# Measurement of the magnetoelectric coefficient using a scanning evanescent microwave microscope

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(Received 8 April 2005; accepted 18 August 2005; published online 6 October 2005)

A quantitative magnetoelectric coefficient measurement method was developed based on scanning evanescent microwave microscopy. This unique technique does not require electrodes and has advantages that it has a high spatial resolution and can simultaneously measure other related properties such as the nonlinear dielectric constant. We have demonstrated that this technique can detect the magnetoelectric coefficient of thin film samples as low as 10 mV/cm Oe. © *2005 American Institute of Physics*. [DOI: 10.1063/1.2093925]

Effective conversion between magnetic and electric signals is extremely desirable for a large number of device applications. Multiferroics, which are simultaneously ferroelectric and ferromagnetic, can exhibit coupling effects between ferromagnetic and ferroelectric properties.[1–5] For instance, spontaneous electric polarization can be modulated by an applied magnetic field, and the spontaneous magnetization can be changed by an applied electric field. In composite multiferroics, the magnitude of such effects is strongly dependent on the efficacy of the elastic coupling between magnetostrictive and piezoelectric components, and in some intrinsic multiferroic materials systems, the exact nature of the magnetoelectric (ME) coupling is not well understood. In order to elucidate and quantify the nature of the ME coupling, it is important to measure the ME coefficient, $\alpha_{ME}$, which can be defined as the ratio of the induced electric field $E$ caused by the applied magnetic field $H$, $\delta E/\delta H$. Common ME measurement techniques are based on the capacitor structures.[6] In such a measurement, an ac magnetic field is applied to the parallel plate capacitor consisting of a magnetoelectric material under a dc magnetic bias, and the coefficient is deduced by monitoring the open voltage or short current across the capacitor with a lock-in amplifier. A significant shortcoming of this technique, especially for thin film samples, is the fact that an interference signal from the magnetic induction due to the finite wire loop could be substantial and comparable to the ME signal. In this letter, we describe a novel technique for measuring the ME effect based on the scanning evanescent microwave microscopy (SEMM).

The schematic diagram of the SEMM (EMP2001, Ariel Technologies, Inc.) is shown in Fig. 1. A SEMM uses near-field electromagnetic interaction between the sample and the sharpened tip mounted to the center conductor of a coaxial resonator for extracting various physical properties of materials on the microscopic scale. The detailed design, operation, and the theory of the SEMM can be found in the literature.[7–9] SEMM has been previously used to measure the complex dielectric constant, nonlinear dielectric constant, conductivity, etc.[10–13] For the present experiment, all the measurements were carried out in the soft contact mode with the contact force between the tip and the sample less than 20 $\mu$N by placing the sample on a tungsten tape cantilever. The microwave frequency was 2.5 GHz.

The principle behind our measurement technique is that the ME coefficient can be extracted from the dielectric constant shift caused by the applied magnetic field. When a magnetic field is applied to a multiferroic, its polarization changes through the ME coupling, but because of the ferroelectricity, its dielectric constant also changes through dielec-

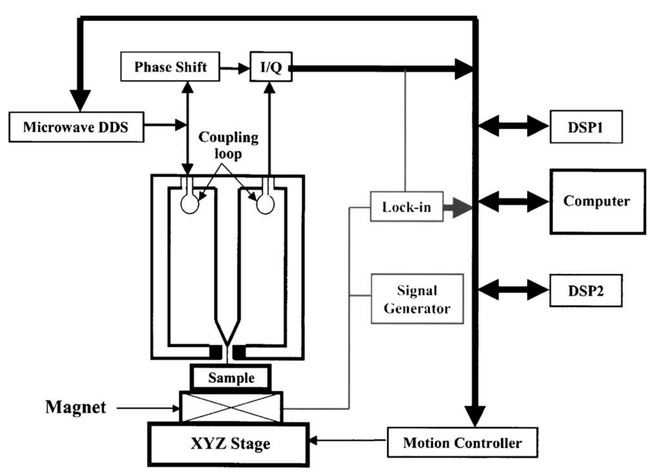


FIG. 1. Modified SEMM system for ME measurement. A signal generator, a lock-in amplifier, and a resonant magnet are integrated into the system.

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tric nonlinearity under the internal electric field induced by the polarization. To measure $\alpha_{ME}=\delta E/\delta H$, we modified the standard SEMM setup by incorporating a signal generator, a resonant magnet and a lock-in amplifier as shown in Fig. 1. The signal detected by the lock-in amplifier upon application of an ac magnetic field can be expressed as

$$v_{ME}=S\frac{\delta f_r}{\delta H}\Delta H=S\frac{\delta f_r}{\delta E}\bullet\frac{\delta E}{\delta H}\Delta H=S\frac{\delta f_r}{\delta E}\bullet\alpha_{ME}\Delta H, \tag{1}$$

where $S$ is the sensitivity of the SEMM on the resonant frequency shift of the microwave probe cavity, $E$ is the internal electric field in the sample induced by the magnetic field $H$ through the ME effect, $\alpha_{ME}$ is the ME coefficient to be measured, and $\Delta H$ is the modulation amplitude of the AC magnetic field. $\delta f_r/\delta E$ is proportional to the dielectric nonlinearity of the material, and it can be obtained through a separate nonlinear dielectric constant measurement as described in Refs. 11 and 12. For this measurement, an electrode on the back of the substrate is typically used for applying an electric field. The nonlinear dielectric signal $v_{\mathrm{nonlinear}}$ can be expressed as

$$v_{\mathrm{nonlinear}}=S\frac{\delta f_r}{\delta E}\bullet\frac{\delta E}{\delta V_n}\Delta V_n, \tag{2}$$

where $E$ is the electric field inside the sample caused by the applied voltage $V_n$ between the back electrode and the tip, and $\Delta V_n$ is the modulation amplitude of the applied voltage. Combining Eqs. (1) and (2), we arrive at

$$\frac{v_{ME}}{v_{\mathrm{nonlinear}}}=\frac{\alpha_{ME}\Delta H}{\dfrac{\delta E}{\delta V_n}\Delta V_n}. \tag{3}$$

Thus, $\alpha_{ME}$ can be obtained as

$$\alpha_{ME}=\frac{\delta E}{\delta V_n}\frac{V_{ME}/\Delta H}{V_{\mathrm{non}}/\Delta V_n}, \tag{4}$$

where

$$\frac{\delta E}{\delta V_n}=\frac{1}{CR_0}\frac{\varepsilon+\varepsilon_0}{2\varepsilon_0}$$

is the ratio of the effective electric field in the sample to the applied voltage calculated using the bulk approximation.[11] Here, $R_0$ is the tip radius, $\varepsilon$ and $\varepsilon_0$ are the permittivities of the sample and air, respectively, and $C$ is an $\varepsilon$ dependent constant which is needed for quantitative microwave microscopy.[11] For thin film samples, the electric field inside the sample produced by the tip is highly nonuniform. As a consequence, the quantitative calculation of nonlinear dielectric constant of film samples relies on a numerical simulation to obtain the constant $C$. An algorithm has been developed for this purpose, and the $C$ value for the present sample configuration was calculated to be $1.67\times10^4$. The details of this algorithm will be published elsewhere.[14]

A common problem in high frequency magnetic measurements is the lack of an appropriate power supply. As the inductance of a coil increases with frequency, a high voltage or a high current amplifier is needed to drive the coil. However, the linearity of such an amplifier is always a nontrivial issue. In our measurement, a resonant magnet technique was used.

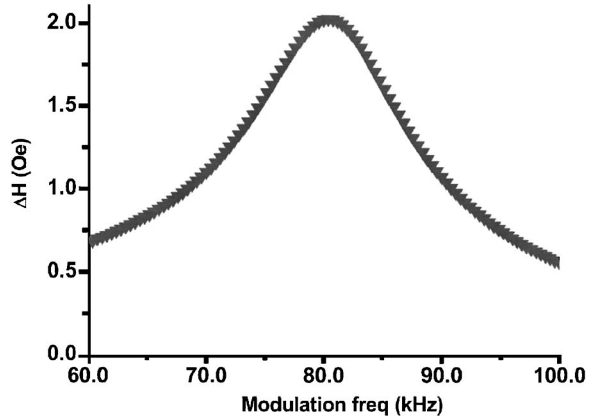


FIG. 2. Frequency dependence of the modulation magnetic field generated by the resonant magnet.

A homemade resonant magnet was used which consisted of a 20 turn coil of $\approx 2\ \mu$H connected in parallel with a 2 $\mu$F capacitor. This magnet has the advantage that it is easy to drive it near the resonant frequency. Generally speaking, the current flowing in the resonant system is $Q$ times that of the power supplied, where $Q$ is the quality factor of the resonant magnet. For the magnet we used, the resonant frequency and $Q$ were measured to be 84.5 kHz and 5, respectively. This magnet has the bandwidth covering the frequency from 60 kHz to 100 kHz. Figure 2 shows the frequency dependence of the magnetic field produced by the coil measured using a small loop probe. The resonant coil can provide a peak magnetic field of 2 Oe at the resonant frequency with the driving voltage and the current of 5 V and 50 mA, respectively. To minimize the signal from the above mentioned inductive effect, all electronics were connected to a floating ground, and the lock-in amplifier was used in the differential mode.

Shown in Fig. 3 is a ME measurement result obtained from a 300 nm thick $(PbTiO_3)_{80}-(CoFe_2O_4)_{20}$ multiferroic nanocomposite film epitaxially grown on a MgO single crystal. Detailed description on the fabrication, microstructural analysis, and multiferroic properties of the film will be published elsewhere.[15] For comparison, a pure $PbTiO_3$ film, a $CoFe_2O_4$ film on MgO substrates and a MgO substrate itself were also measured, and these results are displayed in the

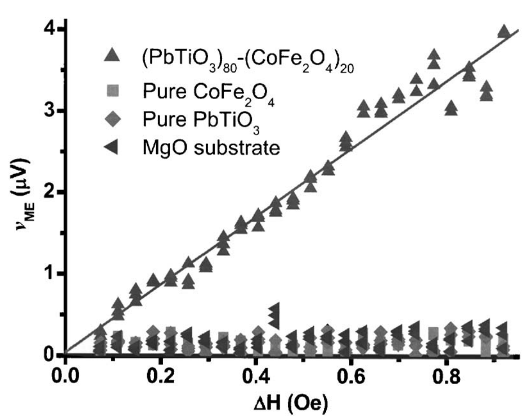


FIG. 3. ME signals of the $(PbTiO_3)_{80}-(CoFe_2O_4)_{20}$ nanocomposite film, pure $PbTiO_3$ film, pure $CoFe_2O_4$ film, and MgO substrate measured as functions of the amplitude of the modulation magnetic field at 92 kHz. The slope of the best fit of $(PbTiO_3)_{80}-(CoFe_2O_4)_{20}$ gives a ME coefficient of 500 mV/cm Oe.

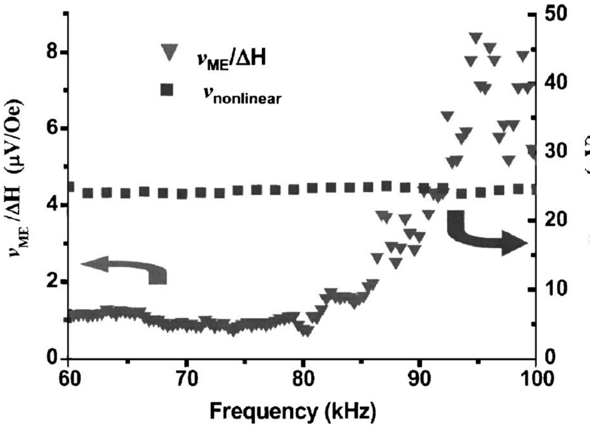


FIG. 4. Frequency dependence of the normalized ME signal (left) and nonlinear dielectric signal (right). Two mechanical resonant peaks were observed in the ME signal while the nonlinear signal is quite flat.

figure. Among them, only $(PbTiO_3)_{80}-(CoFe_2O_4)_{20}$ shows the ME signal, which increases linearly with the field modulation amplitude, attesting to the validity of the measurement. From the least square fitting of the slope in Fig. 3 and the separately measured nonlinear dielectric signal $v_{nonlinear}$, the ME coefficient observed for the present sample at 92 kHz is estimated to be about 500 mV/cm Oe. This value is consistent with that of a $CoFe_2O_4-Pb(Zr,Ti)O_3$ composite thin film sample synthesized using a sol-gel process and measured using the standard capacitive technique.[16] The detection limitation estimated from the noise level in the current measurement setup is about 10 mV/cm Oe.

Shown in Fig. 4 is the frequency dependence of the magnetic-field normalized ME signal and the nonlinear dielectric signal. A scatter in the mechanical resonant response is observed in the ME signal, while the nonlinear signal shows almost no frequency dependence. We believe that the scatter peak around 95 kHz is caused by the dense vibration modes of the MgO substrate, which are most likely the modes with propagation directions along the face diagonal and the cubic diagonal of the rectangular substrate, as the thickness of the substrate is much smaller than the length of the edge of the substrate. The calculated value of the frequency of the lowest resonant mode of the MgO substrate is in fair agreement with the peak position seen in the spectrum. This indicates that the ME effect can be enhanced by exciting the resonant vibration mode of the substrate.[17]

In conclusion, we have constructed a SEMM based ME coefficient measurement technique in the 100 kHz frequency range. This technique has the advantages that it is nondestructive and has a high spatial resolution. It can also be used to obtain other related properties simultaneously. Although the present measurement was carried out at ~100 kHz, which is the limit of the lock-in amplifier used, in principle the operational frequency can be extended to the GHz range. These advantages make the present scheme a unique technique for spatially resolved mapping of magnetoelectric properties of multiferroic films and combinatorial screening of novel magnetoelectric materials.

This work was funded by NSFC, NSRL, MOE (SRFDP program), ONR N000140110761, N00140410085, NSF DMR 0094265 (CAREER), NSF DMR 0231291, and MRSEC DMR-00-80008.


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