

**Current-induced nonlinear magnetoelectric effects in strontium hexaferrite**I. V. Zavislyak,<sup>1</sup> M. A. Popov,<sup>1</sup> and G. Srinivasan<sup>2</sup><sup>1</sup>*Department of Radiophysics, Electronics and Computer Systems, Taras Shevchenko National University of Kyiv, Kyiv 01601, Ukraine*<sup>2</sup>*Physics Department, Oakland University, Rochester, Michigan 48309, USA*

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We report on the observation of nonlinear magnetoelectric effects at room temperature due to a dc current in the ferrimagnetic  $M$ -type strontium hexaferrite platelets. Utilizing microwave measurement techniques and data on the shift in magnetic mode frequencies, it was found that a dc current along the hexagonal  $c$  axis resulted in a significant decrease in the saturation magnetization and an increase in the uniaxial magnetocrystalline anisotropy field. These changes in the magnetic order parameters were directly proportional to the square of applied electric field and were found to be much higher than variations due to Joule heating. A phenomenological theory that takes into account the current-induced magnetobielectric (MBE) effects is proposed. Expressions for coupling coefficients for MBE effects have been obtained and have been calculated from the variations in magnetic order parameters. The electric field  $E$  (or current) tuning of the magnetic modes in  $SrM$  reported here is orders of magnitude stronger than strain mediated  $E$  tuning of magnetic resonance in hexaferrite-ferroelectric composites. The nonlinear magnetoelectric effects in hexaferrite, therefore, open up an avenue for the realization of  $E$ -tunable broadband microwave and millimeter wave ferrite signal processing devices such as resonators and filters.

DOI: [10.1103/PhysRevB.94.224419](https://doi.org/10.1103/PhysRevB.94.224419)**I. INTRODUCTION**

This report is on electric-field control of magnetism in  $M$ -type hexagonal ferrite through nonlinear magnetoelectric interactions. There have been several reports in recent years on current/electric field control of ferroic order parameters in magnetic thin films and in composite structures. Efforts so far could be classified into two broad categories: (i) electric currents or spin currents in ferromagnetic films or composites [1–7] and (ii) influence of applied electric fields in single phase or composite multiferroics [8–16]. A rich variety of phenomena involving electron transport in ferromagnetic films or heterostructures were reported and were associated with the transfer of charge, spin-orbital moment, or spin magnetic moment [17]. Examples include giant magnetoresistance, tunneling magnetoresistance, spin-Hall effect and spin torque transfer that were successfully utilized for control of resistivity, magnetization reorientation, and microwave generation [1–7]. Several useful applications including magnetic sensors, memory devices, and spin torque nano-oscillators have also evolved from such studies [17].

Electric-field control of magnetism is also well known in multiferroic materials with coexisting long-range ordering of magnetic moments and electric dipoles. The magnetoelectric (ME) effect in such materials is defined as an induced magnetization (polarization) in an external electric (magnetic) field [8–12]. In a vast majority of single-phase multiferroics, however, the ME effect is rather weak even at low temperatures [8,9,18–22]. A strong ME coupling, could, however, be realized in a composite consisting of magnetostrictive and piezoelectric phases in which the ME effect is the result of a “product-property”, i.e., the mechanical deformation due to magnetostriction resulting in dielectric polarization due to piezoelectric effects [10–14]. Studies so far have focused on thick films and thin-film heterostructures and bonded bilayers of ferrites, manganites, and metals/alloys for the ferromagnetic phase and several ferroelectrics including lead zirconate titanate and barium titanate [12]. A strong strain mediated

ME coupling was reported in several of these systems. Device applications for the ME effects in the composites include pT magnetic sensors, gyrators, and voltage-tunable microwave ferrite signal processing devices [12–16].

Here we discuss the nature of electric-field-induced nonlinear ME interactions in the  $M$ -type strontium hexagonal ferrite  $SrFe_{12}O_{19}$  ( $SrM$ ) which is an  $n$ -type semiconductor. Linear ME effects are prohibited in  $M$ -type barium and strontium hexaferrites with collinear magnetic arrangement since the crystal symmetry contains spatial inversion element [18]. Such linear ME effects, however, are allowed in heavily doped  $M$ -type and  $Z$ -type hexaferrites in which the dopants replace  $Fe^{3+}$  and weaken the strength of exchange interactions, resulting in a spiral spin structure. Examples include  $Z$ -type  $Sr_3Co_2Fe_{24}O_{41}$  and  $M$ -type  $SrCo_2Ti_2Fe_8O_{19}$  and the application of an electric field to these ferrites induces a change in the magnetization [23–29]. The unit cell of strontium hexaferrite, the ferrimagnetic oxide considered in this study, consists of spinel and hexagonally packed blocks in which iron atoms are distributed among five different types of crystallographic sites (labeled 4f1, 4f2, 2a, 2b, 12k). Ferrimagnetic ordering of magnetic moments (with Fe at the 4f1 and 4f2 sites having their spins aligned antiparallel to the rest of the Fe atoms) gives rise to a net ferromagnetic moment in the material. Although linear ME effect of the type  $EH$  (in the free energy) and nonlinear ME effects of the type  $EHH$  are forbidden in  $SrM$ , coupling of the type  $HEE$  (also called magnetobielectric interaction [18]) and higher-order ME effects of the type  $HHEE$  are allowed [21,30].

This paper is on nonlinear ME interactions in  $SrM$ . An electric field  $E$  applied along the  $c$  axis was found to result in a strong current-induced nonlinear ME interaction that manifests as variations in the magnetic order parameters (the ME phenomenon in this study could be described either as “ $E$  induced” or as “ $I$  induced” since the current is rather large in our semiconducting  $SrM$  platelets). We utilized microwave measurement techniques to estimate the changes in the magnetocrystalline anisotropy field and saturation

magnetization. The  $I$ -induced shift in the magnetic resonance frequency  $\Delta f_r$  in the multidomain state depends only on the change in uniaxial anisotropy field. Data on  $\Delta f_r$  were used to estimate the change in the uniaxial anisotropy field  $H_a$  with  $I$ . Similar measurements were then carried out in the single-domain (or saturated) state for estimating the change in the saturation magnetization  $M_0$ . Our data reveal a decrease in  $M_0$  and an increase in  $H_a$  which scale linearly with the electric power density (or  $E^2$ ). Although the influence of magnetobielectric (MBE) effects on magnetic parameters is somewhat similar to thermal heating, we show that  $I$ -induced variations are much higher than expected changes due to Joule heating.

We have developed a phenomenological model for the nonlinear ME effects from the free-energy considerations. Expressions for variations in the magnetization and anisotropy field have been obtained in terms of the magnetobielectric coupling coefficients. We estimated these coupling coefficients for SrM from the data on changes in the magnetic parameters versus electric power. A theory for the mechanisms responsible for the current control of magnetism in SrM is yet to be developed. According to the density function theory, SrM is a semiconductor with  $n$ -type conductivity with the bottom of the conduction band consisting of electronic states of Fe ions [31]. Iron ions in the 12k sites contribute substantially to the net ferromagnetic moment and Fe in 2b sites to the uniaxial anisotropy constant  $K_u$ . Under a dc current, electrons from these sites are involved in the hopping-type conduction and the hopping occurs via intermediate oxygen atoms [32]. It is essential to model the influence of the conduction current along the  $c$  axis on the magnetic exchange and spin-orbit coupling interactions and the resulting changes in the magnetic parameters

The results presented here are also of technological importance for tunable ferrite signal processing devices. Ferrites are the materials of choice for low-loss high-frequency communication devices. Tuning of such devices has heretofore required the application of a large but variable external magnetic field that provides relatively slow tuning, utilizes multiwatt levels of power which cannot be readily miniaturized or integrated with high-frequency semiconductor (e.g., GaN or

GaAs) devices. However, voltage tuning of ferrite-ferroelectric devices through mechanical strain coupling was demonstrated in recent years, but the tuning was rather small, on the order of 0.01% for hexaferrite-PZT [15,16]. The tuning of resonance modes reported here, however, is one to two orders of magnitude higher than strain mediated voltage tuning. The tuning under a multidomain state, in particular, will facilitate ferrite devices such as resonators and filters that do not require a bias magnetic field. As discussed later in Sec. III, one could potentially achieve GHz tunability by decreasing the ferrite thickness and controlling the sample conductivity. It is therefore possible to realize miniature, broadband current-tunable ferrite devices enabling agile communication systems that can essentially provide rapid frequency hopping between rf bands.

In the sections to follow, we provide details on results of our studies on nonlinear ME interactions in SrM and discuss the results in terms of a phenomenological model.

## II. EXPERIMENT

### A. Electrical characterization

The samples used in this study were single-crystal strontium hexaferrite of the stoichiometric composition  $\text{SrFe}_{12}\text{O}_{19}$  grown by floating zone techniques. Samples were cut into thin platelets with the  $c$  axis perpendicular to the plane. The specimens had lateral dimensions  $a \times b = 2.0 \times 1.5 \text{ mm}^2$  and thickness  $d = 160 \mu\text{m}$ . Electrodes (1.5- $\mu\text{m}$ -thick Pt with 10-nm Ti underlayer) were deposited on the top and bottom surfaces by rf sputtering. The bottom surface of the sample was completely covered with Pt, whereas a 0.75-mm-wide conducting stripe was formed in the middle of the top surface of the sample. The measurement cell (shown in Fig. 1) consisted of a copper plate with the sample bonded to it with a thin layer of an alloy. A small hole was drilled in the Cu plate next to the sample and was used to connect a thin wire to the top electrode. Thus, under an applied voltage a current was established along the  $c$  axis, across the thickness of the ferrite. The bottom Cu plate also served as a heat sink.

The SrM samples were first characterized in terms of electrical properties by measuring the current  $I$  as a function of

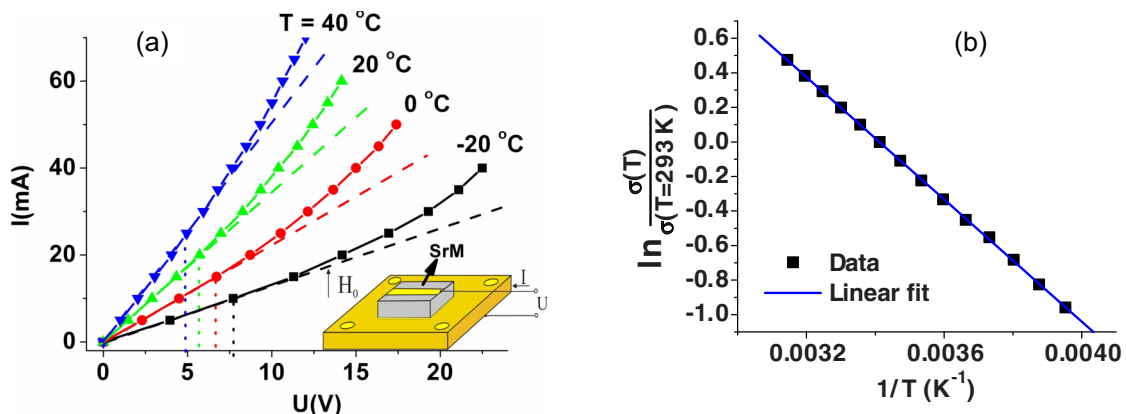


FIG. 1. (a) Current  $I$  vs voltage  $U$  data as a function of temperature  $T$  for a  $c$ -plane platelet of single crystal strontium hexaferrite. The current is directed along the  $c$  axis. Vertical dotted lines show the region of linear  $I$  vs  $U$  behavior. Inset shows the measurement cell with the ferrite platelet with electrodes mounted on a Cu flange. (b) Normalized small-signal conductivity  $\sigma$  vs inverse temperature data for the SrM platelet. The conductivity values were calculated from the linear regime of the data in (a).

applied voltage  $U$  and temperature  $T$ . Such data are essential for information on the nature of the electrical conduction mechanism. The hexaferrite together with a copper plate were placed inside a temperature controlled chamber and  $I$  vs  $U$  data as in Fig. 1(a) were obtained for a series of temperatures. These data reveal a linear behavior for  $U$  in the range 0–7 V and then becomes nonlinear at higher voltages as expected for semiconductors. The conductivities  $\sigma$  were then calculated as a function of temperature from the linear regime of the data in Fig. 1(a) and the results are shown in Fig. 1(b). For  $U > 7V$ , however,  $\sigma$  is dependent on  $U$ . The data in Fig. 1(b) were fitted to the relation  $\sigma(T) = \sigma_0 e^{-q/kT}$  for hopping-type conduction [33]. We estimated an activation energy of  $q = 0.153 \pm 0.001$  eV that is in agreement with reported results for barium hexaferrite, a closely related compound [32]. As described later in Sec. II C, the data in Fig. 1(b) were also used for determining the sample temperature under a current.

### B. Magnetic resonance modes in multidomain and single-domain states

We first characterized the high-frequency modes in the absence of a dc excitation current in order to establish the baseline resonance behavior. For these measurements, the Cu plate with the sample was placed at the open end of a waveguide in order to form a short circuit [see Fig. 2(a)] and profiles of scattering matrix parameter  $S_{11}$  (reflection coefficient) vs frequency  $f$  were recorded and analyzed. The rf power was kept low, approximately 1 mW, to avoid any

sample heating. The waveguide section with the sample was placed between the pole pieces of an electromagnet so that the bias field  $H_0$  was directed perpendicular to the sample plane and parallel to the hexaferrite  $c$  axis.

The measurements were done using two scalar network analyzers, 37–53 GHz and 53–78 GHz frequency bands, and results on mode frequency  $f_r$  as a function of  $H_0$  were then combined to a single plot shown in Fig. 2(b). The data are for the sample in the multidomain ( $H_0$  below magnetic saturation) and single-domain (saturated) states. Several modes were observed in both states. The data in Fig. 2 show modes in the multidomain state that ultimately evolve into (1,1) and (1,3) modes in the single-domain state. Splitting of these (1,1) and (1,3) modes occurs in both multidomain and single-domain states due to the shielding effect of the Pt electrodes. Modes that originate from the sample volume enclosed between the top and bottom electrodes have a higher frequency than the modes from the rest of the sample and are designated as (1,1)' and (1,3)'. Models for  $f_r$  vs  $H_0$  in the multidomain state are rather complicated and sensitive to the actual configuration of the domain structure [34].

The mode with the lowest frequency in the multidomain state is the domain resonance with the frequency  $f_r$  given by [34]

$$f_r(H_0 = 0) = \gamma(H_a - 4\pi M N_{zz}^{\text{eff}}), \quad (1)$$

where the gyromagnetic ratio  $\gamma = 2.66$  MHz/Oe [35],  $H_a = 2K_u/M_0$ , is the uniaxial anisotropy field and  $K_u$  is an anisotropy energy constant, and  $N_{zz}^{\text{eff}}$  is the effective demagnetization coefficient [36]. Under  $H_0 = 0$  the sample with uniaxial anisotropy along the  $c$  axis splits into oppositely magnetized stripe domains of equal volume so that  $4\pi M \sim 0$ . Under these assumptions, the demagnetization term in Eq. (1) is zero and may be neglected. We could, therefore, use data on  $f_r$  for  $H_0 = 0$  to calculate the value of  $H_a$  and its variation with current  $I$ . In the absence of an excitation current, we estimated  $H_a \approx 18.9$  kOe from the data in Fig. 2(b), and it is in good agreement with reported values [37].

In the single-domain (saturated) state, magnetostatic forward volume waves are excited in the sample. The dispersion equation for the modes in a ferrite with one side metallized is given by [38]

$$|k|d = \frac{1}{\sqrt{-\mu}} \arctg\left(\frac{1}{\sqrt{-\mu}}\right), \quad \mu = \frac{f^2 - f_H(f_H + f_M)}{f^2 - f_H^2}. \quad (2)$$

Here  $k$  is a transverse wave number,  $\mu$  is the diagonal element of the rf permeability tensor,  $f_H = \gamma(H_i + H_a)$ ,  $f_M = \gamma 4\pi M_0$ , and  $H_i = H_0 - N_{zz} 4\pi M_0$  (where  $4\pi M_0$  is a saturation magnetization and  $N_{zz}$  is the demagnetizing factor along the  $c$  axis). For our sample we estimated  $N_{zz} = 0.818$  [39]. The mode wave number for a rectangular straight-edge resonator is given by  $k^{(nm)} = \pi \sqrt{(n/a)^2 + (m/b)^2}$ ,  $n, m = 1, 2, 3, \dots$  and the corresponding mode is then designated by the indices  $(n, m)$  [38]. The mode frequencies in the saturated region of Fig. 2(b) with linear  $f_r$  vs  $H_0$  dependence correspond to the two lowest frequency forward volume modes which were identified as (1,1) and (1,3) modes. Estimated frequencies for (1,1) and (1,3) modes are shown for comparison and the

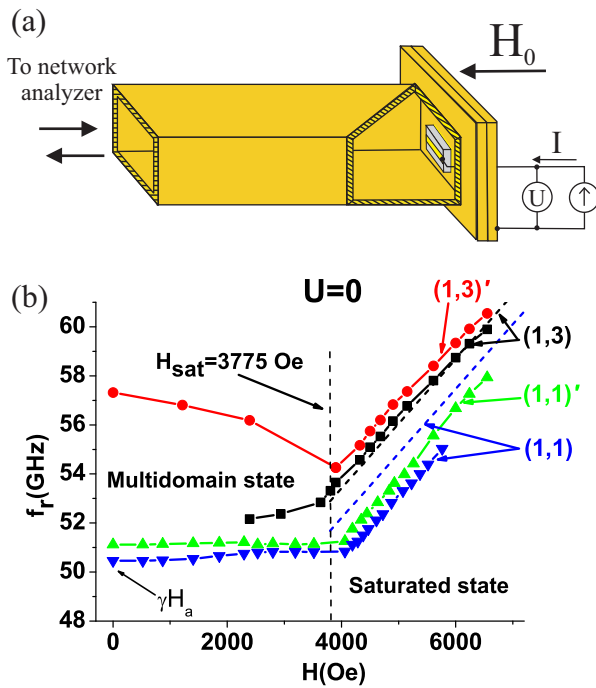


FIG. 2. (a) Diagram showing the experimental setup for measurements on magnetic resonance modes in SrM. (b) Resonance frequency  $f_r$  vs bias magnetic field  $H_0$  dependence for magnetostatic modes of SrM in multidomain and saturated states. Symbols represent data points, solid lines are guides to the eye, and dashed lines are theoretical estimates.

minor discrepancy between theory and experimental values for the (1,1) mode could be attributed to the retardation effect associated with metallized ferrites [38].

Data in Fig. 2 also indicate splitting of the (1,1) and (1,3) modes into doublets labeled (1,1) – (1,1)' and (1,3) – (1,3)', respectively, with the frequency gap between the doublets much smaller than the gap between the two primary modes. We attribute the presence of the doublets to nonuniform metallization of the sample leading to differences in the boundary conditions. It is well known that the top and bottom metallization strongly influence the frequencies of forward volume magnetostatic wave modes [40]. Moreover,

$$\frac{\Delta f^{(nm)}}{\gamma} = \left( \frac{f^2 + f_H^2}{2f_H f} \Delta H_0 + \frac{f^2 + f_H^2}{2f_H f} \Delta H_a + \frac{f_H(f^2 - f_H^2) - f_M N_{zz}(f^2 + f_H^2)}{f_M f_H f} \Delta(4\pi M_0) \right) \Bigg|_{f=f^{(nm)}},$$

where the right-hand side should be evaluated for a given mode frequency. Since for small ( $k^{(nm)}d$ ) values  $f^{(nm)} \approx f_H$  the above expression may be reduced to

$$\frac{\Delta f^{(nm)}}{\gamma} \approx \Delta H_0 + \Delta H_a - N_{zz} \Delta(4\pi M_0). \quad (3)$$

It is clear from Eq. (3) that (i) changes in  $f_r$  are directly proportional to the sum of changes in  $M_0$  and  $H_a$  and, therefore, data on current-induced frequency shift may be used to estimate variations in these magnetic parameters, and (ii) the frequency shift does not depend on frequency of a specific mode. Thus identical shift in frequency is expected for all lowest-order magnetostatic modes. For a specific mode and for constant bias field  $H_0$ , the frequency shift is due to changes in  $H_a$  and  $M_0$ .

The bias field required for the single-domain state  $H_0 = H_{\text{sat}}$  was determined by careful measurement of the mode frequency in the transition region between multidomain and saturated states and was found to be  $H_{\text{sat}}^{\text{exp}} = 3775$  Oe. One expects saturation to occur for  $H_i = 0$  or  $H_{\text{sat}} = 4\pi M_0 N_{zz}$ . Substituting for  $4\pi M_0 = 4652$  G [41] and  $N_{zz} = 0.818$ , we get  $H_{\text{sat}}^{\text{theory}} = 3805$  Oe, in agreement with the experiment.

### C. Effects of Joule heating on magnetic parameters

In order to rule out the influence of Joule heating from the ME effects we wish to study, we measured the electrical resistance of the sample as a function of temperature in a separate experiment. Although the copper flange on which the SrM platelet is mounted acts as a heat sink for the heat generated under a current pulse, sample heating inevitably occurs and will lead to variation in the magnetic parameters and a shift in  $f_r$ . So it was necessary to investigate the current-induced change in the sample temperature. We performed a detailed and systematic study to assess the extent of sample heating and its contribution to changes in  $M_0$  and  $H_a$ . In order to avoid complications associated with temperature measurements using an external probe, we utilized electrical conductivity  $\sigma$  vs  $T$  data in Fig. 1(b) to determine the sample temperature from measured  $\sigma$  values.

double-sided metallization leads to higher mode frequency. In the present case the sample has double-sided metallization only in the central part and has a single-sided metallization elsewhere. Hence, magnetostatic modes that are primarily localized in the center of the sample will differ in frequency from the modes localized away from the center.

Next we consider the variation in mode frequencies due to any change in the magnetic parameters in the single-domain state. For a specific value of wave number  $k = k^{(nm)}$  we have  $\mu = \mu^{(nm)} = \text{const}$  from Eq. (2). Then after differentiating the expression for  $\mu$  in Eq. (2) and equating  $d\mu^{(nm)} = 0$ , we get the following expression for the frequency shift:

Measurements on thermally induced effects were done using the following procedure. A dc current of preset magnitude was supplied to the sample for a prolonged period of  $\sim 10$  min. Then at certain time intervals, varying from 5 s to 1 min, the current was switched off momentarily and sample resistance was measured using the same electrodes used for the current application. After 10 min the current was turned off and the sample resistance was measured at periodic intervals as the sample was cooled. The sample temperature was then determined from  $\sigma$  vs  $T$  data in Fig. 1(b). Figure 3 shows the sample temperature thus obtained as a function of time for currents up to a maximum of 70 mA. The sample temperature initially increases rapidly with time over the interval  $t = 0$  to 100 s. The rate of increase in  $T$  with time over this interval ranges from  $0.02^\circ\text{C/s}$  for  $I = 50$  mA to  $0.04^\circ\text{C/s}$  for  $I = 70$  mA. Then the rate of increase in  $T$  with  $t$  slows down and finally it levels off. Since we are primarily interested in the sample heating characteristics for the low- $t$  region in Fig. 3, we used excitation currents that are pulses of 1 s duration in our experiments as described in the following section. Figure 3 also shows the  $\Delta T$  vs  $t$  profiles for the time

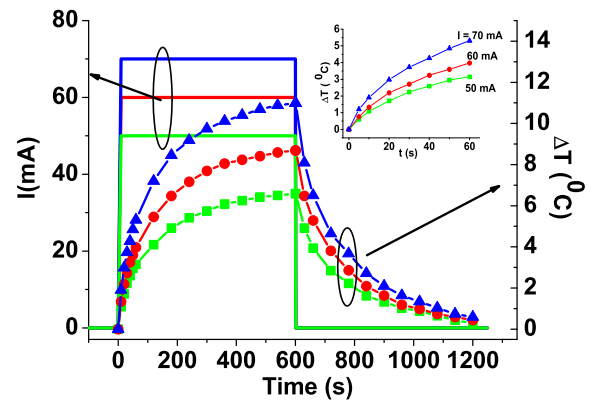


FIG. 3. Change in temperature  $\Delta T$  of SrM as a function of time  $t$  under a current pulse of duration 10 min and different amplitudes. The inset shows increase in sample temperature during the initial 1 min. Also shown is the  $\Delta T$  vs  $t$  data when the current is switched off and the sample cooled down to room temperature.

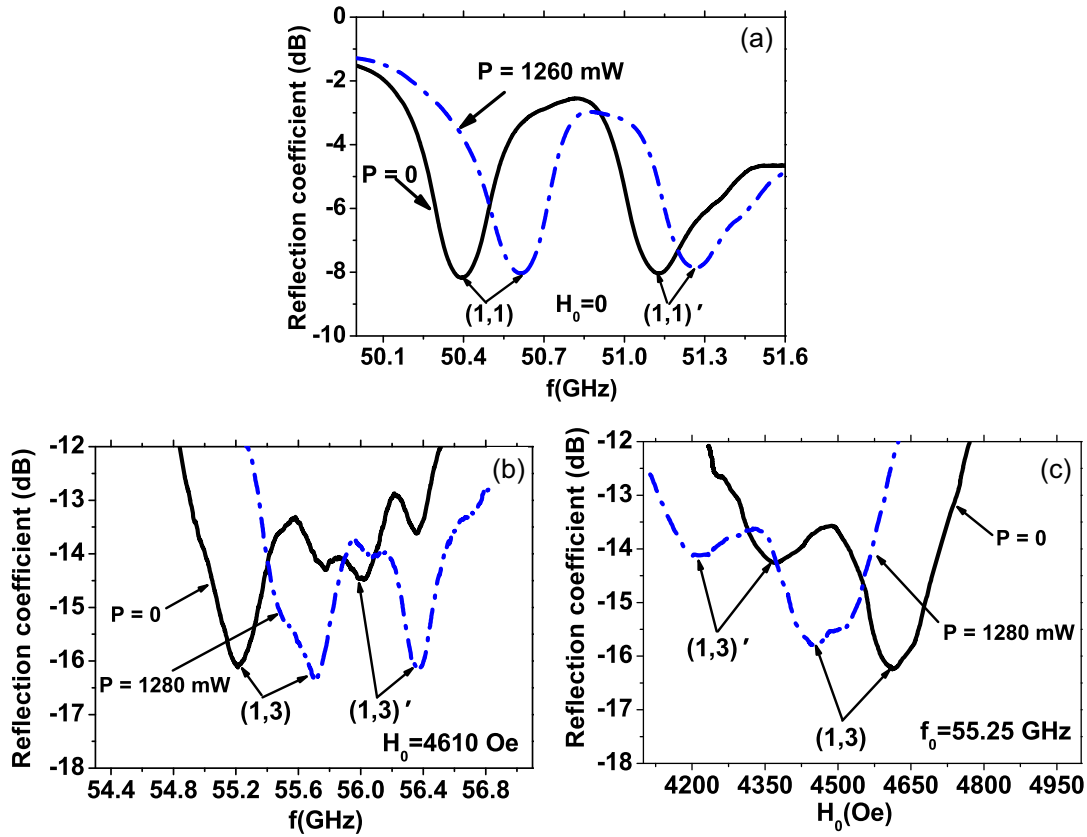


FIG. 4. Reflection coefficient vs  $f$  data as a function of electric power applied to the sample (a) in the multidomain state ( $H_0 = 0$ ) and (b) in the single-domain state. (c) Reflection coefficient vs  $H_0$  for the single-domain state. The current pulses were of 1 s duration,  $\sim <10$  ms rise/fall time and 10–70 mA amplitude.

interval  $t = 0$ –60 s. From the data in Fig. 3 for  $t = 0$ –5 s, we estimated  $\Delta T/\Delta t \approx 0.25^\circ\text{C}/\text{s}$  for  $I = 70$  mA. Therefore, for current pulses of 1 s duration, the temperature rise may not exceed  $0.25^\circ\text{C}$  for the currents used in our experiments.

Following the acquisition of the data in Fig. 3, we estimated the anticipated change in  $f_r$  in the multidomain and single-domain resonances due to the expected increase in sample temperature. Additional experiments, conducted in environmental chamber in the same temperature range, yielded an estimated upshift of  $\Delta f_r = 3.7$  MHz for multidomain resonance for  $0.25^\circ\text{C}$  increase in  $T$  that can only be attributed to an increase in  $H_a$  with increasing temperature [Eq. (1)]. On the other hand, fractional decrease in the saturation magnetization with temperature  $\frac{1}{\Delta T} \frac{\Delta(4\pi M_0)}{4\pi M}$  for SrM was reported to be  $-0.002/^\circ\text{C}$  [42,43]. Hence, for  $4\pi M_0 = 4652$  G [41], we obtain  $\Delta(4\pi M_0)/\Delta T \approx -9$  G/ $^\circ\text{C}$  corresponding to  $\Delta f_r = -\gamma\Delta(4\pi M_0) = 6$  MHz for  $0.25^\circ\text{C}$  increase in  $T$ . Summarizing, the potential contribution of Joule heating to frequency shift during a 1-s, 70-mA current pulse is estimated to be  $\Delta f_r^{\text{thermal}} \approx 4$  MHz for the multidomain state measurements and  $\Delta f_r^{\text{thermal}} \approx 10$  MHz for the saturated-state [Eq. (3)]. As discussed in the section to follow, the frequency shifts in  $f_r$  due to nonlinear ME interactions are at least an order of magnitude higher than the variations originating from Joule heating. One could, therefore, rule out any significant contribution from the Joule heating to variation in the magnetic parameters and a shift in the magnetic mode frequencies.

#### D. Microwave measurements on current induced ME effects

Next we discuss our investigations on the current-induced nonlinear ME effects. During the measurements, short current pulses were applied and the resonance frequency was measured as a function of current (input power). The measurements involved applying rectangular current pulses of short duration. The pulse width was selected short enough to limit the influence of Joule heating as discussed in Sec. II C. Once the current stabilized we recorded the resonance profiles.

One may utilize profiles of either  $S_{11}$  vs  $f$  at constant bias magnetic field or  $S_{11}$  vs  $H_0$  at constant  $f$  for such measurements. For multidomain resonance we are primarily interested in the  $S_{11}$  vs  $f$  profiles due to the fact that it is possible to readily estimate  $H_a$  and its current variation directly from the resonance frequency and using Eq. (1). For single-domain resonance both types of profiles could be used to determine the resonance field and its shift under a current pulse. Resonance profiles under zero current and upon the application of a current pulse are shown in Fig. 4. Profiles in Fig. 4(a) are for the sample in the multidomain state whereas Fig. 4(b) shows similar results for forward volume waves in the sample magnetized to saturation. Figure 4(c) shows a representative example for  $S_{11}$  vs  $H_0$  profiles measured at constant  $f$ . Such profiles are useful since they are less susceptible to broadening due to unwanted electrodynamic ripples that appear in transmission characteristic when using frequency sweeps. However, in the multidomain state  $f_r$  has a

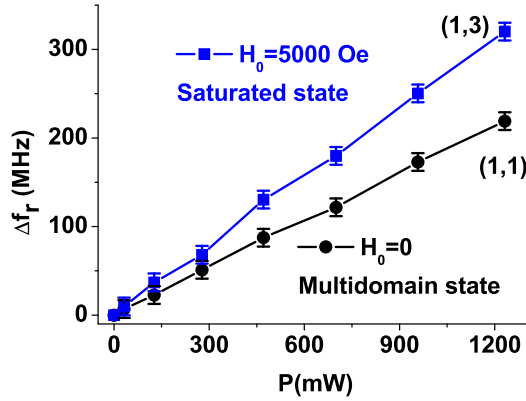


FIG. 5. Frequency shift for magnetic resonance modes  $\Delta f_r$  as a function of applied power  $P$  in multidomain and single-domain states of SrM.

weak dependence on  $H_0$  as seen in the data of Fig. 2(b), and  $S_{11}$  vs  $H_0$  are not appropriate for this specific case. Therefore  $S_{11}$  vs  $f$  profiles are preferred in this study. Finally, such profiles also clearly demonstrate the tunable broadband ferrite device aspect of the phenomenon discussed here.

Data on  $S_{11}$  vs  $f$  profiles were acquired for frequency resolution of 1 MHz. The mode frequency  $f_r$  and the shift  $\Delta f_r$  were determined from the frequency that corresponds to the minimum in the reflected power. The error in  $S_{11}$  for our network analyzers was on the order of  $\Delta S_{11} = 0.03$  dB, corresponding to an absolute error  $\Delta f \approx 20$  MHz in the frequency measurements. The relative error in the mode frequency was much lower than the absolute error and is on the order of 3–4 MHz. In the multidomain state two resonance modes are seen in Fig. 4(a). We identify the modes with the labels (1,1) and (1,1)' since they evolve into these modes (as shown in the data of Fig. 2) in the saturated state. Both the multidomain and single-domain state have a resonance mode that is higher in frequency for two-sided metalized ferrite than for one-sided metalized ferrite [40,44] which allowed us to label the individual modes in Figs. 2 and 4. The two modes in Fig. 4(a) are separated by 750 MHz for zero current. Both modes showed a significant increase in the resonance frequency when a current was applied to the sample. Figure 5 shows the increase in frequency  $\Delta f_r$  vs electric power  $P$ . The frequency shift for the low-frequency (1,1) multidomain mode is shown since Eq. (1) holds true for this mode and the data are sufficient for our analysis. A shift of identical magnitude was measured for the high frequency (1,1)' mode.

Resonance profiles in Fig. 4(b) are for the modes (1,3) and (1,3)' for the sample in the single-domain state. The resonances in the single-domain state are rather broad in comparison to the multidomain case even though one would anticipate a broader resonance absorption in the multidomain case. In thin platelets of ferrites with uniaxial anisotropy, such as SrM, the stripe domains with antiparallel magnetization have equal volumes and are separated by Bloch-type domain walls (DWs). There are two types of eigenmodes in the multidomain state; one is excited by ac magnetic fields parallel to the domain walls and the other by ac field perpendicular to DWs. The mode in our case is the latter one, for which magnetization oscillations

are continuous across the DWs, and hence no high-frequency demagnetizing field appears on DWs. Thus there are no new two- and three-magnon relaxation mechanisms expected in the multidomain resonance and this mode may not have a higher linewidth than for the saturated single-domain state. The broad resonance absorptions in the saturated state in the present case may in part be due to a resonance frequency  $\sim 10\%$  higher than the multidomain resonance and forward volume wave nature of the excitations. This observation needs further investigation.

We also observed splitting of the modes in Fig. 4(b), i.e., ripples, in otherwise smooth  $S_{11}$  vs  $f$  characteristics. The ripples are distortions caused by electrodynamic perturbation of waveguide modes due to the presence of sample. The nature of these ripples are dependent on the sample geometry and location in the waveguide, but their frequency did not change with  $H$  or  $I$ . Figure 5 shows the shift  $\Delta f_r$  in the frequency of the (1,3) mode as a function of the electric power in the saturated state. Values of  $\Delta f_r$  were corrected for the increase in  $T$  due to Joule heating as discussed in Sec. II C. In the saturated state, both (1,3) and (1,3)' modes showed the same frequency shift. We made measurements of  $\Delta f_r$  vs  $P$  for six different static magnetic field values in the range  $H_0 = 4$ –6.5 kOe and found a very weak dependence of  $\Delta f_r$  on  $H_0$  in agreement with the theoretical predictions [Eq. (3)].

The data in Fig. 5 show a much higher current-induced shift in  $f_r$  in the single-domain state than for the multidomain resonance. In the multidomain case any change in  $H_a$  influences the mode frequency, whereas in the saturated state changes in both  $4\pi M_0$  and  $H_a$  contribute to the frequency shift. The  $I$  (or  $E$ -) induced shifts in  $f_r$  in Fig. 5 are the highest reported for any hexagonal ferrite. The maximum frequency shifts in Fig. 5,  $\sim 220$  MHz in the multidomain state and  $\sim 320$  MHz in the saturated state for  $E \sim 1$  kV/cm, are two orders of magnitude higher than shifts of  $\sim 0.6$  MHz in ferromagnetic resonance (FMR) for  $E = 1$  kV/cm in BaM-PZT composites due to strain mediated coupling [16,23].

Next, we utilized the  $\Delta f_r$  vs  $P$  data in Fig. 5 to estimate the changes in the anisotropy field and magnetization with the electric power density  $p = P/V$ , where  $V$  is the sample volume. First, the data for the multidomain state and Eq. (2) were used to estimate the variation in  $\Delta H_a$ . Then we obtained  $\Delta(4\pi M_0)$  vs  $p$  using Eq. (3), the frequency shift data in the saturated state, and previously estimated  $\Delta H_a$  values. Figure 6 shows  $p$  dependence of variations in  $4\pi M_0$  and  $H_a$ . Since there is a linear relationship between the mode frequency and uniaxial magnetic field and magnetization [Eqs. (1) and (3)], the uncertainty in  $\Delta f_r$  corresponds to the uncertainty of 8 Oe in the anisotropy field (or the magnetization) values.

This accuracy sets the size of error bars in Figs. 5 and 6. The anisotropy field in Fig. 6 is found to increase with  $p$ , whereas  $M_0$  decreases with increasing electric power. Both parameters show linear dependence on the electric power density (or  $E^2$ ). Since the uniaxial anisotropy field  $H_a = 2K_u/M_0$ , changes in both  $K_u$  and  $M_0$  will ultimately determine the net variation in  $H_a$ . The variation in the anisotropy field is given by  $\Delta H_a = \frac{\partial H_a}{\partial K_u} \Delta K_u + \frac{\partial H_a}{\partial M_0} \Delta M_0$  and

$$\frac{\Delta H_a}{H_a} = \left( \frac{\Delta K_u}{K_u} - \frac{\Delta M_0}{M_0} \right). \quad (4)$$

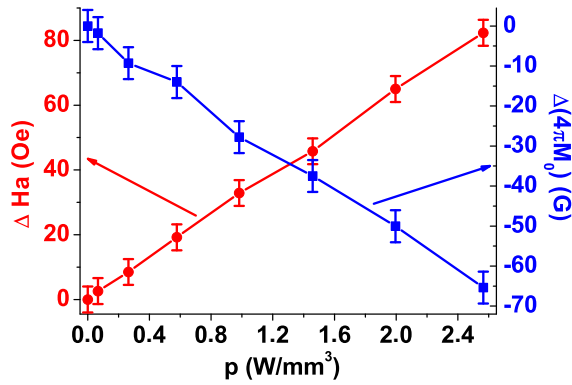


FIG. 6. Current-induced variation in the uniaxial anisotropy field and saturation magnetization as a function of electric power density. Error bars show measurement uncertainty due to the Joule heating.

Although both  $K_u$  and  $M_0$  are expected to decrease with increasing  $p$ , one infers from the results in Fig. 6 that  $|\Delta K_u/K_u| < |\Delta M_0/M_0|$ , so that  $\Delta H_a/H_a > 0$ .

### III. DISCUSSION

The presence of free charge carriers in both spinel and hexagonal ferrites is known to lead to such effects as eddy current losses, magneto-plasmon-polariton formation and propagation, and magnon-electron relaxation processes [45]. Yet, none of these can be the cause of the observed modification of magnetic parameters. The first-principles band-structure calculations for  $SrM$  using the density-function theory show that the electronic structure for this compound is a semiconductor with a direct energy gap of about 0.63 eV and  $n$ -type conductivity along the hexagonal  $c$  axis [31]. Magnetic structure calculations predict that the most stable magnetic structure of the ferrite is ferrimagnet with Fe in the 4f1 and 4f2 sites having the magnetic moments antiparallel to the rest of the Fe.

The electrical conductivity in  $SrM$  is the result of electron hopping between  $Fe^{3+}$  and  $Fe^{2+}$  (“valence exchange”) [32,46], presumably via intermediate oxygen atoms [31]. In stoichiometric  $SrM$  the unit cell is charge balanced and hence the hopping process is impossible. But in ferrites with excess iron or oxygen deficiency, charge imbalance promotes  $Fe^{2+}$  formation and leads to hopping-type electrical conduction [47].

Next we speculate on the possible causes for the current-induced changes in the magnetic order parameters. From the results presented here it is clear that the current conduction process has a significant impact on both the magnetization and anisotropy. In this respect it is similar to effects of Joule heating. The electron hopping, however, is much faster than the heating process and this allowed us to separate the two contributions. As seen in the data of Fig. 6, the observed ME effects are proportional to the power density  $p = P/V = \sigma_{\parallel} E^2 = \sigma_{\parallel} U^2/d^2$ . It is also clear from the  $I$  vs  $U$  data in Fig. 1 that  $\sigma$  is independent of the applied voltage  $U$  only for  $U \sim 0-7$  V and, therefore, changes in  $M_0$  and  $H_a$  are directly proportional to  $E^2$  for low input power. However, nonlinear  $I$  vs  $U$  behavior for  $U > 7$  V needs to be carefully considered for an understanding of the results in Fig. 6 and

for modeling efforts of the nonlinear ME effects in materials with semiconductor-type conductivity. From the expression for  $p$  we expect the strength of ME interactions to increase linearly with  $\sigma$  and decrease with increasing  $d$ . Moreover, a decrease in sample volume will also result in an increase in the sample surface area-to-volume ratio and an efficient transfer of heat generated due to Joule heating to the Cu holder. Thus as the sample size decreases one anticipates weakening of Joule heating related effects on magnetic parameters. Studies on micro- and nanodots of hexaferrites are of interest in this regard.

Next we compare the  $E$  (or  $I$ ) tuning of resonance modes in  $SrM$  with magnetic and  $E$  tuning of resonances in other microwave ferrites and yttrium iron garnet (YIG). Ferrite and YIG devices based on magnetic modes or FMR in general are tuned with a magnetic field produced by an electromagnet or a solenoid. Broadband  $H$  tuning was demonstrated for devices such as resonators and filters [15]. Another tuning procedure involves application of a dc voltage to a ferrite/YIG-ferroelectric composite. Such composites are multiferroics and show coupling between electric and magnetic subsystems that are mediated by mechanical strain and occurs through magnetostriction in the ferrite and converse piezoelectric effect in the ferroelectric. Thus one can control electromagnetic properties of a composite with  $E$  and/or  $H$ . Under the application of an electric field, the ME effect in the composite can be observed in the form of a shift in ferromagnetic resonance (FMR) profile for the ferrite. Voltage tuning of several YIG-PZT and ferrite-PZT devices including resonators, band-pass filters, phase shifters, and delay lines were reported [15]. However,  $E$  tuning is rather weak for hexaferrite-PZT due to low magnetostriction of the former and was on the order of 0.6 MHz of the central frequency for a BaM-PZT resonator for  $E = 1$  kV/cm [16]. In the present study, the tuning is on the order of 250–300 MHz for  $E = 1$  kV/cm (Figs. 4 and 5) and is two orders of magnitude higher than for the strain mediated tuning. Thus this report on nonlinear ME interaction mediated tuning of FMR/magnetic modes is of importance for a different family of broadband  $E$ -tunable hexaferrites devices.

#### A model for the nonlinear ME effects

Next we consider a model for the observed effects. The free-energy density for  $SrM$  (in CGS units) can be expressed as

$$G = G_0 + G_{ME}. \quad (5)$$

Here  $G_0$  is the free energy of the magnetic system in the absence of electric field and includes Zeeman energy and energy associated with exchange interactions, dipole-dipole interaction, and spin-orbital coupling.  $G_{ME}$  is the magnetoelectric energy that is traditionally written as a series with ascending powers of electric and magnetic fields [18,21] and should be consistent with the crystal symmetry of a given material. Specifically, for  $M$ -type hexaferrites with collinear magnetic arrangement both the linear magnetoelectric term  $\alpha_{ij} E_i B_j$  and electrobimagnetic  $\beta_{ijk} E_i B_j B_k$  effects are prohibited [18] since its crystal structure has the center of inversion. For the same reason, all higher-order terms may only contain electric field

in even power. We use  $B$  instead of  $H$  for magnetic field since the former is more appropriate for the magnetically saturated materials with nonzero spontaneous magnetization [18,21]. We therefore express  $G_{\text{ME}}$  as

$$G_{\text{ME}} = -\frac{1}{4\pi} \left( \gamma_{ijk} B_i E_j E_k + \frac{4\pi}{M_0^2} \delta_{ijkl} E_i E_j M_k M_l \right). \quad (6)$$

Here  $\gamma_{ijk}$  denotes the coefficient of the magnetobielectric effect (MBE) and  $\delta_{ijkl}$  is the coefficient for the second-order magnetoelectric term contributing to magnetocrystalline anisotropy constant [30]. Note that ME terms in Eq. (6) are quadratic in  $E$ . That is in agreement with our results in Fig. 6 where all measured changes in magnetic parameters are found to be linearly proportional to electric power density.

Consider first the magnetobielectric term in  $G_{\text{ME}}$  given by

$$G_{\text{ME}}^{\text{MBE}} = -\frac{1}{4\pi} \gamma_{ijk} B_i E_j E_k.$$

The change in the static magnetization due to MBE is given by

$$\Delta M_i^{\text{MBE}} = -\frac{\partial G_{\text{ME}}^{\text{MBE}}}{\partial B_i} = \frac{1}{4\pi} \gamma_{ijk} E_j E_k. \quad (7)$$

The terms  $\gamma_{ijk}$  in the magneto-bielectric tensor are determined by the symmetry properties of the crystal structure of  $\text{SrM}$ . According to the Neumann principle, the  $\gamma_{ijk}$  tensor must be invariant with respect to all transformations connected with the symmetry operation of the specific magnetic point group which is  $6/m\bar{m}'m'$  in the case of  $M$ -type hexaferrites [48]. One needs to consider the  $\gamma_{ijk}$  tensor with all of the 27 coefficients present and then impose the condition that this tensor has to be invariant under all the transformations. Each symmetry operation leads to 27 equations (one for each tensor component) and in turn provides equations relating the tensor coefficients. These equations are then solved to obtain the nonzero tensor components. The magnetobielectric tensor, however, has essentially the same form as the piezomagnetic tensor  $\Lambda_{ijk}$  for this point group [18]. Using the Voigt notation for the indices for the tensor components  $\gamma_{ijk} = \gamma_{il}$  with  $l$  represented by 1 for  $j, k = 11; 2 = 22; 3 = 33; 4 = 23 = 32; 5 = 31 = 13; 6 = 12 = 21$ , one arrives at the following coefficients for the tensor [49]:

$$\gamma_{il} = \begin{pmatrix} 0 & 0 & 0 & 0 & \gamma_{15} & 0 \\ 0 & 0 & 0 & \gamma_{15} & 0 & 0 \\ \gamma_{31} & \gamma_{31} & \gamma_{33} & 0 & 0 & 0 \end{pmatrix}.$$

For the MBE tensor  $\gamma_{ijk}$  we obtained the following nonzero coefficients:  $\gamma_{131} = \gamma_{232} = \gamma_{113} = \gamma_{223}$ ,  $\gamma_{311} = \gamma_{322}$ ,  $\gamma_{333}$ . Hence, the change in magnetization along the sixfold symmetry axis (denoted here with index 3) is

$$4\pi \Delta M_3^{\text{MBE}}(\vec{E}) = \gamma_{311} E_{\perp}^2 + \gamma_{333} E_{\parallel}^2, \quad (8a)$$

where  $E_{\perp}$  and  $E_{\parallel}$  denote static electric-field components perpendicular and parallel to the hexaferrite  $c$  axis. Equation (8a) implies the anisotropic nature of MBE effects in  $\text{SrM}$  with the change in the static magnetization dependent on the direction of electric field. Substituting for  $E$  in Eq. (8a) by  $E_i = \rho_{ij} j_j$ , where  $\rho_{ij} = (\sigma_{ij})^{-1}$  is the anisotropic resistivity tensor [32],

we get

$$4\pi \Delta M_3^{\text{MBE}}(\vec{j}) = \gamma_{311} \rho_{\perp}^2 j_{\perp}^2 + \gamma_{333} \rho_{\parallel}^2 j_{\parallel}^2. \quad (8b)$$

Expression 8(b) does not take into account the dependence of the resistivity tensor components on the electric field and is valid only for materials with ohmic or quasiohmic conductivity. In other cases, e.g., for semiconductors in nonlinear regime, dependence of  $\rho_{ij}$  on  $E$  should be explicitly taken into account.

The input electric power is given by  $P = UI$  with  $U = \int_L \vec{E} d\vec{l}$  and  $I = \int_S \vec{j} d\vec{S}$ . In our case  $\vec{j} = (0, 0, j_{\parallel})$  and therefore  $P = \rho_{\parallel} j_{\parallel}^2 V$ , and sample volume  $V = abd$  ( $a$  and  $b$  are the lateral dimensions of the sample). Thus  $\rho_{\parallel} j_{\parallel}^2 = P/V$  and the magnetization change  $4\pi \Delta M_3$  is quadratic in electric field  $\vec{E}$  and a linear function of electric power density  $P/V$ . The duality of magnetoelectric effect implies that besides electric-field-induced magnetization a magnetic-field-induced polarization should also exist. One obtains for the induced polarization  $\Delta P_i^{\text{MBE}} = -\frac{\partial G_{\text{ME}}^{\text{MBE}}}{\partial E_i}$  and for  $\vec{B} = (0, 0, B_3)$ ,  $\Delta P_i$  is given by

$$4\pi \Delta \vec{P}^{\text{MBE}}(\vec{E}) = 2\gamma_{311} B_3 \vec{E}_{\perp} + 2\gamma_{333} B_3 \vec{E}_{\parallel}. \quad (9)$$

Thus the effect can be treated as a magnetic-field-induced modification of dielectric permittivity:  $\epsilon_{ij} = \epsilon_{ij}^0 + \Delta \epsilon_{ij}(\gamma_{mnp}, \vec{B})$  where the specific form of the  $\Delta \epsilon_{ij}(\gamma_{mnp}, \vec{B})$  term depends on the crystallographic symmetry and direction of flux density vector. Using Eq. (8) and experimental data, one can estimate the value of magnetobielectric coefficient  $\gamma_{333}$ . Our estimates provided  $\gamma_{333} = 5.6 \times 10^{-9} \text{ G m}^2/\text{V}^2$  that corresponds to  $\gamma_{333} = 5.1 \text{ cm/statV}$  in CGS.

Electric-field-induced modification of magnetocrystalline anisotropy is accounted for in a similar way by using the second term in Eq. (6). From symmetry considerations discussed above for the tensor  $\gamma$ , it can be shown that the tensor  $\delta_{ijkl}$  has 21 nonzero coefficients (six independent) for the  $6/m\bar{m}'m'$  point group. The form of tensor  $\delta_{ijkl}$  resembles that of magnetostriction tensor [48]. We obtained the following expression for the free energy in terms of the direction cosines  $\alpha_i = M_i/M_0$ :

$$G_{\text{ME}}^a = -\delta_{ijkl} E_i E_j \alpha_k \alpha_l. \quad (10)$$

Then, by comparing this expression with the uniaxial anisotropy term  $G_o^a = -K_u \alpha_3^2$  (and taking into account the condition  $\sum_i \alpha_i^2 = 1$ ) we find that the change in the uniaxial anisotropy constant is given by

$$\begin{aligned} \Delta K_u &= [\delta_{ij33} - 1/2(\delta_{ij11} + \delta_{ij22})] E_i E_j \\ &= [\delta_{ij33} - 1/2(\delta_{ij11} + \delta_{ij22})] \rho_{im} \rho_{jn} j_m j_n. \end{aligned} \quad (11)$$

Once again this contribution is anisotropic with respect to the electric-field direction. However, corresponding expressions are much more cumbersome than Eq. (8). In our case we have  $\vec{E} = (0, 0, E_3)$ . Using the nonzero tensor coefficients we obtained the following expression for the change in the anisotropy constant:

$$\Delta K_u = (\delta_{3333} - \delta_{3311}) E_{\parallel}^2 = (\delta_{3333} - \delta_{3311}) \rho_{\parallel}^2 j_{\parallel}^2. \quad (12)$$

After substituting for the parameters in Eq. (12) we estimated  $(\delta_{3333} - \delta_{3311}) = -2.97 \times 10^{-2} \text{ erg}/(\text{V}^2 \text{ cm})$  or  $-2.67 \times 10^3$  (dimensionless) in CGS.



#### IV. CONCLUSIONS

In conclusion, observation of room-temperature current/electric-field-induced nonlinear magnetoelectric effects in ferrimagnetic single-crystal  $M$ -type hexaferrite is reported. It is shown that conduction current along the  $c$  axis of the strontium hexaferrite platelet induces variations in the saturation magnetization and uniaxial anisotropy energy. Microwave measurement techniques have been used for estimation of changes in magnetic parameters from magnetostatic mode frequencies for the multidomain ( $H_0 = 0$ ) and single-domain resonances. The measured current-induced frequency shift amounted to  $\approx 220$  MHz in the multidomain state and  $\approx 320$  MHz in the saturated state for  $E \sim 1$  kV/cm ( $I_{dc} = 70$  mA). Changes in the magnetic parameters due to Joule heating were estimated to be negligible in comparison with the observed ME effects. The saturation magnetization is found to decrease and the uniaxial anisotropy field increases linearly with the applied electric power density (or  $E^2$ ). A phenomenological description of the

observed magnetobielectric effects, consistent with intrinsic crystallomagnetic symmetry of hexaferrite, has been proposed and the MBE coupling coefficients have been estimated from data on change in the magnetization and anisotropy field. The MBE effect is of technological importance for current tunable mm-wave ferrite devices such as resonators and filters for use in high-frequency communication systems.

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