

May 16, 2011

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Recommended Citation

Chen, Yajie; Daigle, Andrew; Fitchorov, Trifon; Hu, Bolin; Geiler, Michael; Geiler, Anton; Vittoria, C.; and Harris, V. G., "Electronic tuning of magnetic permeability in Co_2Z hexaferrite toward high frequency electromagnetic device miniaturization" (2011). *Electrical and Computer Engineering Faculty Publications*. Paper 40. <http://hdl.handle.net/2047/d20002210>

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Citation: *Appl. Phys. Lett.* **98**, 202502 (2011); doi: 10.1063/1.3590771

View online: <http://dx.doi.org/10.1063/1.3590771>

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Electronic tuning of magnetic permeability in Co₂Z hexaferrite toward high frequency electromagnetic device miniaturization

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(Received 15 February 2011; accepted 24 April 2011; published online 17 May 2011)

The magnetic and magnetostriction properties of Z-type cobalt-doped barium hexaferrite with perpendicular *c*-axis crystallographic texture are presented. The hexaferrite was utilized as a component in Co₂Z/lead magnesium niobate-lead titanate multiferroic heterostructures whose tunability of permeability with electric field in terms of ferromagnetic resonance shift was supported by experiments and theoretical calculation. A permeability change of 16% was measured by an induced magnetic field of 38 Oe under the application of 6 kV/cm of electric field. These findings lay the foundation for the application of Z-type hexaferrites in tunable rf and microwave devices valued for sending, receiving, and manipulating electromagnetic signals. © 2011 American Institute of Physics. [doi:10.1063/1.3590771]

The rapid and continuous development of wireless communication technologies, especially in mobile communication, has placed significant demands upon both radio-frequency (rf) and microwave devices as well as their constituent materials. Trends toward miniaturization and high performance of high frequency materials include those microwave planar ferrites such as Co₂Z-type ferrite, i.e., Ba₃Co₂Fe₂₄O₄₁.¹ In the Z-type ferrite system the room temperature easy magnetization direction lies in the crystallographic basal plane (*a*-*b* plane) of the hexagonal structure, maintaining a high permeability from ultra high frequency to L-bands (300 MHz–3 GHz) (Ref. 2). It is therefore regarded as a promising candidate for inductor cores, electromagnetic noise absorbers, antenna substrates and a wide range of other microwave devices that send, receive and manipulate electromagnetic signals in these bands.^{3,4}

It is common that microwave devices require magnetic fields to bias and actively tune phase angle or frequency, such as in phase shifters, resonators, tunable filters, inductors, and electronic band gap metamaterials, to name but a few examples.^{5,6} However, it has been a longstanding goal to identify and develop approaches to generating tunable magnetic fields in high frequency miniature electromagnetic devices.

It is noteworthy that the magnetoelectric (ME) coupling effect based on multiferroic (MF) heterostructures has presented great potential for use in multiple technology fields.^{7–10} We propose here that magnetic properties for the Co₂Z-type ferrites can be manipulated with an electric field by means of a MF heterostructure, consisting of a ferrimagnetic Co₂Z ferrite and ferroelectric lead magnesium niobate-lead titanate (PMN-PT) to achieve tunability of permeability and ferromagnetic resonance frequency.

The Co₂Z ferrite powder was synthesized by a chemical process and aligned with a uniaxial pressure within an in-plane rotating magnetic field. The perpendicular *c*-axis oriented hexaferrite was sintered at 1200–1280 °C for a soak time of 2–16 h in air. Details of the sample preparation have

been described elsewhere.⁵ The Co₂Z ferrite was cut and polished to a size of 6 × 4 mm² with a thickness of 170 μm. In order to construct a multiferroic structure, a 0.5 mm-thick ferroelectric PMN-PT crystal was used having a [011] poling with anisotropic in-plane piezoelectric coefficient *d*₃₁ and *d*₃₂.⁸ The Co₂Z slab was plated with Au on both sides then bonded to a PMN-PT slab using a quick curing ethyl cyanoacrylate adhesive. We emphasize that the *c*-axis of the hexaferrite was aligned perpendicular to the plane of ferrite. Ferromagnetic resonance (FMR) measurements were carried out using a microwave cavity excited in a TE₁₀₂ mode at X-band (*f*=9.55 GHz), where both the external field, H₀, and microwave magnetic field, *h*_{*c*}, was always aligned in the plane of the sample. ME coupling measurements were carried out in which H₀ was alternately aligned parallel to the *d*₃₁ (i.e., longitudinal) and *d*₃₂ (transverse) directions of the PMN-PT crystal. The magnetostriction coefficient (λ) of the Co₂Z was measured over a magnetic field range of 0–2000 Oe. In order to obtain a direct magnetic field dependence of permeability, a single ferrite toroid of dimension 7 mm OD × 3 mm ID, where OD and ID are the outer and inner diameters, respectively, was employed to examine microwave permeability (*f*=0.5–5 GHz) by the coaxial air-line technique using an Agilent vector network analyzer. Note, the measurement gives rise to an ac dispersion of permeability, which is a type of nonresonance relaxation (i.e., an ac field is parallel to magnetization), other than FMR, as described in Ref. 11.

A magnetic hysteresis loop for the oriented Co₂Z ferrite as the magnetic field was applied along the plane of the sample is presented in Fig. 1. The oriented ferrite having an average grain size of 4–5 microns showed saturation magnetization, 4πM_s, of 3150 G and a coercivity value of ~5 Oe. As depicted in the inset to Fig. 1, a scanning electron microscopy (SEM) image of the sample top-view illustrates the *c*-axis orientation perpendicular to the sample surface as is evidenced by aligned hexagonal platelets. X-ray diffraction analysis indicated an orientation factor, *f*=0.3–0.5 where *f*=[P–P₀]/[1–P₀] (Ref. 12) for the perpendicular *c*-axis oriented planar ferrite.

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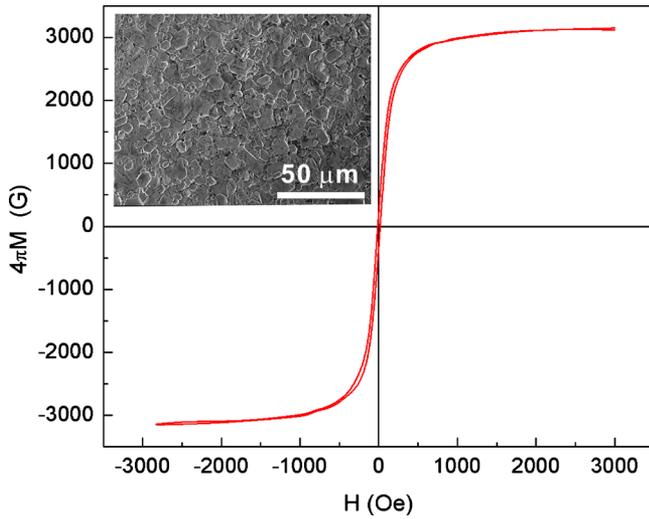


FIG. 1. (Color online) Magnetic hysteresis loop and surface morphology as a SEM image (see inset) of a perpendicular c -axis oriented Co_2Z ferrite compact.

The dependence of strain with magnetic field, i.e., magnetostriction, of the oriented Co_2Z ferrite is depicted in Fig. 2. The longitudinal and transverse orientations represent an external magnetic field aligned parallel and transverse to the direction of strain, respectively. Saturation magnetostriction coefficient is determined to be -6 ppm, in terms of the longitudinal and transversal measurement.

Figure 3 illustrates the measurement of ferromagnetic resonance for the $\text{Co}_2\text{Z}/\text{PMN-PT}$ heterostructure under an applied electric field and the zero field condition, respectively. The magnetic field was applied along the d_{31} direction of the PMN-PT crystal. The FMR line shape is clearly distorted and nonsymmetrical, which may arise from the sweeping field being insufficient to saturate the sample.¹³ Nevertheless, the results indicate that the oriented Co_2Z ferrite has an FMR linewidth of 736 Oe and 755 Oe under an electric field of zero and 6 kV/cm, respectively. Interestingly, a 6 kV/cm electric field applied across the PMN-PT crystal results in a pronounced shift of FMR field (H_r) from 840 to

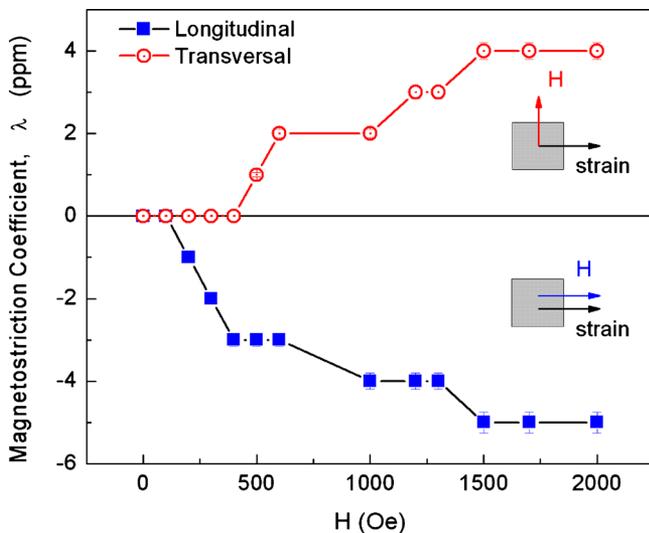


FIG. 2. (Color online) Magnetostriction measurements for a perpendicular c -axis oriented Co_2Z ferrite compact. Longitudinal and transversal represent an applied magnetic field parallel and transversal to the direction of strain observed, respectively.

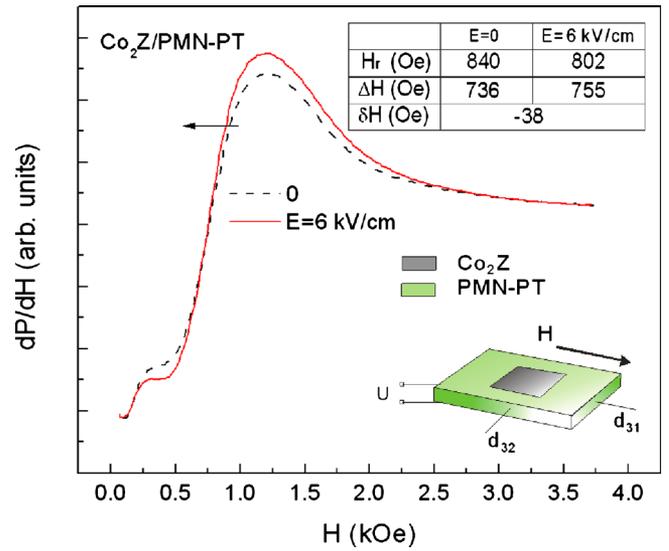


FIG. 3. (Color online) Ferromagnetic resonance measurements for a perpendicular c -axis oriented Co_2Z ferrite slab bonded to a piezoelectric PMN-PT crystal under the application of an electric field of 6 kV/cm and zero electric field condition, respectively. dP/dH represents the derivative of microwave absorption power with respect to the external magnetic field. The inset illustrates the measurement geometry and field configuration of the multiferroic heterostructure. Here, the longitudinal orientation represents the direction upon which the applied magnetic field aligns along d_{31} of the PMN-PT slab.

802 Oe. The lowering of the resonance field implies an enhancement of the induced magnetic field inside the ferrite along the applied magnetic field direction. This result is understood in terms of the relationship between magnetostriction and piezoelectric coefficients as expressed in Eq. (1) (Ref. 14)

$$\delta H_E = 3 \frac{\partial \lambda}{\partial M} \cdot d_{31} \cdot E \cdot Y \cdot (\cos^2 \varphi - \nu \sin^2 \varphi), \quad (1)$$

where δH_E denotes a stress-induced magnetic anisotropy field due to an electric field applied across piezoelectric substrate, φ is the angle between the magnetization and applied stress (here $\varphi=0^\circ$), and ν denotes the Poisson ratio of the ferrite material. E and Y represent the applied electric field, and the Young's modulus of the magnetic material, respectively. It was assumed that the deformation of the ferrite mirrors that experienced by the PMN-PT substrate. Note, Eq. (1) predicts that the stress-induced magnetic field is proportional to the derivative of the magnetostriction coefficient (λ) with respect to the magnetization (M). In the present case, both λ_s and d_{31} are negative, leading to an induced magnetic field parallel to the applied magnetic field. Furthermore, by applying the experimentally determined parameters of $\lambda = -6$ ppm, $M = 250$ G, $Y = 2 \times 10^{12}$ dyn/cm²,¹⁵ $d_{31} = -1500$ pC/N, and $E = 6$ kV/cm, an estimate from Eq. (1) suggests an induced field of 74 Oe. This estimate is nearly double the measured value; a discrepancy attributed to the lack of efficient strain transfer from the PMN-PT to the ferrite slab yielding a measured value of ~ 38 Oe.

For a planar hexagonal ferrite, Equation (2) can be used to describe FMR for a perpendicular c -axis oriented polycrystalline Co_2Z -type ferrite with an isotropic in-plane arrangement of basal plane, whereupon the external magnetic field lies within the a - b plane;¹⁶

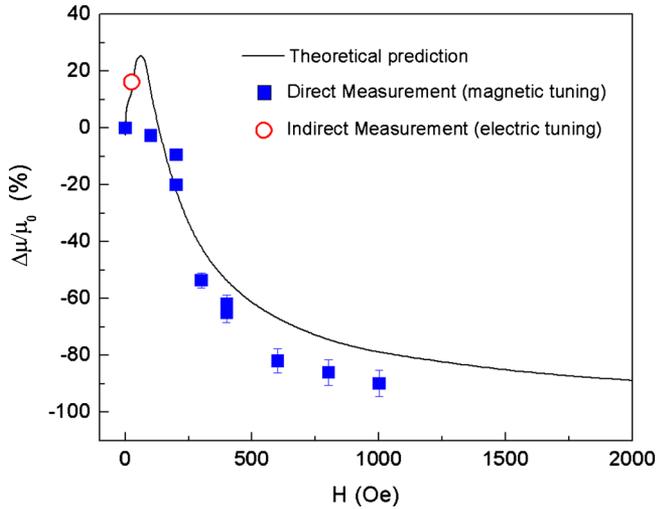


FIG. 4. (Color online) Tunability of permeability with a biased magnetic field. The solid line represents the theoretical prediction derived from Eq. (2); square and circle symbols denote direct measured data tuned by a biased magnetic field and indirect measured value tuned by an electric field, respectively.

$$f = \gamma' \sqrt{H(H + H_A + 4\pi M)}, \quad (2)$$

where H and H_A denote external and perpendicular anisotropy fields, respectively. The FMR measurement indicates $H_A = 9.85$ kOe, whereas the resonance frequency shift is calculated to be 250 MHz at X-band with respect to an E-field of 6 kV/cm, yielding a frequency tunability of 41.6 MHz/kV cm⁻¹, which is comparable to other multiferroic structures operating in the range of 3–15 MHz/kV cm⁻¹.^{17,18}

In order to obtain the tunability of permeability controlled by an electric field applied across the PMN-PT slab, we must determine a relationship between permeability and magnetic field. Therefore, we deduce a mathematical relation describing the change in permeability with magnetic field. At low frequencies, i.e., off-resonance frequency, the permeability can be expressed as,

$$\mu \approx \frac{4\pi M}{H + H_\phi} + 1. \quad (3)$$

Here, we define the permeability tuning factor with magnetic field as,

$$\begin{aligned} \frac{\Delta\mu}{\mu_0} &= \frac{\mu(H) - \mu(0)}{\mu(0)} \\ &= \frac{\frac{4\pi M}{H + H_\phi} - \frac{4\pi M_r}{H_\phi}}{\frac{4\pi M_r}{H_\phi} + 1}. \end{aligned} \quad (4)$$

Note, $M(H)$ is expressed as a function of H , and when $H=0$ the $4\pi M$ in Eq. (3) becomes the remnant magnetization, $4\pi M_r$. According to Eq. (4), we plot the trend of permeability with respect to the bias magnetic field in terms of remnance, $4\pi M_r = 275$ G, and the in-plane anisotropy field, $H_\phi = 20$ Oe. The solid line plotted in Fig. 4 represents the theoretical prediction of the dependence of the rate of change in permeability with respect to the applied magnetic field.

To verify the accuracy of the theoretical prediction, the complex permeability was measured only for a single ferrite toroid with different magnetic fields. As mentioned above, the μ'' absorption is a type of nonresonance relaxation.² The measured data plots closely to the theoretical curve.

Finally, the permeability change tuned by 38 Oe of induced magnetic field under the application of 6 kV/cm of electric field was measured, indicating the permeability changed by 16%, i.e., a permeability tunability of 2.7% kV/cm⁻¹. Since the Co₂Z ferrite has a zero field resonance frequency of 1.6 GHz, the nonresonance relaxation frequency is presumably tuned by 256 MHz due to 16% of permeability change in accordance with the limitation imposed by Snoek's law. Furthermore, the indirectly measured value is also plotted in Fig. 4 as a hollow circle symbol and it is seen to also plot on the theoretical curve.

In summary, we have reported the magnetic and magnetostriction properties of Z-type cobalt-doped barium hexaferrite with perpendicular c -axis crystallographic texture. The Co₂Z hexaferrite was utilized as a component in a Co₂Z/PMN-PT multiferroic heterostructure. A theoretical calculation of permeability tunability under biased magnetic fields predicted accurately the experimental response of both the planar hexaferrite material and heterostructure. The measurements of direct magnetic field tuning and indirect electric field tuning of permeability for Co₂Z ferrite was demonstrated, indicating an electric field tuning of 16% of the permeability which corresponded to a FMR frequency shift of 250 MHz. These findings lay the foundation for the application of Z-type hexaferrites in tunable rf and microwave devices.

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