ISSN 1063-7834, Physics of the Solid State, 2015, Vol. 57, No. 7, pp. 1357–1361. © Pleiades Publishing, Ltd., 2015.
Original Russian Text © A.L. Freidman, A.D. Balaev, A.A. Dubrovskii, E.V. Eremin, K.A. Shaikhutdinov, V.L. Temerov, I.A. Gudim, 2015, published in Fizika Tverdogo Tela, 2015, Vol. 57, No. 7, pp. 1334–1338.

MAGNETISM

Effect of an Electric Field on the Magnetization of a $SmFe_3(BO_3)_4$ Single Crystal

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Received January 12, 2015

Abstract—A change in the magnetization of a $SmFe_3(BO_3)_4$ single crystal in response to an applied alternating electric field has been experimentally observed for the first time. The measurements have demonstrated that the magnetization oscillates not only at a frequency of the applied electric field but also at twice the frequency. The dependences of the magnetoelectric effect on the magnetic and electric fields and temperature have been measured. It has been assumed that the existence of the second harmonic of the magnetoelectric effect is due to the electrostriction.

DOI: 10.1134/S1063783415070112

1. INTRODUCTION

In the recent years, the coexistence of a magnetic order and spontaneous electric polarization in materials called multiferroics is a subject of extensive investigations into the physics of condensed matter. Compounds exhibiting any two or all three types of ordering, namely, a spontaneous magnetic moment, a spontaneous dipole moment, and a spontaneous deformation can be called multiferroics [1].

Among multiferroics, two types can be distinguished: materials in which magnetic and ferroelectric transitions occur independently of each other and materials in which these phase transitions occur simultaneously and correlate with each other. In the latter case, the interaction between the magnetic and ferroelectric subsystems can be very strong [2].

The materials, in which the magnetoelectric effect is observed, demonstrate the dependence of the polarization on the applied magnetic field (so-called ME_H effect) or the change in the magnetization when applying an electric field (so-called ME_E effect) [3].

Among materials exhibiting the magnetoelectric effect, we note the family of $RM_3(BO_3)_4$ borates, where R is a rare-earth ion or Y and M = Al, Fe, Ga, Sc, Cr ions. The crystals of this family have space group $R32$ (see, e.g., [4]), which determines the absence of the inversion center. The sublattice of MO_6 octahedra forms a helicoidal chain along axis c with the exchange interaction between the 3d elements; the rare-earth ions, forming RO_6 prisms, are isolated from each other by BO_3 triangles and, as a result, the R–O–R-type interaction is absent. Both the BO_3 triangles and RO_6 prisms are bound with three MO_6 chains.

In [5], it was shown that the polarization of the $SmFe_3(BO_3)_4$ compound is induced by the antiferroelectric ordering ($T_N \approx 40$ K) of Fe^{3+} ions, which indicates a strong magnetoelectric coupling. The magnetically induced polarization $\Delta P(H)$ (ME_H effect) of this compound was measured in [5], and, in addition, its magnetodielectric effect $\varepsilon(H)$ was studied in [2]; however, up to now, there are no available data on the measurement of the ME_E effect, i.e., the magnetization as a function of electric field $\Delta M(E)$. In this work, the ME_H effect in $SmFe_3(BO_3)_4$ has been measured for the first time as a function of magnetic and electric fields and temperature.

2. SAMPLE PREPARATION AND EXPERIMENTAL TECHNIQUE

The samarium ferroborate $SmFe_3(BO_3)_4$ single crystal was grown from a $Bi_2Mo_3O_{12}$-based solution–melt [6]. We will use the orthogonal system of coordinates (x, y, z), where x and z coincide with the crystallographic directions a and c, respectively, and direction y is perpendicular to an xz plane. The sample was cut as a rectangular plate.

To measure the ME_E effect, we deposited an epoxy-based conducting adhesive on the yz faces of the sample. An alternating-current (ac) electric field E applied to the sample plates causes oscillations of its magnetization with amplitude ΔM and a frequency of the

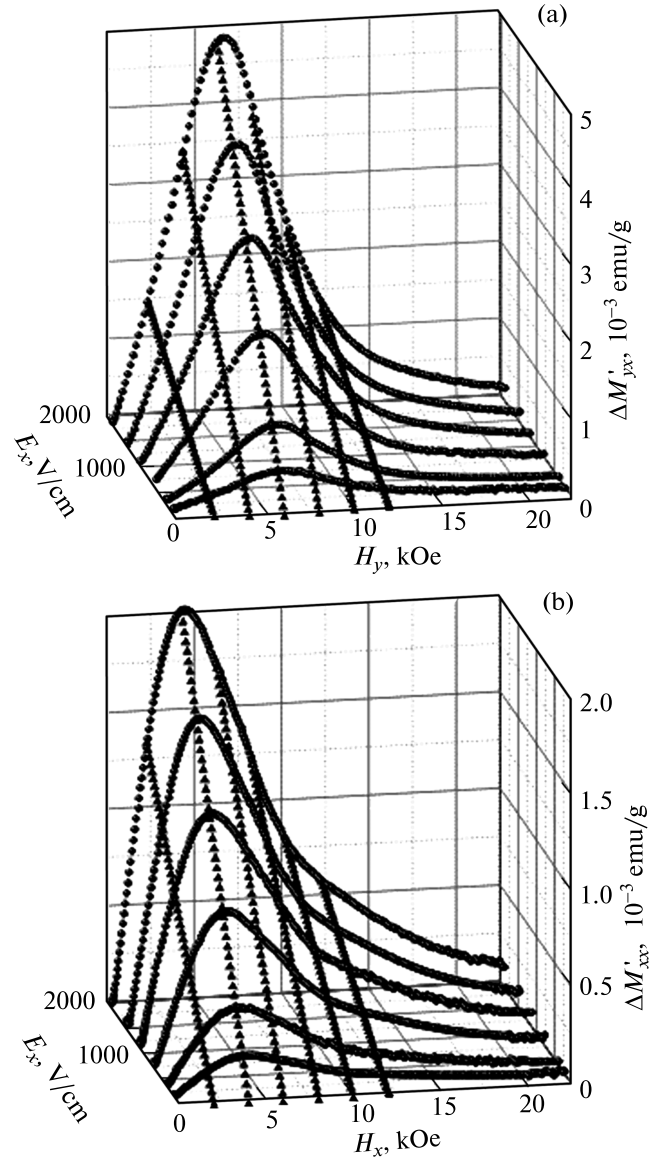

Fig. 1. Dependences (a) $\Delta M'_{yx}$ (E_x, H_y) and (b) $\Delta M'_{xx}$ (E_x, H_{yx}) measured at $T = 4.2$ K. The circles denote the experimental data, and the triangles are the points obtained by the linear approximation.

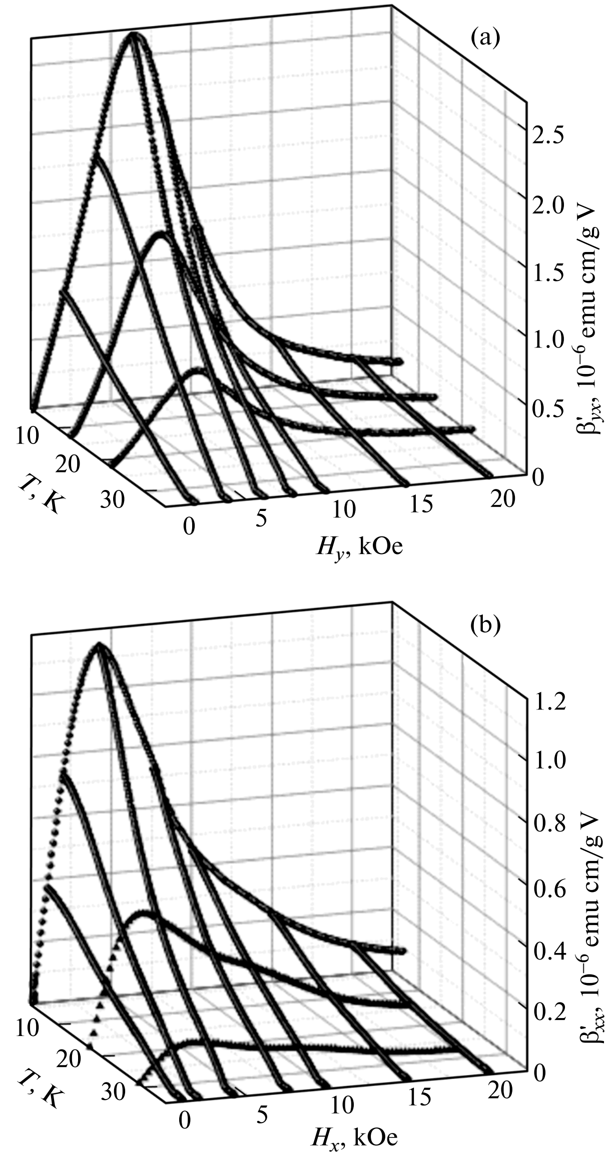

Fig. 2. Dependences of the magnetoelectric susceptibilities of (a) transverse β'_{yx} (H_y, T) and (b) longitudinal β'_{xx} (H_x, T) effects. Circles show the experimental data, and triangles are the points obtained by the spline approximation.

applied voltage (first harmonic), as well as with a frequency that is twice as high as the frequency of field E due to the magnetoelectric (ME_E) effect. The value of ΔM was measured by a Stanford Research Systems Model SR830 DSP Lock-in Amplifier (synchronous detector), which can measure a signal at several harmonics. The technique of measuring the ME_E effect was described in detail in [7, 8].

3. RESULTS

Figure 1 depicts results of the measurement of the first harmonic of the ME_E effect as a function of magnetic field H and the amplitude of ac electric field E. We proposed the denotations $\Delta M'_{ij}$ (or $\Delta M''_{ij}$), where one prime denotes the first harmonic and two primes denote the second harmonic, i is the direction along which the change in the magnetization ΔM was measured (the direction of H also always coincides with i because of the structural features of the device), j is the direction of field E. As is seen from Fig. 1, in order that the ME_E effect to occur, it is necessary that both the magnetic and the electric fields should be nonzero for both the transverse (Fig. 1a) and the longitudinal (Fig. 1b) cases. In this case, function $\Delta M(E, H)$ is lin-

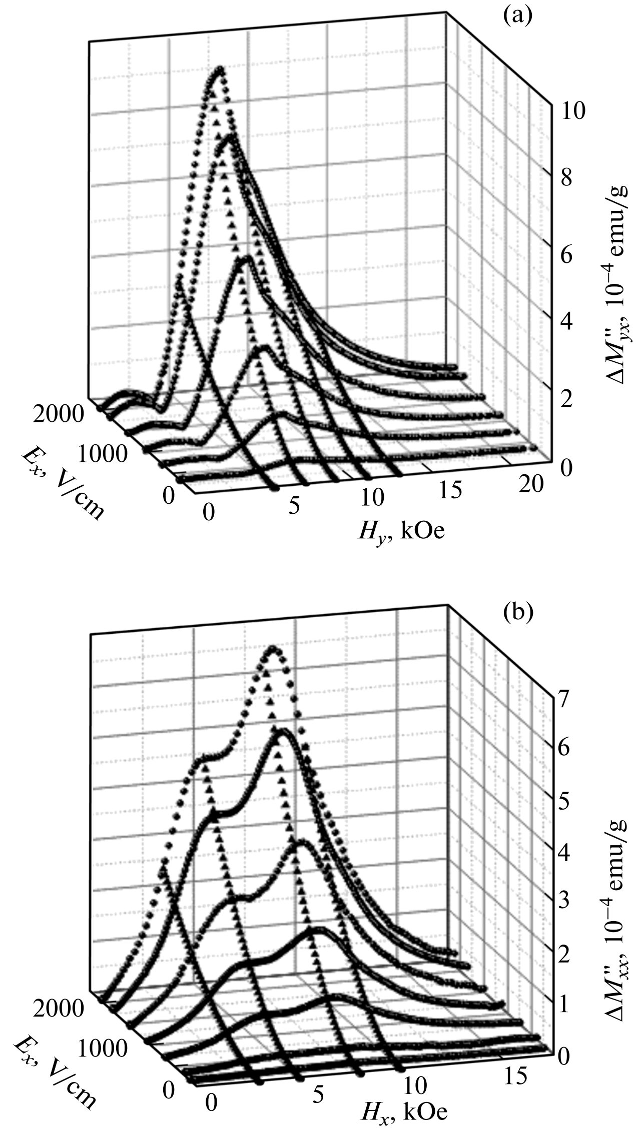

Fig. 3. Field dependences of the second harmonic of (a) the transverse $\Delta M''_{yx}(H_y, E_x)$ and (b) the longitudinal $\Delta M''_{xx}(H_x, E_x)$ magnetoelectric effects, $T = 4.2$ K. Circles show the experimental data, and triangles are the points obtained by the approximation with the second-degree polynomial.

ear with respect to electric field E but nonlinear and nonmonotonic with respect to magnetic field H. The effects are different in the magnitudes and have maxima in magnetic fields 6.4 and 4.2 kOe for the transverse and the longitudinal effects, respectively. The maximum value of $\Delta M'_{yx}$ is almost twice as large as $\Delta M'_{xx}$.

It is interesting that, in the case of ME_H effect, function $\Delta P(H)$ has the highest curvature near 5 kOe [5], while the ME_E effect has a maximum near such magnetic fields. In addition, function $\Delta P(H)$ tends to

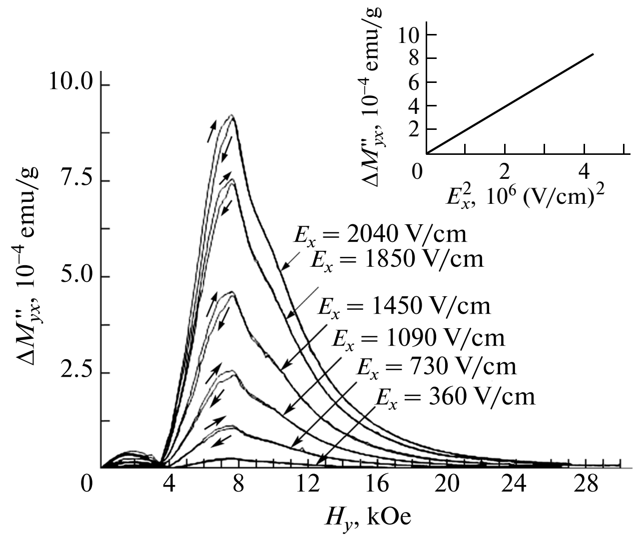

Fig. 4. Dependences $\Delta M''_{yx}(H_y,)$ measured at different amplitudes of the external electric field E_x and $T = 4.2$ K. The inset shows $\Delta M''_{yx}$ as a function of the square of the amplitude of the external electric field E_x at $H_y = 8$ kOe and $T = 4.2$ K.

saturation in fields higher than 10 kOe, while the ME_E effect almost disappears, which corresponds to the formation of an uniform antiferromagnetic ordering [5].

Figure 2 shows the temperature–field dependences of the magnetoelectric susceptibility of the first harmonic of the ME_E effect determined as $\beta'_{ij} = \Delta M'_{ij}/E_j$. In Fig. 2a, the magnetic field is applied along the direction of axis y (transverse effect), and, in Fig. 2b, the magnetic field is directed along axis x (longitudinal effect). As is seen from Figs. 2a and 2b, the maximum of the ME_E effect in dependences $\beta'_{yx}(H_y, T)$ and $\beta'_{xx}(H_x, T)$ decreases and shifts to weaker magnetic fields as temperature increases up to the phase transition at $T = 33$ K.

Moreover, we revealed the changes in the magnetization $\Delta M''$ at the frequency that is twice as high as the frequency of exciting electric field E. Figure 3 shows the dependences of the second harmonic of the ME_E effect as a function of H and E. Whereas the first harmonic of the ME_E effect $\Delta M'$ is linear with respect to the electric field amplitude E, the signal of the second harmonic $\Delta M''$ has a quadratic dependence on E. The dependence of $\Delta M''$ on the magnetic field also becomes more complex.

As magnetic field increases, the second harmonic amplitude of the transverse effect $\Delta M''_{yx}$ (Fig. 3a) increases to the first maximum that is observed in field $H_y \sim 2$ kOe; then, the effect decreases and completely disappears in a field ~ 3.5 kOe. With further increasing field, the value of $\Delta M''_{yx}$ increases once again and goes

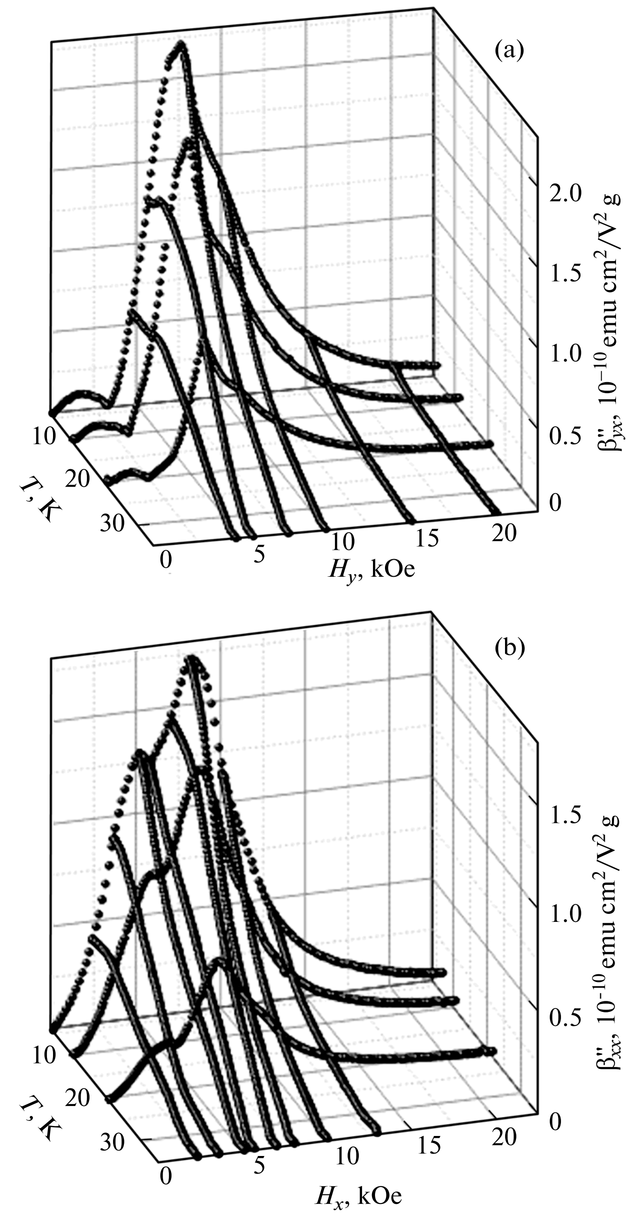

Fig. 5. Dependences (a) β''_{yx} (H_y, T) and (b) β''_{xx} (H_x, T).

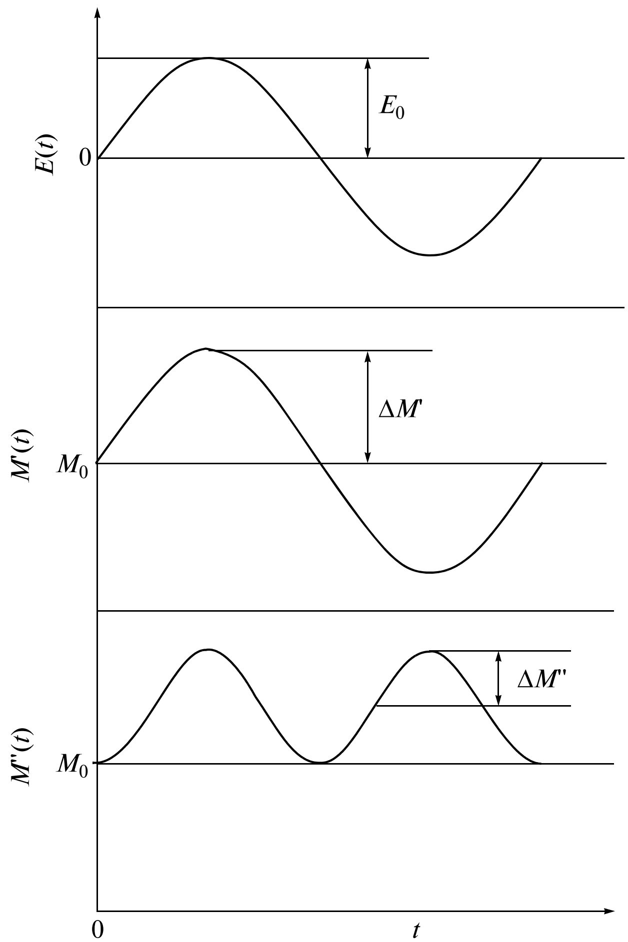

Fig. 6. Schematic representation of the time dependences $E(t)$, $\Delta M'(t)$, and $\Delta M''(t)$.

to the second maximum in field $H_y \sim 7.5$ kOe; then the effect decreases monotonically. If thereafter we begin to decrease external field H_y, we observe a small hysteresis shown in Fig. 4. It should be noted that quantity ΔM has the meaning of the amplitude of variations in the magnetic moment in all the presented plots; however, the measuring method makes it also possible to observe the phase of the detected signal, which is turned to be dependent on the magnetic field. For example, for all measuring configurations and harmonics, the phase is switched by π as the direction of field H is changed to the opposite direction; i.e., when the field passes through zero; in the other words, the effect is odd with respect to H. In addition, the second harmonic phase of the transverse effect $\Delta M''_{yx}(H_y)$ is also switched in field $H_y \sim +3.5$ kOe (Fig. 4). The insert to Fig. 4 shows a strong quadratic dependence of the second harmonic on electric field E, i.e., $\Delta M'' = \beta''(H)E^2$.

The second harmonic of the longitudinal effect $\Delta M''_{xx}$ (Fig. 3b) also demonstrates two maxima in fields $H = 5.2$ and 8.1 kOe at $T = 4.2$ K; however, the phase is only switched in point $H = 0$. The longitudinal effect also demonstrates a weak hysteresis in field H.

Temperature–field dependence of the magnetoelectric susceptibility $\beta''(H, T)$ is shown in Fig. 5 for the transverse (Fig. 5a) and the longitudinal (Fig. 5b) effects. As seen from Fig. 5, the maxima of the second harmonic of the ME_E effect, as is the case with the first harmonic, shift to weaker magnetic fields as temperature increases. In addition, the phase switching point of the transverse effect also shifts to weaker fields with increasing T.

4. DISCUSSION

An ac electric field applied to the crystal excites in it simultaneously the inverse piezoelectric effect and the electrostriction. In this case, when the sample is in a constant magnetic field, the deformation leads to the formation of the magnetoelastic effect, which causes a change in the magnetization. Figure 6 shows schematically the time dependence of the applied ac electric field E and magnetoelectric response of the first $\Delta M'$ and the second $\Delta M''$ harmonics. Since the crystal is in a constant magnetic field H, the sample magnetization oscillates with respect to the constant value $M_0(H)$ with amplitude ΔM. In this case, the first harmonic frequency coincides with the frequency of exciting field E, and the second harmonic frequency is twice as high as the frequency of field E. This implies that the effect leading to oscillations of $\Delta M'$ is odd in E and that inducing oscillations $\Delta M''$ is even in E. According to our measurements, quantity $\Delta M'$ is linear with respect to electric field E and $\Delta M''$ has a quadratic dependence on E, and this fact indicates that the ME_E effect is induced by the piezoelectric and the electrostriction effects.

5. CONCLUSIONS

The ME_E effect in the $SmFe_3(BO_3)_4$ crystal as a function of magnetic and electric fields and temperature was measured for the first time. We separated the linear and quadratic contributions to the magnetoelectric effect under study. An ac electric field applied to the crystal simultaneously induces in it the piezoelectric effect and the electrostriction. It was assumed that the piezoelectric effect is responsible for the existence of the first harmonic of the ME_E effect and the electrostriction effect is responsible for the existence of the second harmonic. In order to better explain the nonmonotonic dependences of the ME_E effect, additional studies are necessary, and they will be performed in future works.

ACKNOWLEDGMENTS

This study was supported by the Russian Foundation for Basic Research (project no. 14-02-00307_a).

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Translated by Yu. Ryzhkov