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Investigation of the magnetoelectric effect driven by a single magnetic field in $Tb_{1-x}Dy_xFe_{2-y}-Pb(Zr, Ti)O_3$ bilayers

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Abstract

We have investigated the magnetoelectric (ME) effect induced by a single dc magnetic field in $Tb_{1-x}Dy_xFe_{2-y}-Pb(Zr, Ti)O_3$ bilayers with a simple technique. As a result, a giant ME voltage is observed in the material. We show that the single field driven ME effect, SFDME, is a significant improvement over the two-field driven ME effect in the same material. The SFDME technique appears to have potential to facilitate applications of the ME effect.

1. Introduction

Magnetoelectric (ME) materials are stimulating increasing interest because of their potential application in the fabrication of sensitive devices [1–3]. Many kinds of ME materials have been found and developed in recent years [4–7]. However, only layered composites based on a product effect, that is a cooperated action of several effects, have shown useful characteristics for applications such as in ferrite (or rare earth alloy)-piezoelectrics. So far, most of the ME studies reported have focused on a procedure driven by using two magnetic fields, a bias field *H* and an ac field δH , and with both parallel to each other [8–11], where the ac field is to provide an alternative stress, which results in an alternating surface electric field for the piezoelectric materials. The ME voltage coefficient α_E ($\alpha_E = \delta V/t\delta H$, where *t* is the thickness of the piezoelectric) was used to characterize the intensity of the ME effect in the studies [12–17]. This behaviour can be called a two-field driven ME effect. To simplify the applications of the ME effect, some efforts have been made in recent years, such as the work of Dong *et al* [18]. However, it has not radically changed the use of TFDME.



Figure 1. Illustration of the single field-driven sensor for ME laminate TDF-PZT and the geometrical arrangement for measurements.

Recently, we found that an evident ME effect can also be observed driven by a single dc magnetic field for some layered ME composites, such as $Tb_{1-x}Dy_xFe-Pb(Zr, Ti)O_3$. In this paper we describe how a single dc field can be used to drive the ME effect, SFDME. We find that the results of SFDME are completely different from those of TFDME. Thus the SFDME technique can be expected to simplify the applications of the ME effect.

2. Sample preparation and characterization

 $Tb_{1-x}Dy_xFe_{2-y}$, TDF, is a newly developed material with good magnetostrictive performance and low anisotropy in magnetic crystals. It is also an ideal material for studies of ME composites due to its high Curie temperature and a desirable magnetostriction property. Pb(Zr, Ti)O₃, PZT, is a typical and an often used piezoelectric material. Samples under investigation are stickup bilayers of TDF and PZT. In fact, TDF–PZT laminates have attracted more attention recently, since they show perfect TFDME [18–20].

Commercial TDF pieces with 10 mm diameter and 1 mm thickness are used here. The Curie temperature (653 K) has been measured with a magnetic balance and the magnetostriction λ under a magnetic field *H* parallel to the sample plane has been investigated by the standard strain gauge technique at room temperature. It was found that, with increasing *H*, the magnetostriction λ increased rapidly when $H \leq 750$ Oe, then increased slowly and approached saturation when $H \approx 3000$ Oe.

The PZT powder fabricated by a sol–gel method was pressed into discs of 9 mm diameter and 1 mm thickness, which then were sintered at different temperatures from 1000 to 1200 °C to obtain PZT samples with different pilling density. The sintered PZT disks were poled in an electric field of 10 kV cm⁻¹ perpendicular to the sample plane over 5 h. The TDF–PZT bilayers were made by bonding both with a kind of slow-dry epoxy.



Figure 2. Magnetic field dependence of ME voltage for TDF–PZT bilayers composed of different temperature sintered PZT in the direction with the field parallel, lower panel, and perpendicular, upper panel, to the sample plane, respectively. The dots are measured data and the lines are guides to the eye.

3. Experimental results and discussion

For holding the surface electric charge or stabilizing the surface electric field of the PZT, we connect in parallel a suitable capacitance to the bilayer as a composite unit as shown in figure 1, which is then placed into a magnetic field H parallel or perpendicular to the sample plane, respectively, and insulated from the ambience.

Calling C_0 and C the capacity of the sample and the capacity in parallel with the PZT, respectively, the ME voltage of samples V_0 can be simply obtained by the relation $V_0 = V_{out}[(C_0 + C)/tC]$, where t is the thickness of PZT and V_{out} is the voltage measured. C and C_0 were measured with an ac electrical bridge.

Figure 2 shows the field dependence of the ME voltage in two directions, with H parallel and perpendicular to the sample plane, respectively, for TDF–PZT bilayers at room temperature. Setting the facial electric field of PZT E (figure 1) along the direction 3, which is perpendicular to the sample plane (see figure 1), the ME voltage with H parallel and



Figure 3. Magnetic field dependence of $dV_0/t dH$ for TDF–PZT bilayers composed of different temperature sintered PZT in the direction with the field parallel and perpendicular to the sample plane, respectively. The dots are measured data and the lines are guides to the eye.

perpendicular to the sample plane can be noted as V_{031} (lower panel of the figure) and V_{033} (upper panel of the figure), respectively.

It was found that (1) as field increases, the ME voltage V_{033} increases to saturation at about H = 9000 Oe, and increases with increasing sintering temperature (T_s) of PZT; (2) just the reverse to V_{033} , V_{031} decreases with increasing H and T_s , and approaches saturation at about H = 3500 Oe; (3) the H-dependence of the ME voltage for the samples does not agree with the magnetostriction of TDF. The magnetostriction of TDF tends to saturation when H is larger than 750 Oe. This characteristic is quite different from that observed in TFDME. For TFDME, the H-dependence of α_{E31} is consistent with the magnetostriction of the ferromagnet material being used; (4) the ME voltages V_{031} and V_{033} can reach 330 and 550 V cm⁻¹, respectively. These values are much larger than usually reported for TFDME; (5) the H-dependence of the ME voltage is independent of the polarity of field; (6) the variation of V_{033} is much larger than that of V_{031} for the same sample. This is contrary to the results obtained from TFDME. In the case of TFDME, the ME voltage coefficient for transverse orientation α_{E31} is often 4–5 times larger than that for the longitudinal orientation α_{E33} . In addition, The H-dependence of the ME voltage shows a better linearity before it reaches saturation. This characteristic is expected to be profitable to the potential applications of SFDME.

However, the present results are not inconsistent with those observed in TFDME. To prove this, we differentiate the curves in figure 2 and show the results, that is, the magnetic field dependent $dV_0/t dH$, in figure 3. As can be seen, the group curves located in the first and the third quadrants are the $dV_{033}/t dH$ versus H and the ones in the second and the fourth quadrants are $dV_{033}/t dH$ versus H for TDF–PZT bilayers composed of different temperature sintered PZT.

Through the study of TFDME, we know that $dV_0/t dH$ is just the ME voltage coefficient α_E . Thus $dV_{031}/t dH$ and $dV_{033}/t dH$ should correspond to α_{E31} and α_{E33} , respectively [21, 22]. Considering ΔH as the magnitude of an ac field, we can replace ΔH or $-\Delta H$ by $|\Delta H|$ and obtain a group of curves similar to that observed from TFDME [21, 23]. Furthermore, it is found that the peak value of $dV_{031}/t dH$ is larger than that of $dV_{033}/t dH$,

and the dc field corresponding to the peak value of $dV_{031}/t dH$ is less than that of $dV_{033}/t dH$. These characteristics are just what are obtained from the study of TFDME.

The theory of TFDME in layered composites has been discussed in detail [24, 25], and has been proved to work well. We do not think that it needs more reasons to explain the present results. If we take δH in TFDME theory as zero, we believe that we can obtain a theoretical result similar to that obtained here. More detailed discussions about this will be presented elsewhere.

4. Conclusions

In the configuration used here, with a magnetoelectric effect generated in $Tb_{1-x}Dy_xFe_{2-y}$ -PZT laminated composites by a single dc magnetic field, the magnetoelectric voltage reached 330 and 550 V cm⁻¹ in the transverse and longitudinal planes, respectively. Compared with that observed in a two-field ME effect in the same material, the present results are larger and easier to obtain. This technique can be expected to simplify the applications of the ME effect.

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