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Nonlinear magneto-electric effects in ferromagnetic-piezoelectric composites

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ABSTRACT

Theory and results of a systematic study on the nature of nonlinear magnetoelectric (ME) interactions in layered ferromagnetic and ferroelectric composites are discussed. The model that considers the nonlinearity associated with magnetostriction of the ferromagnet is to result in (i) a dc component and (ii) frequency doubling when the composite is subjected to an ac magnetic field. In the presence of two ac magnetic fields of different frequencies, nonlinear effects give rise to generation of ME voltages at the sum and difference of the fields frequencies. The efficiencies of nonlinear ME interactions are shown to be a function of the second derivative of the magnetostriction with respect to the bias magnetic field. The predictions of the model are compared with data for bilayers of lead zirconate titanate (PZT) and ferromagnetic layers with wide variations in saturation magnetostrictions and saturation magnetic fields, i.e., an amorphous ferromagnetic (AF) alloy, Ni, or permendur. Under linear excitation conditions an enhancement in the ME voltage is measured when the ac magnetic field is applied at the acoustic mode frequencies. Under nonlinear excitation conditions the mechanical deformation and the ME response occur at twice the excitation frequency and the AF-PZT composite shows a much higher nonlinear ME effects. In addition, the AF-PZT shows an efficient frequency mixing than the samples with Ni or permendur when subjected to two ac magnetic fields. The frequency mixing is shown to be of importance for magnetic field sensor applications.

1. Introduction

Magneto-electric (ME) interaction in layered composites consisting of ferromagnetic (FM) and piezoelectric (PE) phases manifests as a change of P under action of an magnetic field H (direct ME effect) or as a change in magnetization M (or an induced anisotropy field) by means of an electrical field E (converse ME effect) [1,2]. The interaction arises because of combination of magnetostriction of the FM layer and piezoelectricity of the PE layer through mechanical coupling between the layers. The ME effects in the composites opens up possibilities for devices based on mutual conversion of electric and magnetic fields, such as high sensitivity magnetic field sensors, high frequency signal processing devices, data storage elements, and energy harvesting [3,4].

Most of the studies carried out to-date on FM-PE composites were devoted to linear ME interactions in ac magnetic (electric) fields when response of the sample was measured at the frequency of excitation field and the voltage (magnetic) response was a linear function of the ac field. But the FM materials are characterized by nonlinear magnetostriction λ vs field dependence [5]. Similarly PE materials are characterized by nonlinear piezoelectric coefficient d vs electric field dependence [6]. Thus, under high H or E, nonlinear ME effects are expected in the composites. Nonlinear ME effects were reported by several groups [7–13], including frequency doubling of the voltage response at low frequencies and under conditions of acoustical resonance [11,12]. Frequency mixing of the voltage response for magnetic fields with different frequencies applied to ME composites was observed [14–16].

Here we discuss the results of a comparative study on the nature of nonlinear ME interactions associated with magnetostriction in composites with PZT and a variety of FM layers. We also discuss a model that allows estimation of the ME response using known values of material parameters for the composite. The nonlinear effects were studied under the acoustical resonance conditions. Potential applications for the nonlinear ME interactions are also discussed.

In the first part of the manuscript a theoretical model, describing the origin and the basic nonlinear ME effects in composite structures is proposed. The second part of the paper describes the samples and measuring techniques used. Main experimental results of the investigations carried out and comparison of these results with predictions of the theory are given in the third part of

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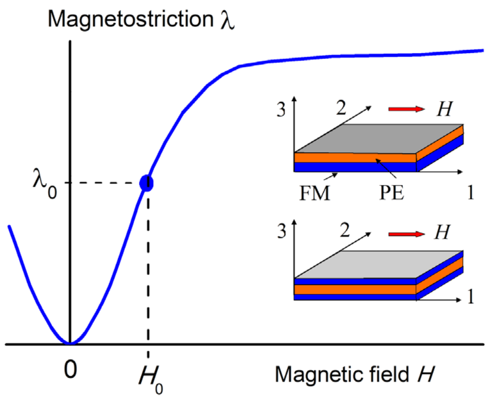

Fig. 1. Typical dependence of longitudinal magnetostriction λ on magnetic field H for a ferromagnet (FM). The insert shows a planar ferromagnetic (FM) - piezo-electric (PE) composites.

the paper. The concluding part of the paper deals with potential utility of nonlinear ME interactions for magnetic field sensing.

2. Theory for nonlinear ME effects

We consider here a model for nonlinear effects in FM-PE composites structures due to nonlinear dependence of magnitostriction λ vs magnetic field H as shown in Fig. 1. A mechanically bonded FM-PE bilayer and a FM-PE-FM trilayer as in Fig. 1 are considered. The layers are in the (1,2) plane with the PE layer of thickness a_p. The FM layer in the bilayer has a thickness a_m, and the FM layers in the trilayer has thickness $a_m/2$ each. The sample is magnetized with a bias magnetic field H which is parallel to the plane and along the direction -1. The strains S, the stresses T, the magnetic field H in the FM layers, the electric field E and electric induction D in the PE layer are related to each other with the following expressions [17]:

$$S_1^p = s_{11}^p T_1^p + d_{31} E_3$$
$$S_1^m = s_{11}^m T_1^m + \lambda(H)$$
$$D_3^p = \varepsilon\varepsilon_0 E_3 + d_{31} T_1^p \qquad (1)$$

The indices p and m in Eq. (1) represent the PE layer and the FM layer, respectively, $\lambda(H)$ is the longitudinal magnetostriction of the FM layer, d_{31} is the piezoelectric coefficient of the PE layer, s_{11}^p and s_{11}^m are the elastic stiffness of the materials, ε is the relative permittivity of the PE layer, and $\varepsilon_0 = 8.85\times10^{-12}$ F/m is the dielectric constant.

The FM-PE bilayer, with width b assumed to be much smaller than the length L, will have a bending deformation under a magnetic field. The strains $S_1^p(z)$and $S_1^m(z)$ will arise in the layers with linear variation along the sample thickness. A neutral-plane with $S=0$ will appear inside the structure at a distance g from the interface boundary. Using Eq. (1), the condition of strain continuity at the interface boundary$S_1^p = S_1^m$, the condition of the sample equilibrium along axis-*1* $a_p T_1^p + a_m T_1^m = 0$, and condition of the structure equilibrium about the rotation around axis 2 passing through the neutral -plane, one gets the stress $T_1^p(z)$in the PE layer. Then, by using the open-circuit condition $D_3^p = 0$, one gets the electrical field $E_3(z)$ in the PE layer. Finally, by integrating $E_3(z)$ over thickness a_p of the PE layer one obtains the expression for the dc voltage u generated in the bilayer in the following form:

$$u = A d_{31}\lambda(H) \qquad (2)$$

Explicit expression for voltage u generated in the bilayer structure is given in the Appendix.

The symmetric FM–PE–FM trilayer will deform only in the (1,2) plane under a magnetic field. In this case the deformations S_1^p and S_1^mwill be uniform across the thickness of the sample. By using Eq. (1), the condition of strain continuity at the interface boundary$S_1^p = S_1^m$, the condition of the structure equilibrium along axis *1*: $a_p T_1^p + a_m T_1^m = 0$, one gets the stress T_1^p in the PE layer. Then, by using the open circuit condition $D_3^p = 0$ one gets the electrical field E_3 in the PE layer. Finally, the voltage u generated by the sample can be expressed as

$$u = B d_{31}\lambda(H). \qquad (3)$$

Explicit expression for voltage u generated by the three-layer is also given in the Appendix.

The coefficients A and B in Eqs. (2) and (3) depend only on the mechanical, geometrical, and electrical parameters of the layers. For more complicated structures, containing several FM or PE layers, the expressions for the generated voltage have the same forms as Eqs. (2) and (3) but with different coefficients. It is important to note that for all composites the ME voltage is a linear function of the magnetostriction $u \sim \lambda(H)$.

Let us consider now the composite response to a magnetic field $H = H_0 + h(f)$, where H_0 is the bias field. Fig. 1 shows typical dependence of the magnetostriction on magnetic field $\lambda(H)$ for ferromagnets. Note that sign of λ is not changed upon reversal of H_0. Upon expanding the magnetostriction as a Taylor series in the vicinity of H_0, we get

$$\lambda(H) = \lambda(H_0) + qh + (1/2)ph^2 + \ldots \qquad (4)$$

where $q = \partial\lambda/\partial H|_{H_0}$and $p = \partial^2\lambda/\partial H^2|_{H_0}$are the first and the second derivatives of λ with respect to the field at $H = H_0$, respectively. In Eq. (4) q is the piezomagnetic coefficient and p is the nonlinear piezomagnetic coefficient. Since this work is concerned with the sample response to simultaneous presence of two ac magnetic fields with frequencies of f_1 and f_2 and amplitudes of h_1 and h_2 (h_1, $h_2 \ll H_0$):

$$h(t) = h_1 \cos(2\pi f_1 t) + h_2 \cos(2\pi f_2 t). \qquad (5)$$

From Eqs. (2)–(4), one obtains the following expression for the voltage generated in the composite

$$u = u^{(0)} + u_1^{(1)} \cos(2\pi f_1 t) + u_2^{(1)} \cos(2\pi f_2 t) + u_1^{(2)} \cos(4\pi f_1 t) + u_2^{(2)} \cos(4\pi f_2 t) + u_{mix} \cos[2\pi(f_1+f_2)t] + u_{mix} \cos[2\pi(f_1-f_2)t]. \qquad (6)$$

The terms in Eq. (6) correspond to the following contributions to the ME voltage:

(a) A dc term $u^{(0)} = Ad_{31}[\lambda(H_0) + (1/4)p(h_1^2 + h_2^2)]$ that describes at $\lambda(H_0) = 0$ the effect of ac magnetic fields. The voltage $u^{(0)}$ is proportional to the nonlinear piezomagnetic coefficient p.

(b) AC components with amplitudes $u_1^{(1)} = Ad_{31}qh_1$ and $u_2^{(1)} = Ad_{31}qh_2$, and frequencies f_1 and f_2, respectively, correspond to the well known direct ME effect. Amplitudes of this components depend on H_0 and reach a maximum at $H_0 = H_m$ for maximum value of the piezomagnetic coefficient q. Efficiency of the linear ME interaction is given by the coefficient $\alpha_E^{(1)} = u^{(1)}/(a_p h_1)$ and measured in the units V cm^{-1}Oe^{-1}.

(c) The ac components $u_1^{(2)} = (1/4)Ad_{31}ph_1^2$ and $u_2^{(2)} = (1/4)Ad_{31}ph_2^2$ with frequencies $2f_1$ and $2f_2$, respectively, describe effect of the frequency doubling [7–13]. Efficiency of the frequency doubling is proportional to p. The efficiency of the frequency doubling is defined as $\alpha_E^{(2)} = u^{(2)}/(a_p h^2)$ with dimension of V cm^{-1}Oe^{-2}.

(d) The components with amplitude $u_{mix} = (1/2)Ad_{11}ph_1h_2$ and frequencies f_1+f_2 and $|f_1-f_2|$ describe the processes of