Journal of Alloys and Compounds 509 (2011) 508–511

Contents lists available at ScienceDirect

Journal of Alloys and Compounds

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

Magnetoelectric properties of Fe–Ga/$BaTiO_3$ laminate composites

Lei Wang [a,*], Zhaofu Du [a], Chongfei Fan [b], Lihong Xu [a], Hongping Zhang [a], Dongliang Zhao [a]

[a] Functional Materials Research Institute, Central Iron and Steel Research Institute, No. 76 Xueyuan South Road, Haidian, Beijing 100081, China
[b] Patent Examination Cooperation Center, State Intellectual Property Office of the People's Republic of China, Beijing 100190, China

ARTICLE INFO

Article history:
Received 22 April 2010
Received in revised form 9 September 2010
Accepted 18 September 2010
Available online 24 September 2010

Keywords:
Magnetoelectric effect
Magnetostrictive alloy
Piezoelectricity
Composites

ABSTRACT

Magnetostrictive and piezoelectric laminate composites of Fe–Ga and $BaTiO_3$ have been studied. The magnetoelectric (ME) coefficients have been characterized for the transversely magnetized and transversely polarized transverse–transverse (TT) mode. At lower frequencies, the ME voltage coefficient of the laminate was 12.5 mV/Oe. Near the natural resonant frequency (~95 kHz) of the laminate, the ME voltage coefficient was found to be dramatically increased to 28.5 mV/Oe. In addition, the induced ME voltages were near linear functions of AC magnetic field.

1. Introduction

The magnetoelectric response is the appearance of an electric polarization P upon applying a magnetic field H (i.e., the direct ME effect, designated as ME_H effect: $P=\alpha H$) and/or the appearance of a magnetization M upon applying an electric field E (i.e., the converse ME effect, or ME_E: $M=\alpha E$) [1]. Materials exhibiting ME effect can be classified into two classes, single-phase materials and composites. The ME effect in a single-phase material is found too weak to be utilized for practical purpose, even at low temperature. The ME effect can also be realized in a composite consisting of ferroelectric and ferromagnetic phases [2]. When a magnetic field is applied to a composite, the magnetic phase changes its shape magnetostrictively. The strain is then passed along to the piezoelectric phase, resulting in an electric polarization. These ME composites have drawn significant interest in recent years due to their potential applications as multifunctional devices such as magneticelectric transducers, actuators, and sensors [3–5]. Some works were devoted to bulk materials, such as $Co_{0.7}Mg_{0.3}Fe_{2-x}Mn_xO_4$–$Sr_{0.5}Ba_{0.5}Nb_2O_6$ [6], $Co_{0.5}Zn_{0.5}Fe_2O_4$–PLZT [7], $NiFe_{1.9}Mn_{0.1}O_4$–$BaZr_{0.08}Ti_{0.92}O_3$ [8], $ZnFe_2O_4$–$BiFeO_3$ [9] and $CoFe_2O_4$–$Ba_{0.8}Sr_{0.2}TiO_3$ [10]. But the ME effect in the bulks has proved to be much weak due to leakage current and low degree of polarization. In recent years, strong ME coupling has been reported in several layered ME composites of magnetostrictive materials, such as $Tb_{0.3}Dy_{0.7}Fe_{1.95}$ [11] or $Tb(Fe_{0.55}Co_{0.45})_{1.5}$ [12] and piezoelectric materials, such as Pb(Zr, Ti)O_3 (PZT) [3] or $Pb(Mg_{1/3}Nb_{2/3}O_3$–$PbTiO_3)$ (PMN-PT) [13] layers.

FeGa alloy (or Galfenol) is a good magnetostrictive material, i.e., high mechanical strength, good ductility, large magnetostriction at low saturation fields, and low cost when compared to rare-earth Fe alloys. A large ME coupling effect has been reported in Fe–Ga/PZT [14] or Fe–Ga/PMN-PT [15,16] laminates. However, PZT or PMN-PT is now facing a big challenge due to the environmental hazard by its toxic lead. Thus alternatively, barium titanate $BaTiO_3$ (BTO) is also a commonly used piezoelectric material and contains nothing injurious to the environment. It provides a choice to synthesize high performance lead-free ME laminate.

In this paper, the ME response of Fe–Ga/BTO laminate has been characterized. The working modes and induced ME voltage of disk-shaped laminate composites were investigated for samples which were magnetized and poled in the transverse direction.

2. Experimental details

The precursor alloy of Fe–20 at.% Ga was arc melted several times under an argon atmosphere to insure homogeneity. Subsequently, the Fe–Ga rod was grown by a Bridgman method in silica crucible. Following crystal growth, the rod was annealed at 860 °C for 8 h and quenched in water. The texture and composition of the rod has been analyzed by X-ray diffraction (XRD) and energy dispersive X-ray spectroscopy (EDS) at room temperature. The magnetostriction was measured by using standard strain gauges. Strain gauges consist of a thin meander shaped films where the resistance changes due to a change of length. This change of resistance can be measured using a bridge circuit.

After then, the Fe–Ga rod was cut into disk-shaped plates of 10 mm in diameter and 1 mm in thickness. Commercial $BaTiO_3$ discs of Ø12 mm × 1.5 mm were used in this study. A conventional disc type three-layer laminate of Fe–Ga/BTO/Fe–Ga was assembled using a conductive epoxy resin, in which the magnetization and polarization were both oriented in its thickness directions, i.e., the (TT) mode.

* Corresponding author. Tel.: +86 010 62183484; fax: +86 010 62183484.
E-mail address: wl_6234@hotmail.com (L. Wang).

0925-8388/$ – see front matter
doi:10.1016/j.jallcom.2010.09.083

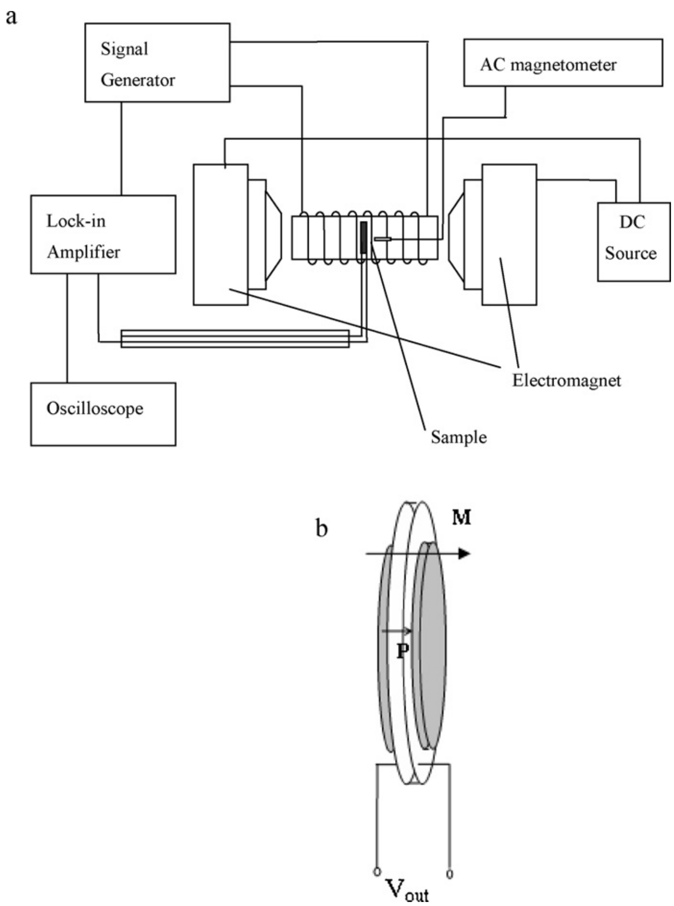

Fig. 1. (a) Schematic diagram of magnetoelectric measurement setup and (b) illustration of the ME laminate composite.

The ME coefficient, α_{ME}, is the most critical indicator for the ME function of a material. The ME coefficient is defined as $\alpha_{ME} = dE/dH = dV/(t\ dH)$, where V is the induced ME voltage, H is the magnitude of the exciting magnetic field, and t is the thickness of the material [1]. There are three methods of measuring ME output [17]. (i) In the static method, ME output (charge or voltage) is measured as a function of increasing magnetic field using an electrometer having high input impedance. Charges which have accumulated at the grain boundaries while poling may move toward the electrode over time giving erroneous measurements [18]. (ii) For more precise measurement, a quasi-static method may be employed. This is done by applying a time varying DC magnetic bias and measuring the polarization response with a high impedance electrometer. The output is measured as a function of time. But this method cannot be employed for a polycrystalline material as the charge build up at the grain boundaries still affects the ME output [19]. (iii) In the dynamic method, dH is produced by superimposing a small AC magnetic field onto a DC magnetic bias in the same direction. Here, dV is also an AC signal. The application of an AC magnetic field prevents charges from migrating toward the electrodes, thereby giving more accurate measurement of the ME response [20].

A dynamic method of measuring ME output has been used to characterize the ME behavior of FeGa/BTO laminate composites. Fig. 1(a) illustrates the measurement system. The sample was put into a small solenoid, and placed between the poles of an electromagnet. The DC magnetic bias was then controlled by a DC source [GP-HP-L/±65A]. The magnetic field strength was pre-calibrated from a Tesla meter [T-6 TYPE, ISAS]. The small solenoid coils produced a small AC magnetic field which was supplied by a signal generator [CA1640-02, CALTEK] and detected by an AC induction magnetometer [CCG-1000]. The electric signal produced by the sample was input to a lock-in-amplifier [LI 5640]. At the same time, the signal generator sent a signal synchronized with the coil excitation signal to the lock-in amplifier as reference. The polarization response was measured along the thickness of the samples, and the magnetic fields (AC and DC) were applied along the thickness as shown in Fig. 1(b).

3. Results and discussion

Firstly, the characterizations of Fe–Ga rod have been performed. XRD results in Fig. 2 confirm that the Fe–Ga rod preserves the bulk A2 structure with three major peaks having hkl values of (1 1 0), (2 0 0), and (2 1 1). The EDS analyses confirm the uniformity of $Fe_{80}Ga_{20}$ phase over the entire length of the rod. The magnetostrictive strain of the Fe–20% Ga rod (in the free condition) was measured as a function of magnetic bias H_{dc}, as shown in Fig. 3. Data are shown for a longitudinal strain where H_{dc} is applied along the length of the Fe–Ga rod. The maximum strain of the Fe–Ga rod was 100 ppm at 1000 Oe applied magnetic field. The insert of Fig. 3 illustrates the induced shape changes of the specimen. Subsequently, when the Fe–Ga rod was cut into disks, the ME saturation field of its laminate composites maybe increased, due to the demagnetisation effects caused by the TT-configuration.

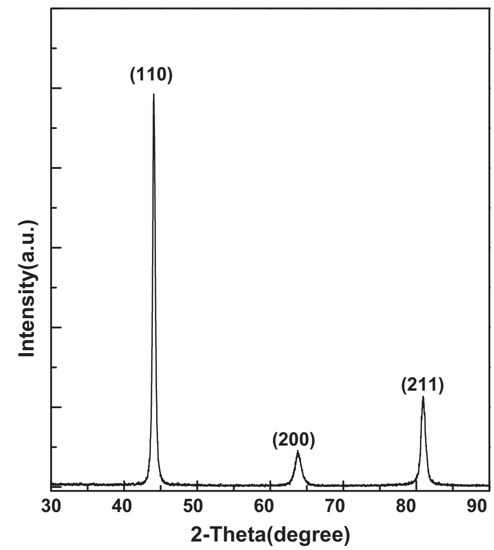

Fig. 2. X-ray diffraction result of Fe–Ga rod.

ME properties of the proposed Fe–Ga/BTO laminate composite were characterized at room temperature using the dynamic method. The ME voltage induced across the laminate composite was measured as a function of frequency, as shown in Fig. 4. The results show that the Fe–Ga/BTO laminate has a much enhanced ME response when operated near the resonance frequency of $f_r = \sim 95$ kHz. The maximum ME voltage at resonance for the T–T mode was ∼28.5 mV (or 190 mV/(cm Oe) for the ME coefficient), which is ∼7× higher than that in the low-frequency range.

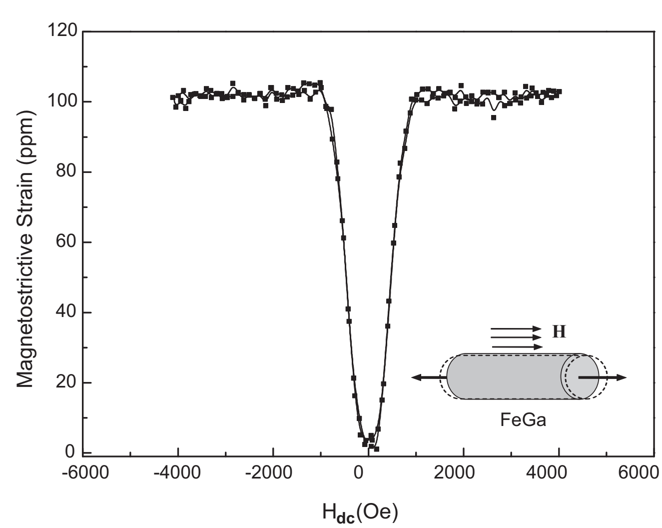

Fig. 3. Magnetostrictive strains of Fe–Ga rod. Inset illustrates the induced shape change of the specimen.

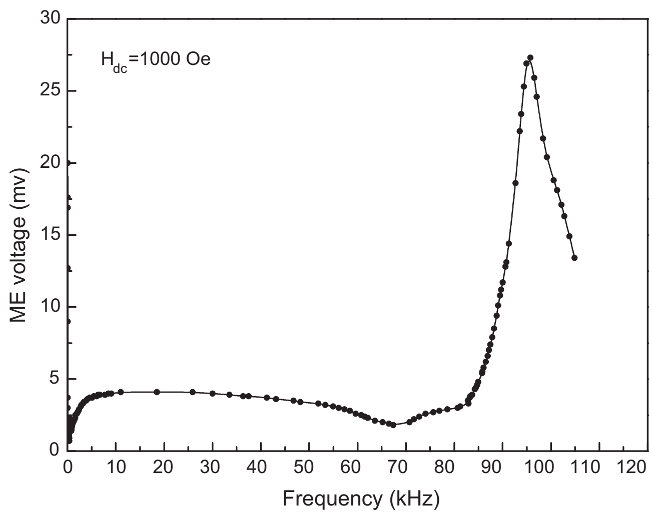

Fig. 4. Induced ME voltage as a function of AC magnetic field frequency for Fe–Ga/BTO laminate. These data were taken under a constant DC magnetic bias (H_{dc} = 1000 Oe) and the amplitude of AC magnetic field H_{ac} = 1 Oe.

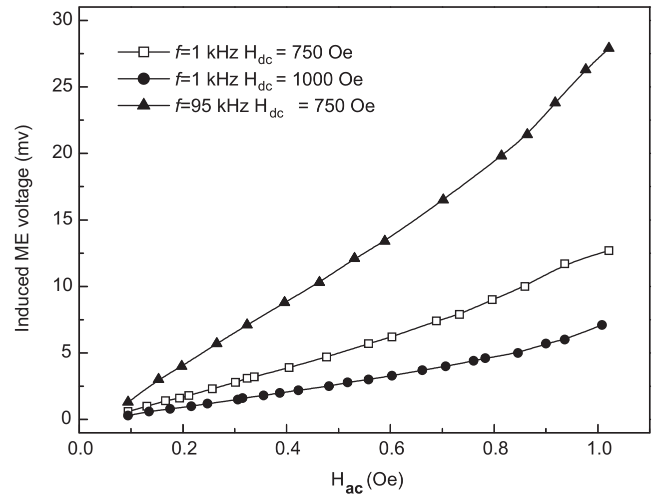

Fig. 6. Induced ME voltages as a function of AC magnetic field H_{ac} for Fe–Ga/BTO laminate, taken at various DC magnetic biases. The operate frequency of H_{ac} is low frequency (1 kHz) or resonance frequency (95 kHz).

Fig. 5 shows the ME voltage coefficient dV_{ME}/dH as a function of DC magnetic bias H_{dc}. These data were taken at frequency of f = 1 kHz and a drive of H_{ac} = 1 Oe. The value of dV_{ME}/dH can be seen to be strongly dependent on H_{dc}. In the DC magnetic bias range $0 < H_{dc} < 750$ Oe, the ME voltage coefficient of the T–T mode of Fe–Ga/BTO laminate increased with increasing H_{dc}, reaching a maximum ME effect of $dV_{ME}/dH \sim 12.5$ mV/Oe at H_{dc} = 750 Oe (or, correspondingly, $dV_{ME}/dH = \sim 84$ mV/(cm Oe)). For $H_{dc} > 750$ Oe, dV_{ME}/dH decreased dramatically with increasing H_{dc}, as Fe–Ga layers of the laminate approached saturation of its magnetostriction. Saturation magnetostriction of the FeGa layers leads to the loss of piezomagnetic effects and ME coupling. In this report, the ME voltage coefficient dV_{ME}/dH was observed to be antisymmetric about H_{dc}. Most previous works [14] only report ME voltage amplitude as a function of H_{dc}.

The dependence of induced ME voltages on H_{ac} for the Fe–Ga/BTO laminate is shown in Fig. 6. It can be seen that the induced ME voltages are near linear functions of H_{ac}. At low frequency (f = 1 kHz), for H_{dc} = 750 Oe, the value of ME voltage was ~12.5 mV under an AC magnetic field of 1.0 Oe; however for H_{dc} = 1000 Oe, ME voltage was decreased to ~7.5 mV. As a comparison, Fig. 6 also illustrates the induced ME voltage of the Fe–Ga/BTO laminate at resonance frequency (f_r = 95 kHz) under a H_{dc} = 750 Oe. A much higher ME voltage of 28–30 mV was obtained at resonance. Dong et al. [21] and Ryu et al. [22] previously studied the Terfenol-D/PZT laminates in TT mode, which exhibited a large ME coupling. Although the low-frequency ME performance of the Fe–Ga/BTO laminate is not as good as that of Terfenol-D/PZT laminate, its resonance ME performance is quite good, due to a high Q-factor.

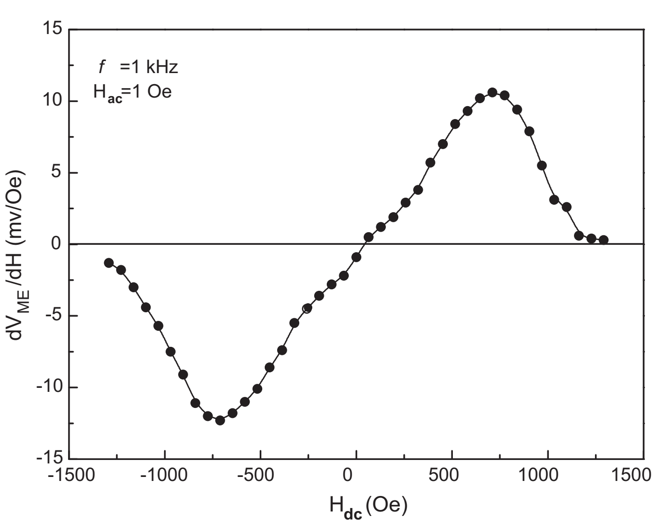

Fig. 5. ME voltage coefficient dV_{ME}/dH as a function of DC magnetic bias H_{dc} for Fe–Ga/BTO laminate.

The compressive stress in the BTO layer can be given in Eq. (1) [22]:

$$\sigma = \frac{2E_f E_b t_f \, \Delta\varepsilon_0}{(1-\nu)(2E_f t_f + E_b t_b)}, \quad (1)$$

where E, t, $\Delta\varepsilon_0$, and ν are the elastic modulus, thickness, the linear strain of the Fe–Ga layer, and Poisson's ratio, respectively. The subscript f or b represents Fe–Ga or BTO, respectively. As shown in this equation, the compressive stress in the BTO layer is increased with increasing the linear strain of the Fe–Ga layer. At H_{dc} = 750 Oe, the slope of Fe–Ga magnetostriction reaches the maximum. Therefore, when an AC magnetic field H_{ac} is superimposed over the DC magnetic bias H_{dc}, the change of magnetostriction is the biggest. With further increasing DC magnetic bias, the slope of Fe–Ga magnetostriction is decreased, thus, the compressive stress in the BTO layer is decreased. The induced ME voltage from the composite is proportional to the stress of BTO layer. Therefore, higher ME voltage may be obtained when the compressive stress in the BTO layer is higher. From Eq. (1), it can be seen that this is achieved when the strain rate of the magnetostrictive layer is maximized, i.e., largest $d\varepsilon/dH$.

4. Conclusions

Laminate of magnetostrictive Fe–Ga and piezoelectric BTO has been characterized. The ME laminate was operated in a transverse magnetized/transverse polarized (TT) mode. An experimental setup has been developed for the measurement of dynamic ME effect, using a time varying DC magnetic bias on which an AC magnetic field is superimposed. The results have shown a maximum value of dV_{ME}/dH = 12.5 mV/Oe at a DC magnetic bias of 750 Oe, when operated in low frequency. A dramatic enhancement in the ME response was observed near the resonance frequency of the laminate. In addition, a linear coupling between the ME voltage and H_{ac} for laminate was observed for various DC magnetic biases.

Since Fe–Ga has high sensitivity in low magnetic field and BTO contains nothing injurious to the environment, it can be a new choice of the laminate for using to make ME devices.

References

[1] C.W. Nan, M.I. Bichurin, S.X. Dong, D. Viehland, G. Srinivassan, J. Appl. Phys. 103 (2008) 031101.
[2] J. Van Suchtelen, Phillips Res. Rep. 27 (1972) 28.
[3] N. Cai, Y. Zhao, X.H. Geng, S.W. Or, J. Alloys Compd. 448 (2008) 89.
[4] D.R. Partil, A.D. Sheikh, C.A. Watve, B.K. Chougule, J. Mater. Sci. 43 (2008) 2708.
[5] D.T. Huong Giang, N.H. Duc, Sens. Actuators A: Phys. 149 (2009) 229.
[6] S.R. Jigajeni, S.V. Kulkarni, Y.D. Kolekar, S.B. Kulkarni, P.B. Joshi, J. Alloys Compd. 492 (2010) 402.
[7] A.S. Fawzi, A.D. Sheikh, V.L. Mathe, J. Alloys Compd. 493 (2010) 601.
[8] R.C. Kambale, P.A. Shaikh, C.H. Bhosale, K.Y. Rajpure, Y.D. Kolekar, J. Alloys Compd. 489 (2010) 310.
[9] P. Uniyal, K.L. Yadav, J. Alloys Compd. 492 (2010) 406.
[10] C.M. Kanamadi, J.S. Kim, H.K. Yang, B.K. Moon, B.C. Choi, J.H. Jeong, J. Alloys Compd. 481 (2009) 781.
[11] Y.J. Wang, X.Y. Zhao, J. Jiao, Q.H. Zhang, W.N. Di, H.S. Luo, C.M. Leung, S.W. Or, J. Alloys Compd. 496 (2010) L4.
[12] N.H. Duc, D.T.H. Giang, J. Alloys Compd. 449 (2008) 214.
[13] Y.J. Wang, X.Y. Zhao, J. Jiao, L.H. Liu, W.N. Di, H.S. Luo, S.W. Or, J. Alloys Compd. 500 (2010) 224.
[14] A.A. Bush, K.E. Kamentsev, V.F. Meshcheryakov, Y.K. Fetisov, D.V. Chashin, L.Y. Fetisov, Tech. Phys. 54 (2009) 1314.
[15] G. Srinivasan, Annu. Rev. Mater. Res. 40 (2010) 153.
[16] D. Seguin, M. Sunder, L. Krishna, A. Tatarenko, P.D. Moran, J. Cryst. Growth 311 (2009) 3235.
[17] M.M. Kumar, A. Srinivas, S.V. Suryanarayana, G.S. Kumar, T. Bhimasankaram, Bull. Mater. Sci. 21 (1998) 251.
[18] A. Hanumaiah, T. Bhimasankaram, S.V. Suryanarayana, G.S. Kumar, Bull. Mater. Sci. 17 (1994) 405.
[19] Z.G. Ye, J.P. Rivera, H. Schmid, M. Heida, K. Kohn, Ferroelectrics 161 (1994) 99.
[20] J. Lu, D. Pan, B. Yang, L. Qiao, Meas. Sci. Technol. 19 (2008) 045702.
[21] S.X. Dong, J.F. Li, D. Viehland, J. Appl. Phys. 95 (2004) 2625.
[22] J. Ryu, A.V. Carazo, K. Uchino, H. Kim, Jpn. J. Appl. Phys. 40 (2001) 4948.