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# An easy way to measure accurately the direct magnetoelectric voltage coefficient of thin film devices

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ABSTRACT

$Tb_xDy_{1-x}Fe_2/Pt/Pb(Zr_x, Ti_{1-x})O_3$ thin films were grown on $Pt/TiO_2/SiO_2/Si$ substrate by multi-target sputtering. The magnetoelectric voltage coefficient $\alpha_{ME}^{H}$ was determined at room temperature using a lock-in amplifier. By adding, in series in the circuit, a capacitor of the same value as that of the device under test, we were able to demonstrate that the magnetoelectric device behaves as a voltage source. Furthermore, a simple way to subtract the stray voltage arising from the flow of eddy currents in the measurement set-up, is proposed. This allows the easy and accurate determination of the true magnetoelectric voltage coefficient. A large $\alpha_{ME}^{H}$ of 8.3 V/cm. Oe was thus obtained for a Terfenol-D/Pt/PZT thin film device, without DC magnetic field nor mechanical resonance.



## 1. Introduction

The search of new magnetoelectric (ME) device has been the subject of a considerable number of experimental works, as attested by the abundant literature. Beside the use of single phase multiferroic materials like $BiFeO_3$, design of short-term operational multiferroic devices stimulated investigation on multiferroic composites prepared from magnetostrictive and piezoelectric materials. In such devices, ME coupling is achieved through transmission of mechanical strain between both materials. Thus, giant room temperature ME responses were reported with the $Tb_xDy_{1-x}Fe_2/Pb(Zr_x, Ti_{1-x})O_3$ composite [1–3].

To achieve integration of the ME composite in electronic devices, thin films must be grown on metallized Si substrate. Several attempts were recently reported for this purpose, especially involving $CoFe_2O_4$ [4–7] and $Tb_xDy_{1-x}Fe_2$ (Terfenol-D) [8–14] for the magnetic layer, and $Pb(Zr_xTi_{1-x})O_3$ (PZT) for the ferroelectric one. The usual experimental set-up employed to measure the ME voltage coefficient ($\alpha_{ME}^{H}$) consists in putting the sample in a Helmholtz coil producing a small ac magnetic field, possibly set into a second DC electro-magnet, and recording the voltage induced across the sample with a lock-in amplifier synchronized at the frequency of the generator supplying the magnetic field. However, the determination of $\alpha_{ME}^{H}$ is difficult in the case of thin films because of possible stray voltages that may add to the ME signal. Thus we showed recently that eddy currents induced in the connecting wires by the applied ac magnetic field, led to high additional voltage across the device [15]: at low frequencies (10 Hz to 1 kHz), this voltage is much larger than the classical electromotive voltage $v_{EMF}(\omega, t)$ induced by the small loops delimited by the connecting wires. By measuring the eddy currents in a second sample containing only the PZT layer, we were able to subtract this unwanted voltage to the total voltage recorded on the Terfenol-D/PZT device, hence to get the true $\alpha_{ME}^{H}$.

The purpose of this paper is to propose a new and very simple method to determine accurately the ME coefficient by the use of an original electrical circuit during the measurement that enables to subtract the stray voltages arising from magnetic induction in the connecting wires.

## 2. Materials and methods

Magnetron sputtering was used to grow each layer of the Pt/Terfenol-D/Pt/PZT/Pt structure on the silicon substrate. First, $Pb(Zr_{0.56}Ti_{0.44})O_3$ layer (400 nm in thickness) was deposited at 150 °C on $Pt(120\ nm)/TiO_2(5\ nm)/SiO_2(100\ nm)/Si(400\ \mu m)$ substrate by multitarget sputtering in mixed argon (95%) – oxygen (5%) gas. The PZT layer was then annealed in conventional furnace at 650 °C during 30 min in air for crystallization. Heating and cooling ramps were 2 °C/min for all annealing treatments employed in this study. Then, a thin Pt layer (100 nm) was sputtered in pure argon at room temperature, followed by annealing in air at 625 °C for 30 min,

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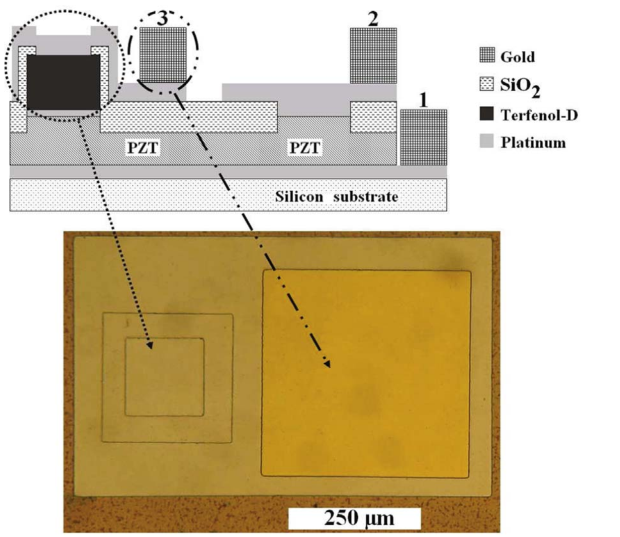


**Fig. 1.** Top: Cross sectional schematic drawing of the device as prepared by UV photolithography. Bottom: front view of a Terfenol-D/Pt/PZT/Pt device, the larger square corresponds to the gold pad for wire bonding, and the smaller square is the active device. Electrical measurements were recorded between pad 1 and pad 3 for the ME device, and between pad 1 and pad 2 for the Pt/PZT/Pt stack.

before deposition of the Terfenol-D layer. The intermediate Pt layer was employed in order to reduce oxygen diffusion from the PZT to the Terfenol-D layer. This Pt layer is also expected to limit diffusion of iron atoms from Terfenol-D to PZT. In this study, Terfenol-D was annealed at 350 °C in air instead of 300 °C in our previous works [14,15].

Discrete PZT/Pt/Terfenol-DME devices and PZT samples were then patterned by etching and lift-off. Argon ion milling was used to etch the sample on which $250 \times 250\ \mu m^2$ pads were protected by a photoresist (1.5 µm in thickness). A sketch of the device showing the different layers is shown in Fig. 1.

The active part of the device consists in a stacking of platinum bottom electrode (120 nm), PZT layer (400 nm), platinum intermediate layer (100 nm) and Terfenol-D upper layer (400 nm). The top contact pad has been shifted away from the active area in view of connecting very small devices (down to micron size), and also to prevent device degradation during bonding of electrical wires. For this purpose, a $SiO_2$ passive layer (250 nm) ensures electrical insulation between the gold pad (400 nm) and the etched areas. The $SiO_2$ layer was deposited by sputtering in mixed argon (95%) – oxygen (5%) gas. We checked that the resistivity of this insulating layer was very high ($10^{13}\ \Omega$ cm for an applied field of 100 kV/cm) and that its relative permittivity was 4 as expected. A thin platinum layer (120 nm) allows to connect the gold pad to the device area and also to ensure sufficient adhesion of gold on the $SiO_2$ layer. Due to the low permittivity (and very high resistivity) of the $SiO_2$ layer, the measured capacitance of the PZT or Terfenol-D/PZT stacks (typically around 1–1.5 nF) were only slightly influenced by the capacitance of the contacts lying above the $SiO_2$ (typically 0.07 nF).

The important point is that the sample (surface of typically 1 $cm^2$) was divided into two parts: one half containing ME thin film devices (several individual squares of $250 \times 250\ \mu m^2$), and the other half containing PZT thin film capacitors only (squares of also $250 \times 250\ \mu m^2$ without the Terfenol-D layer). This enables to compare, for a given sample, a ME device and a reference PZT capacitor in which no ME response is expected. The intermediate Pt layer, grown between the PZT and the Terfenol-D layers in order to limit the atomic diffusions, was 100 nm in thickness. The thickness and the composition of the films was checked for, using an FEI XL30 scanning electron microscope (SEM) equipped with a field emission gun and OXFORD energy dispersive spectroscopy (EDS) elemental analysis system.

The direct ME coefficient $\alpha_{ME}^{H}$ of the device was measured using a lock-in amplifier after setting the sample in the core of a solenoid producing an ac magnetic field of 2.4 Oe driven at a frequency of 800 Hz. The application of a DC magnetic field superimposed to the AC field showed that the ME coefficient was almost constant in the range 0–1000 Oe [12], as also observed by Wan et al. [6]. Therefore all the measurements presented in this work were performed without DC field.

## 3. Calculations

In our previous paper [15], we considered the ME device as a current source $i_{ME}$. The corresponding equivalent electric circuit is shown in Fig. 2(a).

By assuming the input impedance of the lock-in is high enough ($Z_{LI}(\omega)$ of 100 MΩ typically), the measured voltage $v_{Li1}(\omega,t)$ is:

$$v_{Li1}(\omega, t) = v_{EMF}(\omega, t) + Z_p(\omega) \times i_{eddy}(\omega, t) + Z_p(\omega) \times i_{ME}(\omega, t)) \quad (1)$$

In this equation, $v_{EMF}(\omega, t)$ corresponds to the electromotive force, $Z_P$ is the impedance of the sample given by $R_p/(1+j\omega C_p R_p)$, where $C_p$ and $R_p$ are the capacitance and the leakage resistance of the sample respectively. $i_{eddy}$ and $i_{ME}$ are the eddy current and the ME current sources, respectively. Note that with the typical values of $R_p$ (10 MΩ) and $C_p$ (1 nF), $Z_P(\omega)$ may be simplified in $1/jC_P\omega$ in the range of frequencies (10–1000 Hz) investigated in this study (then the influence of $R_p$ is negligible).

At low frequencies $v_{EMF}(\omega,t)$ is very small (typically around 10 µV at 800 Hz), thus negligible with respect to the other terms of (1) so that:

$$v_{Li1}(\omega, t) = Z_p(\omega) \times (i_{eddy}(\omega, t) + i_{ME}(\omega, t)) \quad (2)$$

The question we want to raise in this paper is whether the ME device behaves as a current or a voltage source. This point seems indeed important for the further design and optimization of the electric circuits to be connected to the device in some applications

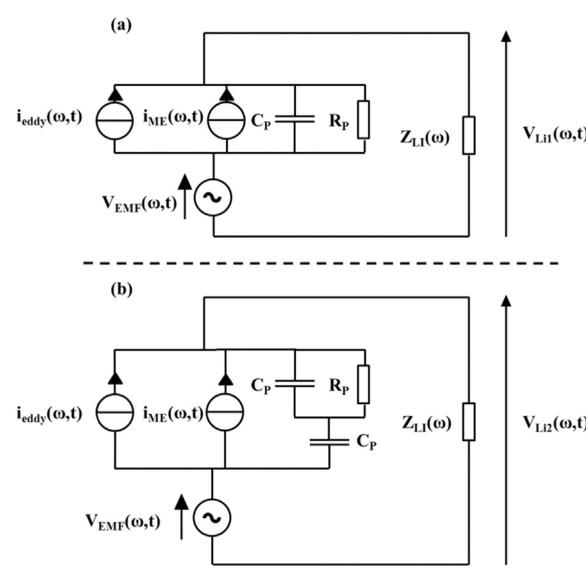


**Fig. 2.** (a) Equivalent circuit assembly where the ME device is considered as a current source and the lock-in amplifier (input impedance $Z_{LI}(\omega)$) is used in the voltmeter mode. $i_{eddy}$ is the eddy currents source. $C_p$ and $R_p$ are the capacitance and the leakage resistance of the ME device, respectively. The voltage source $v_{EMF}(\omega, t)$ represents the total electromotive force induced by small loops delimited by the connecting wires. (b) *Same equivalent circuit assembly including a standard capacitor connected in series.*