Composite Structures 155 (2016) 107–117

Contents lists available at ScienceDirect

Composite Structures

journal homepage: www.elsevier.com/locate/compstruct

Symmetric relationships between direct and converse magnetoelectric effects in laminate composites

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ARTICLE INFO

Article history:
Received 15 March 2016
Revised 30 July 2016
Accepted 3 August 2016
Available online 05 August 2016

Keywords:
Laminates
Electrical properties
Magnetic properties

ABSTRACT

Direct magnetoelectric (DME) and converse magnetoelectric (CME) effects are two opposite processes in magnetoelectric (ME) materials. An ME theoretical model about the longitudinal vibration was proposed to research their relationships in a laminate composite based on equivalent circuit. Following the piezoelectric and magnetostrictive constitutive equations, we deduced magnetic-mechanical-electric equations, and then built a symmetric equivalent circuit about the ME coupling, based on which we analyzed the CME and DMD effects equivalently. A simple laminate composite was prepared by bonding Terfenol-D and Pb(Zr,Ti)O_3 plates together for a experimental research. Theoretical calculations meet the experimental results very well. The CME and DME effects exhibit enhanced energy transitions at the resonance frequency with a large phase shift of about π. The ME composite shows CME and DME frequency multiplying behaviors and inhomogeneous CME and DMD responses. The CME and DME as well as their resonance frequencies present nonlinear characteristics with the bias magnetic field. The CME resonance frequency is lower than the DME resonance frequency in a same sample. CME decreases while DME increases with the magnetostrictive layer. The theoretical model plays an important role in the comprehensive understanding of ME properties, especially the CME effect, and design of ME devices such as magnetic-electric field sensors, energy harvesting transducers and solid tunable transformers.

1. Introduction

Electric field (***E***) and magnetic field (***H***) are closely linked to each other by Maxwell's equations in classical electromagnetism. This link is dynamic in essence, as moving charges generate a magnetic field and a changing magnetic field produces an electric field. Such connection between magnetism and electricity has had a tremendous impact on science and technology. In a solid, a similar coupling between the magnetization ***M*** and electric polarization ***P*** was first considered by Pierre Curie. The mutual control of magnetism and electricity is named as magnetoelectric (ME) effect, which is characterized as an electric polarization induced by an external magnetic field or a magnetic response to an applied electric field, named as direct magnetoelectric (DME) and converse magnetoelectric (CME) effects, respectively [1]. The ME effect facilitates as effective conversion between electric energy and magnetic energy. The ME coupling in single phases is largely a consequence of the relativistic spin-orbit coupling that links the electron spin to the field generated by the motion of the electron. Hence, most materials that order either electrically or magnetically exhibit a weak coupling between magnetic and electric properties [2,3]. Besides, the ME effect in the single phase commonly appears below room temperature, which hinders their practical applications. Alternatively, ME composites enjoy several orders of ME magnitude larger than that in the single phases resulting from the effective stress-mediated ME coupling between the piezoelectric and piezomagnetic phases at room temperature [1], drawing an increasing interest due to their physical basis and potential applications in magnetic field sensors, transducers, microwave devices and energy harvester [4–8].

The two phases in the ME composites must have large magnetostrictive and piezoelectric effects, respectively. High ME effect has been found in the composites of Pb(Zr,Ti)O_3 (PZT) [5,6,8–10] or BaTiO_3 [11–14] as a piezoelectric part, and Terfenol-D [6,9,10], Ni [5], or ferrites [12–15] as a magnetostrictive part. The composites are obtained by combing the individual ferroic phase through different connections, including particulate composite ceramics [11,12,14], laminate composite [5–7,10,13], column structure [15], and ring-type structure [9,16]. It is more flexible to tune ME parameters in the composites than that in the single phases by

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http://dx.doi.org/10.1016/j.compstruct.2016.08.009

selecting component materials, changing component ratio and improving composite structures. The particulate composites are limited in performance by atomic diffusion, mechanical defects and leakage currents, while the laminate composites can overcome these problems. Then, many experimental and theoretical researches have been performed to optimize the ME output in the laminate composites for their relative higher ME coupling. The high DME coefficients have been achieved in the laminate structures [8,17] based on the extensive researches. The high CME efficient was obtained in a multi-push-pull mode laminates [18], a composite of Rosen-type piezoelectric transformer and a Terfenol-D plate [19], and a composite of Terfenol-D and multilayer vibrator [6].

Theoretically, many methods have been developed to research the ME properties of the laminate composites, including average-field method [20], Green's function approach [21] and Landau-Ginzburg-Devonshire thermodynamic theory [22,23]. Equivalent circuit is an effective method for both the electromechanical coupling [24] and magnetomechanical coupling [25,26]. Dong et al. introduced it to model the ME behavior of laminate [27] and finished some basic works [28–30]. Recently, frequency, nonlinear effect [31–36] and thermal expansion [37,38] were considered in the models. However, these methods focus on the DME characteristics while the CME theory is scarce in the literature [39,40], in which the DME and CME coefficients were obtained respectively. This is incomprehensive to understand the DME and CME correlations deeply. The Maxwell equations show the mutual relationship between $\boldsymbol{E}$ and $\boldsymbol{H}$. DME and CME are two opposite phenomena in the ME materials. A theoretical method should simultaneously describe the DME and CME behaviors, i.e., the relationships of $**P \sim H$ and $B \sim E$. In fact, ferromagnetic and ferroelectric parts exhibit symmetrical characteristics during their phase transitions [41]. DME and CME share the same unit s/m while expressed as $\alpha_{\mathrm{DME}} = (\partial P/\partial H)_E$ and $\alpha_{\mathrm{CME}} = \mu_0(\partial M/\partial E)_H$ [10,42], by which the magnitudes of DME and CME can be compared conveniently. On the other hand, the magnetic part was simply coupled by a coefficient in the ME equivalent circuit [27–30,32–35,38], which can not reveal the ME coupling deeply. Another issue is the unclear reason about the two very close but significantly different resonance peak positions of DME and CME, which were observed in some experimental results [40,43–46]. These suggest the relationship between DME and CME must be studied further.

In this article, we achieve symmetrical magnetic-mechanical-electric equations about electric and magnetic parts based on the piezoelectric and piezomagnetic constitutive equations [47,48] in a typical long-type laminate composite. Then, we build a symmetrical equivalent circuit, from which both the DME and CME characteristics are researched with thickness ratio and frequency. In addition, frequency multiplying behaviors are discussed. Finally, we measured the DME and CME characteristics in a bilayered composite of PZT and Terfenol-D, confirming the symmetrical relationship between the DME and CME effects.

2. Experimental

2.1. Sample preparation

The bilayered ME composites were prepared for comparison with the theoretical model. The PZT ceramic and Terfenol-D (Tb_x-$Dy_{1-x}Fe_2$, $0.68 \leqslant x \leqslant 0.73$) alloy were bonded together with M610 glue at 150 °C under a pressure of 5 MPa for 30 min to obtain good mechanical coupling and strong bonding. The PZT layer has a dimension of $30 \times 4.45 \times 4\ \mathrm{mm}^3$ and Terfenol-D $30 \times 4.45 \times 2.2\ \mathrm{mm}^3$ as shown in Fig. 1. The PZT component was polarized along its thickness direction (z direction) in silicon oil under a poling field of 2 kV/mm. $d_{33,\mathrm{p}}$ of the PZT ceramic chip is about 500 pC/N. The thickness of Terfenol-D layer was kept 2.2 mm and PZT was polished to 3.6, 3.2, 2.8, 2.4, 2.0, 1.6 and 1.2 mm in thickness for comparison.

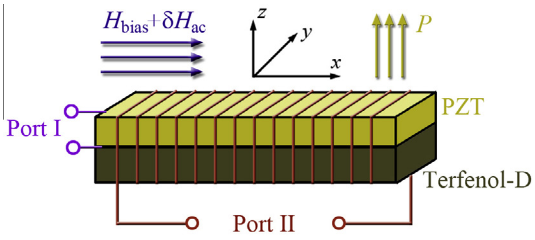

Fig. 1. Schematic diagram of the laminate composite for the model and the experimental measurements. The piezoelectric multilayer is polarized along the z direction and the magnetic field was applied along the x direction.

2.2. Magnetoelectric measurements

Fig. 1 presents the measurement and model of the laminate PZT/Terfenol-D composite. A Cu wire about seventy turns was wound on the composite. The DME measurement was performed by using a dynamic method [6,43,45]. The sample was applied with a small ac magnetic sine signal δH_{ac} superimposed on a dc magnetic field H_{bias} (300 Oe) along the longitudinal direction (x direction). H_{bias} was generated with an electromagnet and δH_{ac} was generated by ac electric current in the Cu wire driven by a power amplifier on the port II, whose sine signal was from a signal generator. The inspired DME charge δQ on the port I and the phase between charge δQ and δH_{ac} were collected by a charge amplifier connected to an oscilloscope. Then the DME effect is characterized by a DME coefficient α_{DME} ($=\delta P/\delta H = \delta Q/S\delta H_{ac}$) with unit s/m, where S is the sample surface area. Another common DME coefficient is α_{ME} ($=\delta E/\delta H$) with unit V/(cm·Oe). Their relation is $\alpha_{\mathrm{DME}} = \varepsilon_r\varepsilon_0\alpha_{\mathrm{ME}}$ and the unit conversion is $1\ \mathrm{s/m} = 2.37 \times 10^7\ \mathrm{V/(cm.Oe)}$, where $\varepsilon_r = 3800$ is used. Then, the electrode at the PZT surface was divided to small uniform electrodes to research the DME contributions from different positions.

The CME coefficient was measured with an induced method. The laminate sample was placed in a dc bias magnetic field of 300 Oe. The PZT layer was applied with a sine electric field δE via the port I along the thickness direction. The induced magnetization δM in Terfenol-D was detected by the Cu coil via the port II. The search coil was connected to an amplifier, then an oscillograph. The phase between the induced magnetization and the input electric field was also recorded. The CME coefficient was obtained by the magnetic induction response to an applied ac electric field, i.e., $\alpha_{\mathrm{CME}} = \mu\delta M/\delta E$ [19,49,50]. Then, another little coil of 10-turn Cu wire with 1.5 mm in length was wrapped around the composite to collect the CME signal at different positions of Terfenol-D [6,19].

3. Theoretical analysis model

Equivalent circuit approaches to both the electromechanical coupling [24] and magnetomechanical coupling [25,26], and it is also an effective method to understand the ME interaction [27], which we develop to discuss the DME and CME characteristics. We consider a rectangular bilayered composite as shown in Fig. 1. In fact, our mode is suitable for the multilayer composite. There are two silver electrodes, which are covered on the piezoelectric surfaces. The two parts are coupled together by stress-strain. The piezoelectric and magnetostrictive layers have the same areas but different thicknesses. The z axis originates the thickness direction and x axis along the length direction. Following the pre-