



# Theoretical study on nonlinear magnetoelectric effect and harmonic distortion behavior in laminated composite



Yang Shi <sup>a, b</sup>, Yuan-Wen Gao <sup>a, b, \*</sup>

<sup>a</sup> Key Laboratory of Mechanics on Disaster and Environment in Western China attached to the Ministry of Education of China, Lanzhou University, Lanzhou, Gansu 730000, PR China

<sup>b</sup> Department of Mechanics and Engineering Sciences, College of Civil Engineering and Mechanics, Lanzhou University, Lanzhou, Gansu 730000, PR China

## ARTICLE INFO

### Article history:

Received 23 April 2015

Received in revised form

13 May 2015

Accepted 20 May 2015

Available online 20 June 2015

### Keywords:

Magnetoelectric effect

Temperature effect

Quadratic harmonic

Harmonic distortion

## ABSTRACT

A theoretical model is established for nonlinear magnetoelectric (ME) effect and harmonic distortion behavior in laminated composite, in which the complex nonlinear mechanical-thermo-magnetic characteristics of magnetostrictive phase are taken into consideration. Both the low-frequency and the resonant ME effects are derived by the elastic mechanics method. In this model, the output ME voltage is divided into three parts: a dc term, a linear ac term and a nonlinear ac term. Considering the temperature effect, this model is adopted to predict the dependences of ac voltages on bias magnetic field, frequency of ac magnetic field, the volume fraction of piezoelectric phase and interfacial parameter. Then, the competition relationship between linear and nonlinear ac voltages is discussed. To clearly describe the nonlinear characteristics of the proposed sample, a distortion factor  $\theta$  is used for characterizing the harmonic distortion behaviors. The dependences of  $\theta$  on the frequency of ac magnetic field and temperature under different ac magnetic fields are also studied. Finally, the predictions of harmonic distortion behaviors with changing bias magnetic field are obtained. The calculated linear ME coefficient shows a good agreement with the previous experimental data. It showed that: linear ac voltage and linear piezomagnetic coefficient present the same trend as the bias magnetic field increases, and nonlinear ac voltage shows the same trend as the absolute value of nonlinear piezomagnetic coefficient with increasing bias magnetic field; with temperature increasing, the output ME voltages decrease; operating temperature has a minimal effect on the required magnetic field corresponding to the optimal value of the output ME voltages, while it cannot alter the optimal value of piezoelectric phase's volume fraction; the nonlinear ac voltage presents more resonant frequencies than the linear one in the same range. Also, a definite multiple relationship is observed between the corresponding resonant frequencies of linear and nonlinear voltages; a larger  $\theta$  leads to a more serious harmonic distortion, which is influenced by the frequency of ac magnetic field and temperature; bias magnetic field can be applied to adjust the harmonic distortion behaviors.

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## 1. Introduction

Magnetoelectric (ME) composites can achieve magneto-electricity conversion by mechanical coupling between magnetostrictive materials and piezoelectric materials, which have great potential applications in magnetic sensors, actuators, energy harvesters, memories and other ME devices [1–5]. Compared with the

single-phase materials and bulk materials, such as  $\text{Ni}_{0.92}\text{C}-0.03\text{Mn}_{0.05}\text{Fe}_2\text{O}_4-\text{BaTiO}_3$  [6] and  $\text{Ni}_{0.8}\text{Co}_{0.1}\text{Cu}_{0.1}\text{Fe}_2\text{O}_4-\text{PbZr}_{0.5}\text{Ti}_{0.5}\text{O}_3$  [7], the laminated composites exhibit much stronger ME coupling at the room temperature and thus arise much attention [8–11].

To enhance ME coefficient, some giant magnetostrictive materials (GMM) were employed to manufacture ME composites due to their large magnetostrictive strains (about 1000 ppm) [12–17]. To our knowledge, giant magnetostrictive materials show some complex nonlinear mechanical-thermo-magnetic characteristics, causing the nonlinear ME coupling in laminated composites. In the past decades. Many researches have been only conducted on ME

\* Corresponding author. Key Laboratory of Mechanics on Disaster and Environment in Western China attached to the Ministry of Education of China, Lanzhou University, Lanzhou, Gansu 730000, PR China.

E-mail address: [ywgao@lzu.edu.cn](mailto:ywgao@lzu.edu.cn) (Y.-W. Gao).

coupling based on a linear magnetostrictive constitutive relation [18,19]. Although they have revealed the effects of volume fraction, frequency of magnetic field on ME effect, the nonlinear relationship between applied magnetic field and ME effect cannot be described properly. A simple quadratic constitutive relation was selected to obtain the high-order ME effect and discuss the effects of the applied magnetic field [20,21]. However, their results were inaccurate at the high range of magnetic field. Besides, some high-order theories for piezoelectric and piezomagnetic materials were presented to evaluate the nonlinear piezoelectric effects and investigate the nonlinear magnetolectric effects [22–24]. For example, Yang and Batra [22] proposed the second-order theory for piezoelectric materials with symmetry class 6 mm and class mm2. Feng et al. [23,24] developed second-order theories for both piezoelectric and piezomagnetic materials. Their works form the basis for, respectively, the future nonlinear analysis of dynamically loaded structures composed of piezoelectric ceramic and magneto-electroelastic materials. These nonlinear models should be useful in the investigation of the mechanics and physics of novel materials undergoing large nonlinear deformations. However, the effects of pre-stress and temperature on the properties of magnetostrictive materials cannot be studied by theories mentioned above. Zheng and her co-workers [25,26] proposed some more accurate constitutive relations to describe the nonlinear behavior of GMM, which were widely applied to study nonlinear ME coupling [27–32]. For example, Wang and Zhou [28] developed a nonlinear magneto-electric coupling model for the magnetostrictive–piezoelectric trilayer, in which the magnetic–mechanical coupling behavior is considered. Zhang and Gao [29] established a theoretical model for circular-shaped magnetolectric layered structure based on a 2D nonlinear mechanical–magnetic coupling constitutive relation. Based on these works, Zhou et al. [30,31] studied the effects of pre-stress and temperature on ME effect. Also, they established a model of resonance mechanical loss in magnetolectric laminated composite, which considers both the bias field and pre-stress effects [32]. Nevertheless, all these works have not systematically discussed the frequency multiplying behaviors and the dependence of nonlinear ME effect on the applied ac magnetic field as well as they are not suitable for the study of high order ME response under a large ac magnetic field.

Recently, a lot of experimental researches have reported some new phenomena in ME response, such as the appearances of frequency multiplying behaviors, high order harmonic and harmonic distortion. K. E. Kamentsev et al. [33] first found frequency doubling at very low frequencies ranging from 1 mHz to 1 Hz, widening the application range of ME effect. Ma et al. [34] reported a novel frequency multiplier based on frequency doubling behavior. Their devices can work in a broad frequency range and be switched by a low bias magnetic field, offering potential opportunities for frequency multipliers in electrical applications. Zhang et al. [35] demonstrated the dependences of frequency multiplying behavior on the dc magnetic field and the ac magnetic field frequency in a Metglas/PZT laminate composite. These phenomena open up possibilities for the applications in frequency doubling devices and other tunable nonlinear ME devices. Most recently, Fetisov et al. [36] observed strong nonlinear ME effects in a composite of ferromagnetic alloy and a PZT bimorph in the absence of bias magnetic field. Later, Burdin et al. [37] discussed the nature of nonlinear ME interactions in layered ferromagnetic and ferroelectric composites. Xu et al. [38] investigated the nonlinear harmonic distortion behavior, and they revealed that the ME distortion is the competition result of linear piezomagnetic and quadratic magnetostrictive effect. Despite all this, with these interesting observations, there is only few works aiming to explain the complex nonlinear ME coupling, and discuss the quadratic harmonic and

harmonic distortion theoretically.

In this paper, a theoretical model of nonlinear ME coupling effect is established by using a mechanical-thermo-magnetic magnetostrictive constitutive relation and a linear piezoelectric constitutive relation. Considering the temperature effect, we systematically study both the low-frequency and the resonant ME effects. For the low-frequency case, the dependences of the applied magnetic field, volume fraction and interfacial parameter on linear and nonlinear ME voltage are obtained. For the frequency-dependent case, the dependences of different order ME voltages on the frequency and ac magnetic field are revealed. Further, the competitive relation between linear and nonlinear ME voltages are discussed to understand the nonlinear harmonic distortion.

## 2. Theoretical framework

### 2.1. Theoretical model

In this section, we consider a theoretical L-T model [5] for the magnetolectric laminated composite shown in Fig. 1. Directions 1, 2, and 3 are along the x-, y-, and z-axes, respectively. The length, width and total thickness of the sample are denoted as  $L$ ,  $w$  and  $t$ , respectively. The thicknesses of magnetostrictive and piezoelectric phases are  $t^m$  and  $t^p$ . The operating principle of the model can be described as: when a combined magnetic field  $H_{dc} + H_{ac}\cos(2\pi f\tau)$  is applied along the direction 1, an induced electric field  $E_3$  will generate along the thickness direction in the piezoelectric phase due to the bonding effect between two phases. Assuming that  $L \gg w \gg t$ , we consider the stress and strain have only one component. To describe the nonlinear characteristics of magnetostrictive material, Zheng–Sun model [26] is selected as follows:

$$\varepsilon_1^m = \frac{\sigma_1^m}{E_s} + \alpha^m \Delta T - \frac{\beta \Delta T M_1^2}{M_s^2} + \begin{cases} \lambda_s \tanh\left(\frac{\sigma_1^m}{\sigma_s}\right) + \frac{\lambda_s}{M_s^2} \left[1 - \tanh\left(\frac{\sigma_1^m}{\sigma_s}\right)\right] M_1^2, & \frac{\sigma_1^m}{\sigma_s} \geq 0 \\ \frac{\lambda_s}{2} \tanh\left(\frac{2\sigma_1^m}{\sigma_s}\right) + \frac{\lambda_s}{M_s^2} \left[1 - \frac{1}{2} \tanh\left(\frac{2\sigma_1^m}{\sigma_s}\right)\right] M_1^2, & \frac{\sigma_1^m}{\sigma_s} < 0 \end{cases} \quad (1)$$

$$H_1 = \frac{1}{\eta} f^{-1} \left(\frac{M_1}{M_s}\right) + \frac{2\beta \Delta T \sigma_1^m M_1}{\mu_0 M_s^2} - \begin{cases} \frac{2\lambda_s}{\mu_0 M_s^2} \left\{ \sigma_1^m - \sigma_s \ln \left[ \cosh\left(\frac{\sigma_1^m}{\sigma_s}\right) \right] \right\} M_1, & \frac{\sigma_1^m}{\sigma_s} \geq 0 \\ \frac{2\lambda_s}{\mu_0 M_s^2} \left\{ \sigma_1^m - \frac{\sigma_s}{4} \ln \left[ \cosh\left(\frac{2\sigma_1^m}{\sigma_s}\right) \right] \right\} M_1, & \frac{\sigma_1^m}{\sigma_s} < 0 \end{cases} \quad (2)$$

where, the right superscript  $m$  and the subscript 1 refer to magnetostrictive phase and direction, respectively;  $\varepsilon_1^m$ ,  $\sigma_1^m$ ,  $M_1$ ,  $H_1$ , and  $\Delta T$  represent strain, stress, magnetization, magnetic field and the change in temperature relative to 0 °C, respectively;  $\lambda_s$ ,  $\sigma_s$ , and

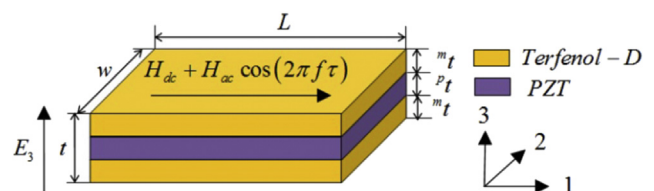


Fig. 1. Configuration of magnetolectric laminated composite.

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