns of 0.2 mm thick wire.

The internal coil, fed by sinusoidal current  $I_x(f_x)$  of frequency  $f_x = 10$  Hz-10 kHz, generated an alternating magnetic field  $h_x \cos(2\pi f_x t)$  of amplitude  $h_x = 0-50$  Oe. The external coil driven by current  $I_y(f_y)$  of frequency  $f_y = 10$  Hz-10 kHz produced an alternating field  $h_y \cos(2\pi f_y t)$  of amplitude  $h_y = 0-50$  Oe. The feeding currents were synchronized so that the phase difference between them was  $90^\circ$ . When the frequencies and amplitudes of the fields were equal, the coils produced a field of constant magnitude  $h = h_x = h_y$ , rotating with frequency  $f = f_x = f_y$  in the sample plane (i.e. circularly polarized, or, in other words, rotating excitation field). At  $f_x = f_y = f$  and  $h_x \neq h_y$  the coils produced elliptically polarized excitation field. During measurements the excitation coils with the sample inside were placed between two Helmholtz coils producing a dc magnetic field  $H_0 = 0-100$  Oe aligned parallel to the sample plane along the  $x$ -axis. The field was measured with a LakeShore 421 Gaussmeter with an accuracy of 0.1 Oe. The ac voltage  $u$  generated by the sample was measured using a digital oscilloscope TDS 3032B and its frequency spectrum was evaluated using a low-frequency SR770 FFT Network Analyzer.

## 3. Experimental results

Figure 2 shows typical dependences of the generated ME voltage  $u$  on frequency  $f$  (amplitude-frequency response) when the structure was excited by an ac magnetic field of amplitude  $h = 1.5$  Oe for three different orientations:  $h // H_0$ ,  $h \perp H_0$ , and rotating  $h$ . It is seen that frequency dependence of the ME response significantly depends on orientation of the excitation field. In the case of  $h // H_0$  (Fig. 2a), there is a low-frequency peak near  $f \approx 0.93$  kHz of amplitude  $u \approx 25$  mV and with quality factor  $Q \approx 13$ , two peaks at frequencies  $\sim 2.45$  kHz and  $\sim 4.36$  kHz, and a wide peak in the frequency region of  $\sim 6-8$  kHz. In the case of  $h \perp H_0$  (Fig. 2b), a weaker low-frequency peak and two clearly resolved peaks near the frequency  $\sim 4$  kHz can be



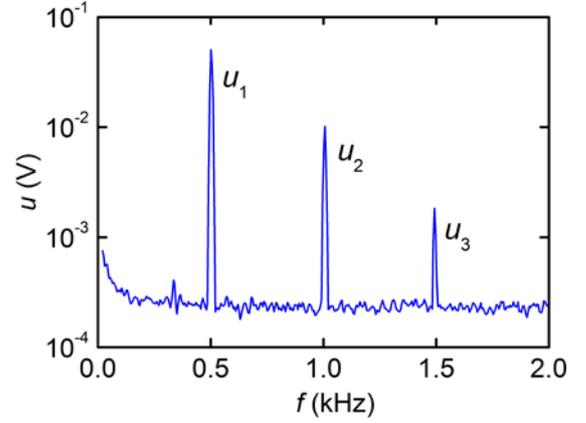
**Figure 2.** Dependences of ME voltage  $u$  on frequency  $f$  of excitation field for different orientations of magnetic fields: (a)  $h // H_0$ , (b)  $h \perp H_0$ , and (c) rotating field  $h$ .

seen. If the structure is excited by the rotating field (Fig. 2c), the frequency response is the sum of the responses corresponding to the two orthogonal linear polarizations.

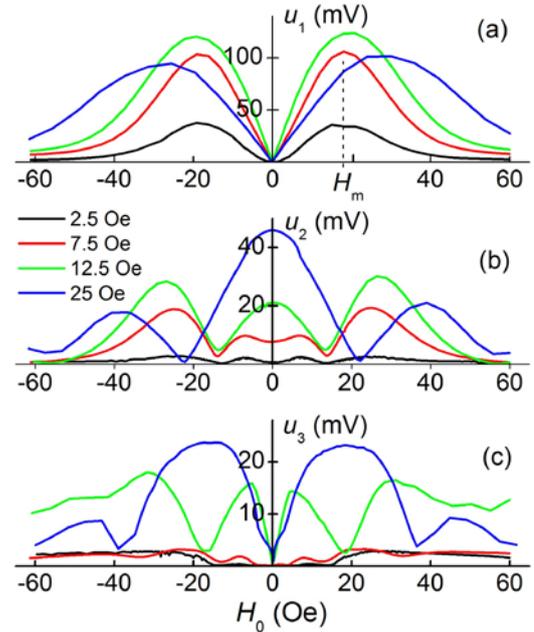
Estimation of the normal mode frequency for a free-standing thin disk with the same geometrical parameters as in our experiments, using formulas from Ref. 26, gave  $f_{cal} \approx 3$  kHz, which is close to the measured values. This corresponds to the excitation of the lowest mode of flexural vibrations in the composite disk. All further measurements were carried out in an off-resonant regime at the excitation frequency  $f = 500$  Hz for different amplitudes of the excitation field  $h$  and different  $H_0$ . The frequency of the excitation field was chosen from the condition that it and its harmonics do not coincide with any acoustic resonance in the structure.

Figure 3 shows a typical spectrum of the voltage generated by the composite structure when it is excited by a rotating field of frequency  $f = 500$  Hz and amplitude  $h = 7.5$  Oe for dc bias field  $H_0 = 20$  Oe. Apart from the fundamental harmonic of amplitude  $u_1$  at 500 Hz, which is equal to the frequency of the excitation field, the spectrum contains the 2<sup>d</sup> harmonic of amplitude  $u_2$  and the 3<sup>rd</sup> harmonic of amplitude  $u_3$ , signifying a high nonlinearity of the ME effect in the described structure.

Figure 4 shows the measured dependences of harmonics' amplitudes on bias magnetic field  $H_0$  for different amplitudes of the rotating excitation field  $h$ . The amplitude of the fundamental harmonic  $u_1(H_0)$  with increasing  $H_0$  behaves in approximately the same way as when the structure is excited by a unidirectional ac field of amplitude  $h$ . Initially  $u_1$  linearly grows from zero with increasing  $H_0$ , reaching its maximum at  $H_m$ , and then asymptotically tends to zero as the magnetostriction of the FM layer saturates. When  $h \ll H_0$ , the field  $H_m$  corresponds to the maximum of the linear piezomagnetic coefficient of the FM layer  $\lambda^{(1)}(H) = \partial \lambda / \partial H$ , where  $\lambda(H)$  is its field dependent



**Figure 3.** Frequency spectrum of ME voltage excited by rotating magnetic field  $h = 7.5$  Oe of frequency  $f = 500$  Hz.



**Figure 4.** Dependences of harmonic amplitudes  $u_1$ ,  $u_2$ , and  $u_3$  on bias field  $H_0$  for different magnitudes of the rotating excitation field  $h = 2.5, 7.5, 12.5,$  and  $25$  Oe.

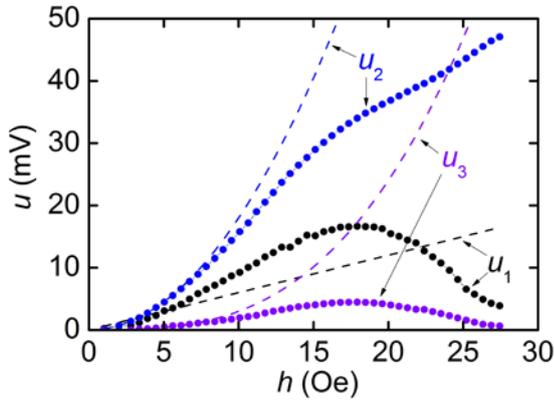
magnetostriction. As  $h$  increases, the harmonic amplitude at the maximum increases and  $H_m$  shifts to larger fields, in agreement with Ref. 21. The efficiency of the linear ME conversion at the optimal bias field  $H_m \approx 18$  Oe and  $h \approx 2.5$  Oe was found to be  $\alpha_E^{(1)} = u / (h \cdot a_m) \approx 700$  mV/(cm·Oe).

In contrast, field dependence of the 2<sup>d</sup> harmonic amplitude  $u_2(H_0)$ , is qualitatively different from that in the case of excitation with a linearly polarized ac field  $h$  (see Fig. 4b). At low  $h = 2.5$  Oe,  $u_2$  is zero at  $H_0 = 0$ , then it increases, reaching a local maximum at  $H_0 \sim 7.5$  Oe. After that it drops again to zero at  $H \approx H_m$ , then reaches the second local

maximum at  $H_0 \sim 26$  Oe, and finally asymptotically decreases to zero. As  $h$  increases to 12.5 Oe,  $u_2$  at  $H_0=0$  increases too, and the heights of the side maxima grow as well. The characteristic fields corresponding to the lateral minima increase. Only at largest excitation field  $h = 25$  Oe, the dependence  $u_2(H_0)$  becomes qualitatively the same as when the structure is excited by a linearly polarized ac field:  $u_2$  is maximum at  $H_0 = 0$ , drops to zero at  $H_0 \sim H_m$ , then reaches a local maximum, and after that tends to zero. For large  $h$ , the field dependence of the 2<sup>d</sup> harmonic  $u_2(H_0)$  qualitatively replicates the field dependence of the nonlinear piezomagnetic coefficient  $\lambda^{(2)}(H) = \partial^2 \lambda / \partial H^2$ . The maximum efficiency of the 2<sup>d</sup> harmonic generation at  $h = 12.5$  Oe and  $H_0 = 27$  Oe was  $\alpha_E^{(2)} = u / (a_p h^2) \approx 9.6$  mV/(cm·Oe<sup>2</sup>).

Behaviour of the 3<sup>rd</sup> harmonic amplitude  $u_3(H_0)$  (see Fig. 4c) is even more complicated. For rotating excitation fields with magnitudes in the range of 2.5 - 25 Oe,  $u_3(0)$  is equal to zero at  $H_0=0$ . Also in this case the characteristic fields of local minima and maxima move to higher  $H_0$  with increasing  $h$ . The maximum efficiency of the 3<sup>rd</sup> harmonic generation for  $h=12.5$  Oe and  $H_0=30$  Oe was  $\alpha_E^{(3)} = u / (a_p h^3) \approx 0.4$  mV/(cm·Oe<sup>3</sup>).

Figure 5 shows, as an example, dependences of amplitudes of the first three harmonics of the ME voltage on magnitude  $h$  of the rotating excitation field at dc bias field  $H_0 = 1$  Oe. The dashed curves in the figure show approximations of the experimental dependencies by power

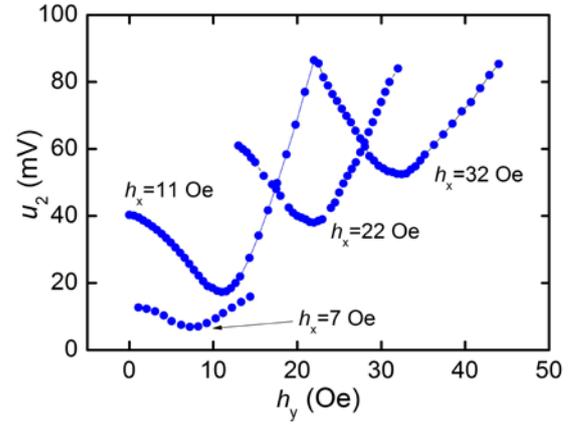


**Figure 5.** Harmonic amplitudes as a function of magnitude of the rotating excitation field at  $H_0 = 1$  Oe. The dots indicate experimental data while the dashed lines show approximations by power functions.

functions  $u_n \sim h^n$ , where  $n = 1, 2, 3$ . It can be seen that in the region of small fields  $h < 7$  Oe, the relations  $u_1 \sim h$ ,  $u_2 \sim h^2$ , and  $u_3 \sim h^3$  are fulfilled. For larger  $h$ , the amplitudes of all harmonics become saturated, and the amplitudes of the 1<sup>st</sup> and 3<sup>rd</sup> harmonics even fall.

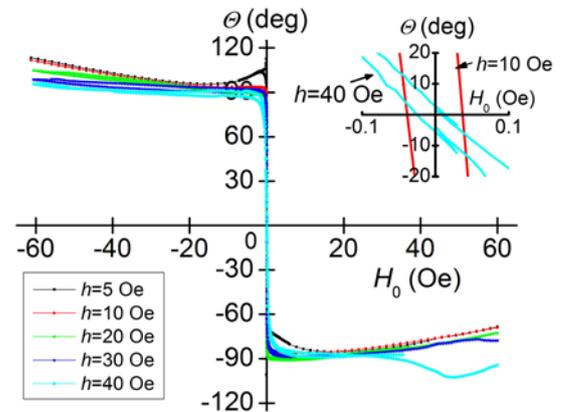
Figure 6 demonstrates how ellipticity of the excitation field affects the amplitude of the generated ME voltage. Here, as an example, the measured dependences of the 2<sup>d</sup> harmonic amplitude  $u_2$  on the component  $h_y$  of elliptically polarized field  $h$  are shown for different  $h_x$  and for the excitation frequency  $f = 500$  kHz and  $H_0 = 0$ . For each curve, minima are achieved when  $h_x \approx h_y$ , i.e. for circular polarization of the excitation field. Similar dependences were observed for the 1<sup>st</sup> and 3<sup>rd</sup> voltage harmonics.

The phase of the 1<sup>st</sup> harmonic of the ME voltage was found to change abruptly from approximately  $-90^\circ$  to



**Figure 6.** Second harmonic amplitude at  $H_0=0$  as a function of  $h_y$  component of elliptically polarized excitation field  $h$  for different  $h_x$ .

approximately  $+90^\circ$  when the bias dc magnetic field  $H_0$  was reversed. Figure 7 shows such dependences for several magnitudes of rotating excitation field  $h$  and for the frequency of 500 Hz. The phase reversal steepness near  $H_0 \approx 0$  was 970 grad/Oe for  $h = 5$  Oe and monotonously decreased to 200 grad/Oe with increasing  $h$  to 40 Oe. At the same time, the width of the hysteresis decreased from  $\sim 0.075$  Oe to



**Figure 7.** First harmonic of the ME voltage as a function of bias field  $H_0$  for different magnitudes of rotating field  $h$ . The inset is a zoomed-in view of the central area.

$\sim 0.03$  Oe (see inset), indicating a substantial suppression of the ME effect hysteresis with increasing the amplitude of the excitation field [22].

#### 4. Discussion

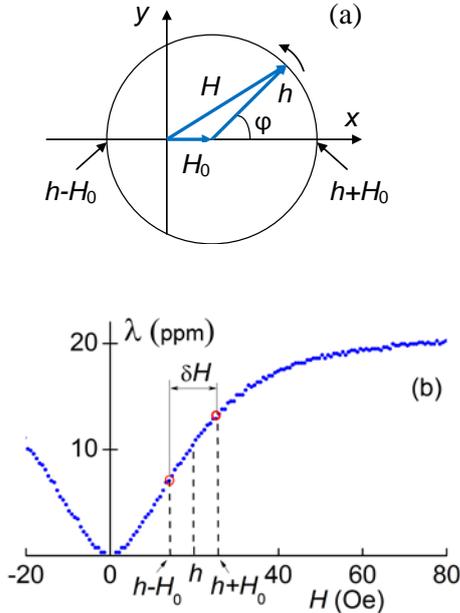
To explain the results, we used the approach proposed in [28], modifying it for the case of rotating excitation field. Let the composite structure be subjected to both a dc bias field  $H_0$  and a rotating field  $h \cos(2\pi ft)$ . First, we consider the case of the FM film being isotropic in the plane and the magnitude of magnetostriction depending only on the magnitude of the resulting field  $H$ , but not on its direction. Figure 8a shows the vector addition diagram for the fields in the "x-y" plane of the sample.

The magnitude of the resultant field acting on the structure at each moment of time is given by the formula

$$H(t) = \sqrt{H_0^2 + h^2 + 2H_0h \cos(2\pi ft)}, \quad (1)$$

where the angle  $\varphi = 2\pi ft$  is counted from the  $x$ -axis. The direction of magnetostrictive deformation of the FM layer coincides with the direction of  $H$  and follows it when the field is rotated. Figure 8b shows the measured deformation of a FeBSiC layer as a function of field  $H$ . Dashed lines in Fig. 8b denote the range of variation of the resultant field  $\delta H$  for  $H_0 = 5$  Oe and  $h = 20$  Oe. When the excitation field rotates, the working point moves along the magnetostriction curve  $\lambda(H)$  within the specified field range.

Let us find the voltage  $u$  generated by the structure for two limiting cases: for small and for large amplitudes of the excitation field  $h$ . To do this, we represent Eq. (1) as a power



**Figure 8.** (a) Field addition diagram. (b) Magnetostriction of a FeBSiC layer as a function of resultant magnetic field  $H$ .

series up to the first order terms and get

$$H(f) \approx H_0 + h \cos(2\pi ft) \text{ for } h \ll H_0 \text{ and}$$

$$H(f) \approx h + H_0 \cos(2\pi ft) \text{ for } h \gg H_0. \quad (2)$$

The amplitude of the voltage generated by the PE layer of the structure due to both magnetostriction and piezoelectric effects is equal to [22]

$$u = Ad_{31}\lambda(H), \quad (3)$$

where  $A$  is a constant that depends on the mechanical parameters and dimensions of the layers of the structure, and  $d_{31}$  is the piezoelectric module of the PE layer.

By expanding  $\lambda(H)$  into a Taylor series up to the third-order term, substituting (2) into (3), and rearranging the terms, we get the following expression

$$u(f) = u_0 + u_1 \cos(2\pi ft) + u_2 \cos(4\pi ft) + u_3 \cos(6\pi ft) + \dots \quad (4)$$

The first term quickly drops to zero due to the finite conductivity of the PE layer, so we will neglect it in our consideration.

In the case of small amplitude of the excitation fields  $h \ll H_0$ , the amplitudes of the first  $u_1$ , second  $u_2$ , and third  $u_3$  voltage harmonics are equal, respectively,

$$u_1 = Ad_{31}\lambda^{(1)}h, \quad u_2 = (1/4)Ad_{31}\lambda^{(2)}h^2 \quad \text{and} \\ u_3 = (1/24)Ad_{31}\lambda^{(3)}h^3. \quad (5)$$

Here  $\lambda^{(1)} = \partial\lambda / \partial H|_{H=H_0}$ ,  $\lambda^{(2)} = \partial^2\lambda / \partial H^2|_{H=H_0}$  and  $\lambda^{(3)} = \partial^3\lambda / \partial H^3|_{H=H_0}$  are the first, second, and third derivatives of magnetostriction with respect to magnetic field, taken at  $H = H_0$ .

In the case of high amplitude of the excitation field  $h \gg H_0$ , the amplitudes of the first, second, and third harmonics are equal, respectively

$$u_1 = Ad_{31}\lambda^{(1)}H_0, \quad u_2 = (1/4)Ad_{31}\lambda^{(2)}H_0^2 \quad \text{and} \\ u_3 = (1/24)Ad_{31}\lambda^{(3)}H_0^3. \quad (6)$$

where the derivatives  $\lambda^{(1)}$ ,  $\lambda^{(2)}$  and  $\lambda^{(3)}$  are taken at  $H=h$ .

Now we consider the case when magnetostriction of the FM layer is still isotropic in the plane, but the excitation field, rotating at frequency  $f$ , has an elliptical polarization (i.e.  $h_x \neq h_y$ ). Then, when the resultant field  $H$  rotates in the structure plane, even for  $H_0 = 0$ , the working point will move along the magnetostriction curve  $\lambda(H)$  and the structure will generate an alternating voltage.

A situation is possible when magnetostriction of the FM film is anisotropic in the plane, for example  $\lambda_x(H) \neq \lambda_y(H)$ . Then, when the structure is excited by a circularly polarized field, even at  $H_0 = 0$ , the working point will move from one magnetostriction curve to another and the structure will also generate an alternating voltage.

The above consideration enabled us to formulate differences and similarities between the characteristics of the ME effect in a disk-shaped ferromagnet-piezoelectric structure when it is excited by a rotating magnetic field, and the characteristics of the ME effect under excitation with a unidirectional ac field.

- The amplitude-frequency response of the ME effect upon excitation with a rotating (i.e. circularly polarized) field is comprised of two contributions corresponding to the excitations with two orthogonal linearly polarized ac fields (see Fig. 2).

- In the region of small amplitudes of the excitation field,  $h \ll H_0$ , the characteristics of the ME effect upon excitation with a rotating field are similar to those upon excitation with an ac field (see Fig. 4, Eq. (5) and formulas (4) in Ref. 27).

- For a high-amplitude rotating field,  $h \gg H_0$ , the amplitudes of voltage harmonics are proportional to the powers of  $H_0$  (see Eqs (6)), rather than the powers of the excitation field  $h$ .

- For a small-amplitude rotating field,  $h \ll H_S$ , the amplitudes of the ME voltage harmonics are power-law functions  $u_n \sim h^n$  of the field (see Fig. 5 and Eq. (5)).

- For  $H_0 \approx 0$ , the harmonics' amplitudes depend on the excitation field ellipticity and are minimal for a circularly polarized field (see Fig. 6). Non-zero values of voltage in minima in Fig. 6 are most likely caused by uniaxial anisotropy of magnetostriction in the textured FeBSiC layer that was used to fabricate the structure.

- When the magnitude of the excitation field is comparable with the magnitudes of both dc bias field and saturation field of the FM layer (i.e.  $h \sim H_0 \sim H_S$ ), the amplitudes of the ME voltage harmonics can be found using the numerical method described in Ref. 27.

If the shape of the FM-PE structure is different from disk, for example, it is rectangular, the field dependences of the generated voltage will be significantly affected by demagnetization effects. A difference between demagnetizing factors of the structure along the "x" and "y" axes will lead to an anisotropy of magnetostriction in the structure plane and, as a result, to a generation of harmonics, even when the excitation field is circularly polarized and  $H_0 = 0$ , which was observed experimentally [14].

It should be noted that the described nonlinear ME effects in ferromagnetic-piezoelectric layered structures can be utilised in magnetic sensors, in which an applied magnetic field is converted into a voltage response [1,2,4,29]. In particular, when the structure is excited by a rotating

magnetic field, the first harmonic can be used to measure permanent magnetic fields (see Fig. 4a), while the second harmonic can be used to measure alternating magnetic fields (see Fig. 5). Such ME sensors of variable fields, as can be seen from Fig. 4b, can work without a bias field (i.e.  $H = 0$ ), which simplifies their design. The use of the nonlinear, rather than linear, ME effect in the sensors would allow to reduce magnetic noise [25] and suppress hysteretic phenomena [30].

## 5. Conclusion

Thus, the ME effect in a disk-shaped composite structure containing an amorphous ferromagnetic FeBSiC layer and a piezoelectric PZT layer, excited by a circular polarized magnetic field  $h$  rotating in the structure plane, was observed and investigated. It is shown that the characteristics of the ME effect can be controlled by applying an additional dc bias magnetic field  $H_0$  in the structure plane. At comparable values of the excitation and dc bias fields (i.e.  $h \sim H_0$ ), the nonlinear dependence of the FM layer magnetostriction  $\lambda(H)$  leads to a generation of voltage harmonics. A theory is proposed that qualitatively describes the ME effect characteristics in the limits of small  $h \ll H_0$  and high  $h \gg H_0$  excitation fields. The described effect can be used to make sensors of permanent and rotating magnetic fields.

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