Low magnetic field magnetoelectric studies of Ni/PZT/Ni composite sensor

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ABSTRACT

A composite material when placed under the external magnetic/electric fields exhibits voltage/induced magnetization is known as magnetoelectric (ME) composite. Such composite materials should have ferroelectric and ferro/ferri magnetic phases as constituents. The magnetoelectric output is exhibited as a product property. Magnetoelectric composite sensors are being used for variety of applications including resonators, filters, phase shifters, optical isolators, actuators and magnetic field sensors. Metal/ferroelectric/metal magnetoelectric composite sensor using Ni and PZT as constituent phases has been fabricated in 2-2 composite pattern to study its product property. The paper presents magnetoelectric studies of Ni/PZT/Ni composite using low dc magnetic field magnetoelectric set-up. Using this ME set-up ME output of Ni/PZT/Ni composite is studied as a function of dc magnetic field. The results were analyzed to identify the useful magnetic field (dc and ac) range in which Ni/PZT/Ni sensor can be utilized for applications.

Keywords: Composite, magnetoelectric, ferroelectric, sensors

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1 INTRODUCTION

Magnetoelectric effect is defined as exhibition of electrical polarization in presence of external magnetic field or magnetization in presence of external electric field. Many materials including single phase and composites have been reported to exhibit magnetoelectric effect[1, 2]. In case of single phase materials there exists simultaneous ferroelectric and ferro/ferrimagnetic ordering due to which magnetoelectric effect is observed [3]. While, in composites of ferro/piezoelectric and ferro/ferri/piezo magnetic phases, ME output is observed as a product property of two phases which is absent in either of the phases. The deformation of piezomagnetic /ferrite phase causes the polarization of piezoelectric particles of the composites material and on the other hand the electrical polarization of the piezoelectric material causes the change in the magnetization of the piezomagnetic / ferrite phase due to the mechanical coupling of the piezomagnetic and piezoelectric phase.

In recent years, there has been renewed interest in the area of magnetoelectrics due to their wide applications in the field of sensors and actuators [4-6]. However, due to their low field magnetoelectric output single phase materials are not widely used for applications. In turn composites were observed to show high ME outputs useful for applications[7]. In order to further improve the output various researchers attempted fabrication of composites with different constituent phases [8-22]. The authors have previously successfully fabricated Metal/PZT/Metal composites in 2-2 connectivity for the first time, which exhibited improved output [23]. The present paper is aimed at studying magnetoelectric output of Ni/PZT/Ni composite system at low dc bias fields, in order to ascertain the useful magnetic field range of this composite.

2 EXPERIMENTAL

2.1 Materials and methods

In the present study Ni was chosen as the magnetic phase. The desired dimensions of Ni specimens were cut from spec pure (99.9+%) metals obtained from M/s Chempure, USA. The piezoelectric phase chosen for the studies was PZT, which is known to show high piezoelectric constants, to prepare the laminate composite. The PZT disks were cut from the samples obtained from M/s Concord Electroceramics Ltd, New Delhi, India.

2.2 X-ray diffraction

X-ray diffraction studies have been carried out using a PANalytical (X'Pert) diffractometer with Cu-K_α radiation. The X- ray diffractogram of PZT is shown in figure 1. The peaks were tallied with the earlier reports in literature [24]. The characteristic peak is observed at 2θ of 30.72^o and the pattern is indexed based on the data of $Pb(Zr_{0.48},Ti_{0.52})O_3$ (ICDD No-00-033-0784). From the comparison of XRD pattern with the standard one, the lattice parameters were evaluated. These lattice parameters indicate that the structure of PZT is tetragonal with a = 4.063±0.005 Å, c = 4.123±0.004 Å. The volume of the unit cell is found to be 68.074 $Å^3$

2.3 Electrical poling

In general the piezoelectric/ferroelectric ceramics are insulators; and due to the random distribution of the dipoles in different domains, they have zero remanant polarization. However, in poling process the domains get oriented in the direction of field. When an external poling field is higher than the coercive field of a ferroelectric sample, most of the domains will be reoriented. In the present study PZT pellets were coated silver paint on the both large surfaces to ensure good ohmic contact. In electric poling process, the PZT sample was heated to 100^oC in an external electrical field of 30 kV/cm, kept at this temperature for about 30 minutes, and then cooled to room temperature in the presence of the field at the rate of $10^oC/min$. Proper electric poling is necessary to obtain magnetoelectric output in composites.

2.4 Laminate fabrication

PZT pellets were machined to thicknesses of 1mm a diameter of 5 mm. Ni discs were machined to dimensions of Φ 5mm x 1mm. Figure 1 illustrates the structure of the two phase laminate composite of NMF and PZT. This laminate consists of one layer of thickness-polarized PZT disc sandwiched between two identical layers of Ni disc along the thickness direction. To construct the Ni/PZT/Ni laminate composite, a PZT disc was bonded between two Ni discs along the thickness direction using a silver epoxy adhesive at 80 ^{o}C cured for 1 hour and 100 ^{o}C for ½ hour to ensure good mechanical coupling between the discs. To ensure proper contact between two phases external load was applied on the laminates during curing. The electrical wires were connected to the outer surfaces of both the Ni discs to form the output terminals.

2.5 Magnetic poling

In magnetic poling technique, a sufficiently high magnetic field (5 k Oe to 10 k Oe) is applied to the composite sample such that domains with in the individual crystallites are reoriented and more or less aligned in the direction of the magnetic field. In general, samples are magnetically poled in the same direction as that of the electrical poling. The sample under test (Ni/PZT/Ni laminate) was kept between the pole pieces of the electromagnet magnetically poled in a dc field of 6 kOe for 60 minutes. A proper poling strategy would ensure magnetic domains to be oriented in the direction of the field. This is necessary for realizing higher magnetoelectric output in the composites.

2.6 Magnetoelectric measurements

Magnetoelectric measurements were preformed adopting the dynamic method. In the dynamic method the samples were kept between the pole pieces of a dc magnet, which can generate the dc magnetic field up to 5 kOe. The ME output was recorded at fixed ac magnetic fields of 2-64 Oe (f =1.008 kHz), superimposed on a varying dc field (in the range of 0.7-5 kOe).

3 RESULTS AND DISCUSSION

Figure 3 shows the ME output vs. dc magnetic field for Ni/PZT/Ni sample. It is observed that with increase in dc bias field, the ME output is found to decrease. For the present Ni/PZT/Ni sample, the output recorded was 173.5mV/cm at 0.7 kOe dc bias field, and fixed 64 Oe ac field. The ME output values are found to be 27.5 mV/cm at fixed 3 Oe, and 160 mV/cm at fixed 54 Oe ac fields.

The maximum ME output obtained is 173.5mV/cm at a fixed 64 Oe ac magnetic field (1.008 kHz frequency) and 0.7 kOe dc magnetic field. The value of ME output is higher than that reported in literature for 40% $Ni_{0.97}Co_{0.03}Mn_{0.01}Fe_{1.9}O_4$ + 60%$BaTi_{1.02}O_{3.04}$ [25].

The variation ME output as a function of ac bias field for Ni/PZT/Ni laminate composite is shown in figure 4.The ME output is found to increase linearly with ac magnetic field. The ME output of Ni/PZT/Ni samples is almost constant with the dc magnetic field in the range 1 kOe to 4.5 kOe. The slope of the plot (magnetoelectric coefficient $\Delta E/\Delta H$) is found to be 2.48mV/cm-Oe for the present sample .

Magnetoelectric effect in Metal PZT composites is found to be higher than particulate composites. Moreover, the fabrication of composites in 2-2 laminates is found to be effective for realizing higher magnetoelectric output. The ME output in these composites is attributed to effective transfer of stress from magnetostrictive phase to piezoelectric phase due to face to face contact of two phases (due to higher contact surface). The effective mechanical coupling resulted in higher charge transfer from piezoelectric phase to metal for short duration of time. Due to this metal being highly conducting phase, ME output is found to be constant in the dc bias field, where as in the ac bias field higher charge accumulation was observed. The combination is observed to be metal/insulator/metal capacitor configuration with charge being accumulated due to bias ac field.

4 DESIGN AND FABRICATION OF ME SET-UP FOR LOW FIELDS

Figure 5 shows a schematic and figure 6 is the photograph of the testing apparatus. The sample is magnetically loaded perpendicular to the solenoid having dimensions of 7.56 cm inner diameter, 15.28 cm outer diameter a length of 30 cm and 3600 turns. The magnetic field strength produced by dc current through the coil at the center is measured using an axial hall probe of Digital Gauss Meter (Model No 202).The solenoid produces a dc magnetic field of 0-500 Oe in positive direction and -500 Oe in negative field. The ac Helmholtz coils each having 32 turns are mounted on bakelite cylinder. This cylinder is inserted inside the solenoid along its axis such that the Helmholtz coils are placed at its centre. The sample is loaded at the centre of Helmholtz coils normal to the field. These coils are connected to the Lock-in- Amplifier (Stanford SR-530) through the power amplifier (TZA-4000) as shown in block diagram. Magnetic testing was performed under various dc magnetic biases with a superposed ac magnetic field of 1.008 kHz frequency.

The magnetic field strength H, produced by ac Helmholtz coil pair is calculated by the formula $H \approx 0.899NI/R$, where H is the magnetic field in Oersteds, N is the total number of the turns per coil, I is the current through the Helmholtz coil and R is the radius in cm. The current through the Helmholtz coils is measured using Keithley 199 DMM.

Using the above experimental setup, ME measurements were carried out as function of ac and dc magnetic fields at room temperature.

Figure 7 shows the ME output data for increasing and decreasing dc bias magnetic field for the sample Ni/PZT/Ni [1:1:1] tri layer composite. As the dc bias field increases from 0 Oe to +500 Oe in positive direction, + 500 to decreases to – 500 Oe through 0 Oe, then increased -500 Oe to 0Oe. A fixed 1.5 Oe ac magnetic field (f = 1.008 kHz) was superimposed on the dc bias field. As the dc magnetic field increases from zero, the ME output is found to increase and reaches a maximum at 60 Oe, with further increase in dc magnetic field, ME output shows a decrease. The ME output recorded was 39.02 mV/cm at 60 Oe dc bias field and 1.5 Oe (f = 1.008 kHz) ac magnetic field. This reveals the ME output dependence on the low dc and ac magnetic fields. The Ni/PZT/Ni composite showed no hysteresis, indicating low loss behaviour of ME composite in the bias magnetic field. This result demonstrates the utility of this composite as switching sensor in low dc bias fields.

5 CONCLUSIONS

Finally, it is concluded that the ME output can be realized in the laminates of Metal/FE composites, and both the participating phases need not be oxides. Thus, a systematic search of combination of various metallic materials, intermetallics and met glasses with piezoelectrics may result in a better combination of a composite, which would show higher ME output for applications.

The 2-2 laminate composite of Ni/PZT/Ni was fabricated by choosing Ni as magnetic phase and PZT as ferroelectric phase. The study yields an ME output of 173 mV/cm for Ni/PZT/Ni composite at 0.7 kOe dc bias field and fixed 64 Oe ac field (1.008 kHz). Low dc magnetic field magnetoelectric set-up was fabricated and successfully utilized to study ME output of Ni/PZT/Ni system. By using the low field set-up the ME output recorded was 39.02 mV/cm for Ni/PZT/Ni[1:1:1] composite at 60 Oe dc bias field and 1.5 Oe (f = 1.008 kHz) ac magnetic field.

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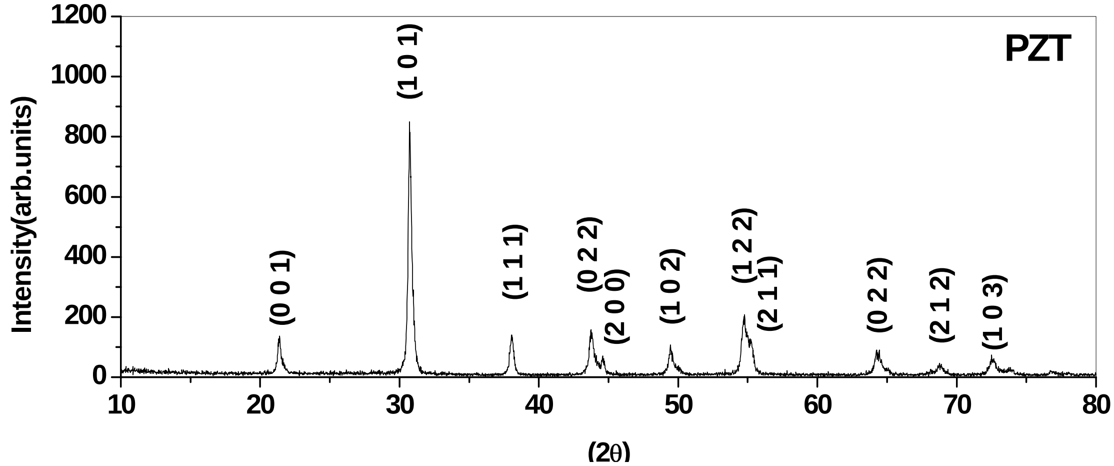

Fig.1. X-ray diffraction pattern of PZT C5 [PZT]

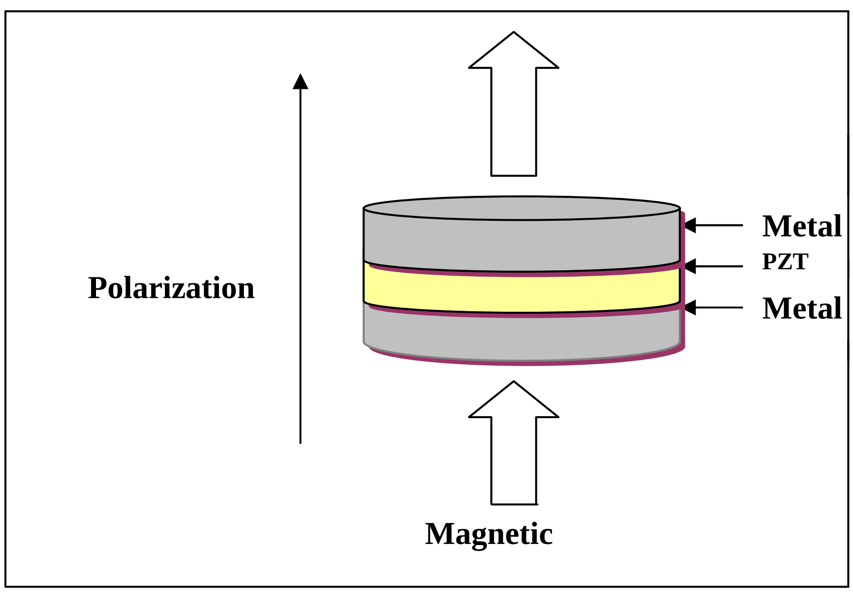

Fig.2. The schematic structure of a laminate composite

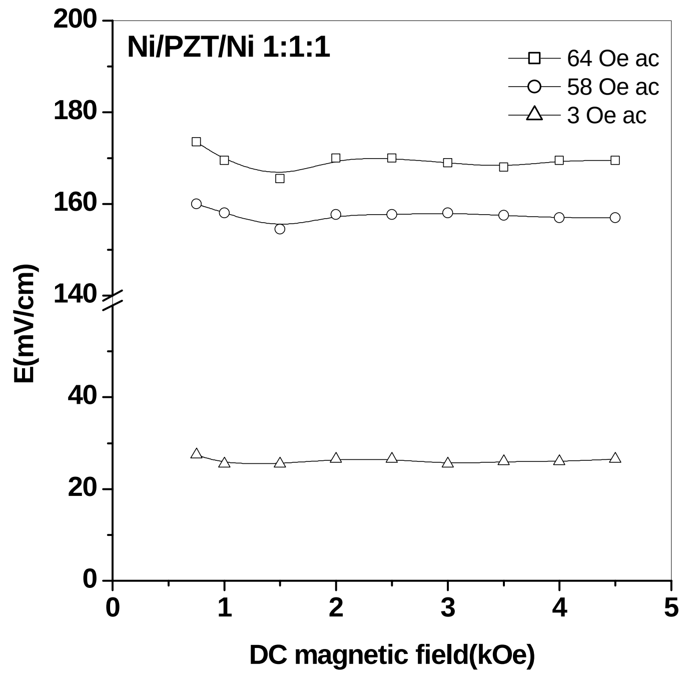

Fig.3. Variation of magnetoelectric output as a function of dc magnetic field for Ni/PZT/Ni 1:1:1 laminates

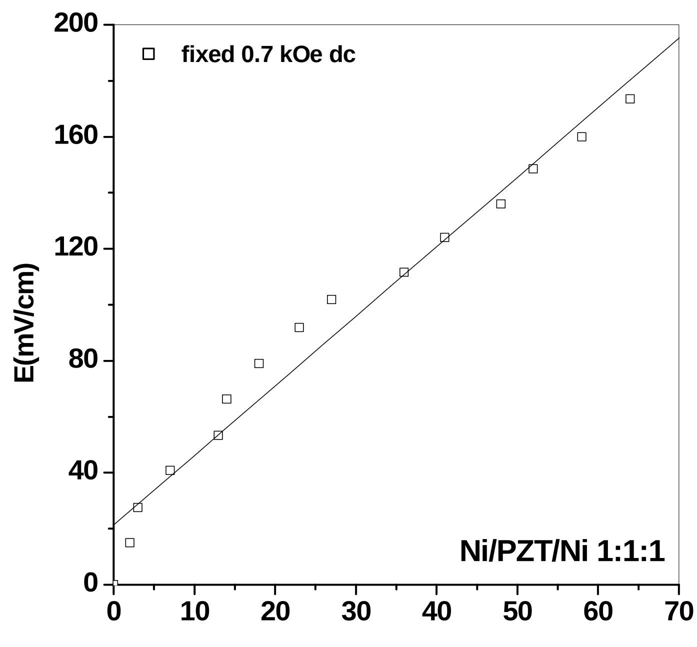

Fig.4. Variation of magnetoelectric output as a function of ac magnetic field for Ni/PZT/Ni 1:1:1 laminates

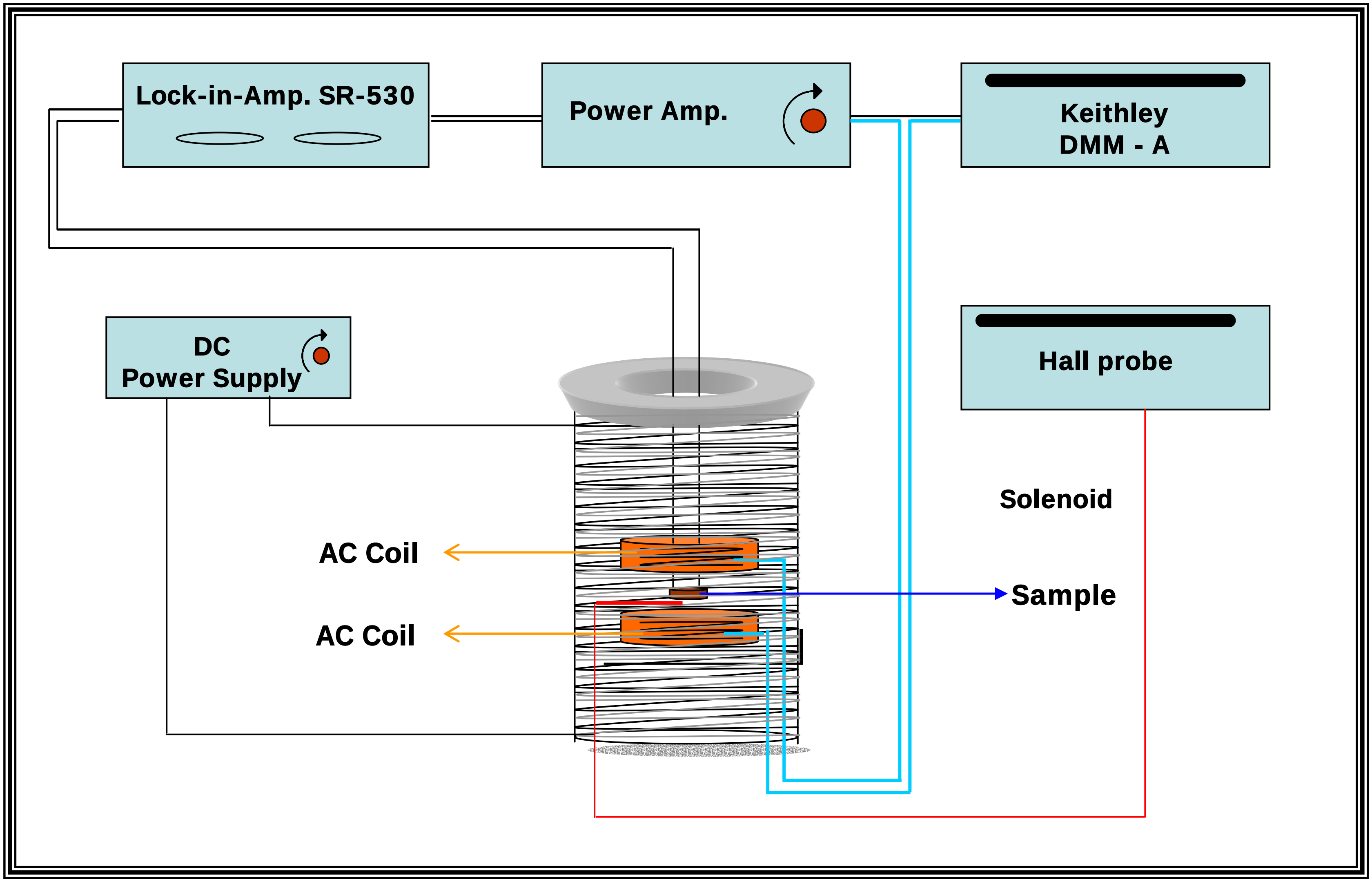

Fig.5. The block diagram of ME set-up working under low fields

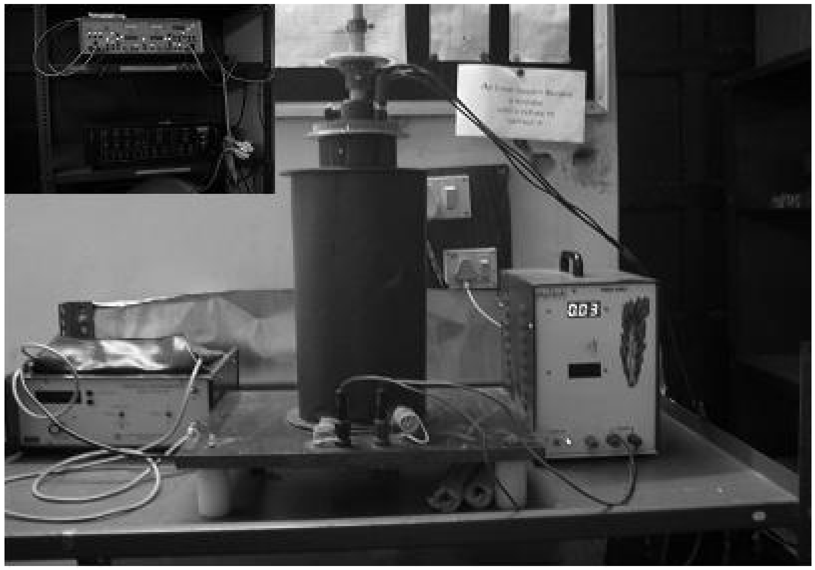

Fig.6. Photograph of low field ME set-up

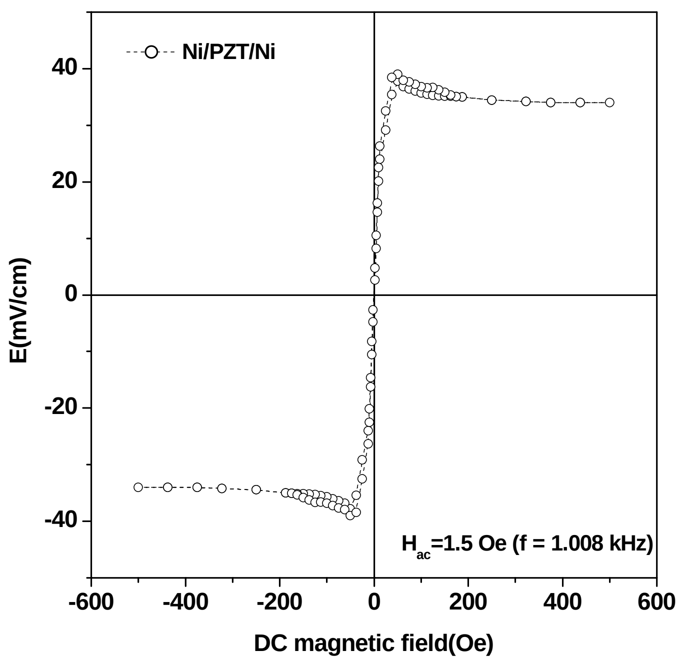

Fig.7. Variation of ME output as a function of dc magnetic field for Ni/PZT/Ni 1:1:1 laminates