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Colossal magnetoelectric effect induced by parametric amplification

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We describe the use of parametric amplification to substantially increase the magnetoelectric (ME) coefficient of multiferroic cantilevers. Parametric amplification has been widely used in sensors and actuators based on optical, electronic, and mechanical resonators to increase transducer gain. In our system, a microfabricated mechanical cantilever with a magnetostrictive layer is driven at its fundamental resonance frequency by an AC magnetic field. The resulting actuation of the cantilever at the resonance frequency is detected by measuring the voltage across a piezoelectric layer in the same cantilever. Concurrently, the spring constant of the cantilever is modulated at twice the resonance frequency by applying an AC voltage across the piezoelectric layer. The spring constant modulation results in parametric amplification of the motion of the cantilever, yielding a gain in the ME coefficient. Using this method, the ME coefficient was amplified from 33 V/(cm Oe) to 2.0 MV/(cm Oe), an increase of over 4 orders of magnitude. This boost in the ME coefficient directly resulted in an enhancement of the magnetic field sensitivity of the device from $6.0\,\mathrm{nT}/\sqrt{\mathrm{Hz}}$ to $1.0\,\mathrm{nT}/\sqrt{\mathrm{Hz}}$. The enhancement in the ME coefficient and magnetic field sensitivity demonstrated here may be beneficial for a variety actuators and sensors based on resonant multiferroic devices. © *2015 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4935332]

Multiferroic materials are widely explored for application where electrical signals are transduced into magnetic signals and vice-versa. In most composite multiferroics, the coupling between electrical and magnetic order is obtained through mechanical coupling at the interface between piezoelectric and magnetostrictive materials. This use of composite structures gives researchers freedom to independently engineer the performance of piezoelectric materials, magnetostrictive materials, and the coupling between them. The relatively strong magnetoelectric (ME) coupling and design flexibility available in composite multiferroics has been leveraged for a variety of applications, such as magnetic field sensors,[1] energy harvesters,[2] phase shifters,[3] and transformers.[4] Hu *et al.* recently demonstrated voltage induced ferromagnetic resonant field shift with multiferroic heterostructures.[5]

To improve the performance of multiferroic materials in these applications, researchers have been making efforts to increase the ME coefficient in both single phase and composite multiferroics.[6–9] Recently, a relatively high ME coefficient of 20 kV/(cm Oe) was reported by Kirchhof *et al.* for a device based on FeBSiC/AlN heterostructure that was operated at a resonance frequency of 152 Hz.[10] For resonant ME devices, it is often desirable to obtain a high mechanical quality factor, as this enhances the ME coefficient. In this study, we explore the possibility of using parametric amplification both to increase the mechanical quality factor of our device and to significantly boost the ME coefficients.

Parametric amplification is a powerful way to increase both the gain and the quality factor of resonant systems. With mechanical parametric amplification, the amplitude of an oscillator at frequency ω_0 can be amplified when the spring constant of the oscillator is modulated at a frequency $2\omega_0/n$, where n is an integer. The greatest increase in amplitude is obtained when $n = 1$, and thus, a modulation of the spring constant at $2\omega_0$ is used here. The spring constant modulation, which we will refer to as the pumping signal, acts to offset damping effects in the oscillator, thus increasing the effective quality factor. Studies have shown that parametric amplification can be very effective in boosting the performance of sensors and actuators in piezoelectric systems with optical measurements[11,12] and MEMS integrated circuit resonators.[13,14]

The device we studied was a heterostructured multiferroic cantilever that consisted of a 500 nm $Fe_{0.7}Ga_{0.3}$ layer, a 500 nm $Pb(Zr_{0.48}Ti_{0.52})O_3$ (PZT) layer, a 35 nm Pt layer, and a 3.2 μm supporting silicon oxide/nitride/oxide (ONO) stack on a Si substrate. Fabrication of the device has been discussed in detail elsewhere.[2] The FeGa layer is magnetostrictive, enabling the cantilever to be actuated by an applied magnetic field. The FeGa layer and Pt layers act as electrodes on the PZT layer, enabling it to be used as a deflection sensor (by reading the voltage across the PZT layer as the cantilever is deflected) or as an actuator (by applying an external voltage to the PZT), or both.

For a linear mechanical oscillator that is driven by an external periodical driving force at ω_d and pumped by a spring constant modulation at $2\omega_d$, we have the equation of motion

$$\underbrace{m\ddot{x} + \frac{m\omega_0}{Q}\dot{x} + [m\omega_0^2 +}_{\text{Classic damped harmonic oscillator}} \underbrace{k_p \sin(2\omega_d t + \theta)]x}_{\text{Parametric pumping}} = \underbrace{F_0 \sin(\omega_d t + \Phi)}_{\text{Driving force}}, \quad (1)$$

where m is the equivalent mass of the cantilever, x is the deflection of the cantilever, ω_0 is the natural resonance frequency of the cantilever, Q is the quality factor without pumping ($k_p = 0$), k_p is the pumping amplitude, ω_d is the frequency of the driving force, θ is the pumping phase, F_0 is the amplitude of driving force, and ϕ is the driving phase.

Eq. (1) is the well-known Mathieu equation, which was solved by Rugar and Griitter for the case of PZT cantilevers,[11] where only the driving phase variation was taken into account. In fact, one can also vary the pumping phase to tune the parametric amplification. The parametric gain can be expressed as a function of both driving phase and pumping phase

$$\mathrm{Gain} = \frac{A_{\mathrm{pump-on}}}{A_{\mathrm{pump-off}}} = \frac{\sqrt{1 + \left(\frac{k_p}{k_t}\right)^2 + \frac{2k_p}{k_t}\cos(2\Phi - \theta)}}{\left|1 - \left(\frac{k_p}{k_t}\right)^2\right|}, \quad (2)$$

where $k_t = 2m\omega_0^2/Q$.

Eq. (2) shows that the gain is sensitive to both driving phase Φ and pumping phase θ, with a period of 180° and 360°, respectively. We see that the gain is maximized when $2\Phi - \theta = n \times 360°$ for integer n, and Eq. (2) becomes

$$\mathrm{Gain} = \frac{A_{\mathrm{pump-on}}}{A_{\mathrm{pump-off}}} = \frac{1}{\left|1 - \frac{k_p}{k_t}\right|}. \quad (3)$$

In Eq. (3), as the pumping amplitude (k_p) approaches the parametric instability threshold (k_t), the gain approaches infinity. According to the Mathieu equation, when the pumping amplitude exceeds the parametric instability threshold, the oscillator becomes unstable, and any small oscillation will grow to infinite amplitude. In practice, non-linearities come into play at large amplitude oscillation and limit the gain.[15] The competition between k_p and k_t is, in fact, the competition between energy getting pumped into the system through parametric pumping and energy getting dissipated through damping. Thus, when the pumping amplitude is set to just below the instability threshold, a significant increases in gain can be expected.

Fig. 1 shows a schematic of our experimental setup. The multiferroic cantilever is mounted in a vacuum chamber

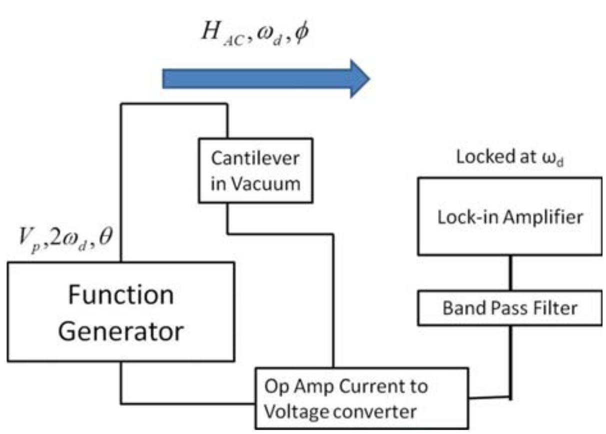

FIG. 1. Block diagram showing a multiferroic cantilever driven by a magnetic-field at angular frequency ω_d and parametrically pumped by a voltage at $2\omega_d$. The response of the cantilever at angular frequency ω_d is measured by a lock-in amplifier.

with a vacuum pressure of approximately 5×10^{-7} Torr. Vacuum is used to enhance the quality factor of the cantilever (and, in turn, the ME coefficient) by reducing the effect of air damping to a negligible level in comparison with the mechanical damping intrinsic to the materials of the cantilever.[2,16–18] A pair of Helmholtz coils is placed outside of the vacuum chamber. The coils provide the AC magnetic field that the device is designed to detect. The magnetic field is aligned with the long axis of the cantilever. The dimensions of the cantilever (950 μm $\times$ 200 μm $\times$ 0.5 μm) are small compared with the Helmholtz coils (10 cm diameter), resulting in a magnetic field that is approximately constant across the device. Since we are operating our device in the magnetic field sensing mode, it is desirable to minimize the background magnetic field noise. To this end, the coils and the vacuum chamber are located inside a rectangular Mu-metal box that screens out environmental magnetic field noise sources with frequencies above 20 Hz. The Mu-metal box also sits on top of a sand box which reduces mechanical vibration noise from the floor.

Parametric amplification is implemented by applying an AC electric field across the PZT layer of the cantilever using a function generator. To achieve optimal parametric amplification, the frequency of the AC electric field is set to twice the frequency of the AC magnetic field and the phase is optimized as described below. The deflection of the cantilever is monitored by measuring the current across the cantilever. The current output from the cantilever was converted to a voltage using a homemade transimpedance amplifier with an amplification that matches the impedance of the device. The input impedance of the amplifier was chosen to match the electrical impedance of the cantilever device at resonance. The output of the transimpedance amplifier was low-pass filtered to remove the parametric pump signal, then measured using a lock-in amplifier. The clocks on the two function generators and the lock-in were synchronized, and the lock-in was configured to use the lower frequency function generator as a reference signal.

As shown in our previous work, the spring constant of the cantilever can be modulated by applying a voltage across the top and bottom electrodes of PZT.[18] In other words, k_p is proportional to V_p.

Then, the maximum gain expression from Eq. (3) becomes

$$\mathrm{Gain} = \frac{A_{\mathrm{pump-on}}}{A_{\mathrm{pump-off}}} = \frac{1}{\left|1 - \frac{V_p}{V_t}\right|}, \quad (7)$$

where V_t is the threshold pumping voltage.

One of the most important characteristics of parametric amplification is that the gain is a function of the source phases. As Fig. 2 indicates, due to parametric amplification, both the device signal and the gain are periodically modulated by the driving phase, with a period of 180°. One can also tune the parametric gain periodically by tuning the pumping phase and fixing the pumping phase. After all, it is the (2ϕ-θ) that matters, as shown in Equation (2). We plot the phase vs. the gain measured at the resonance frequency in Fig. 2(b). We also plot the theoretical fit of the gain in the

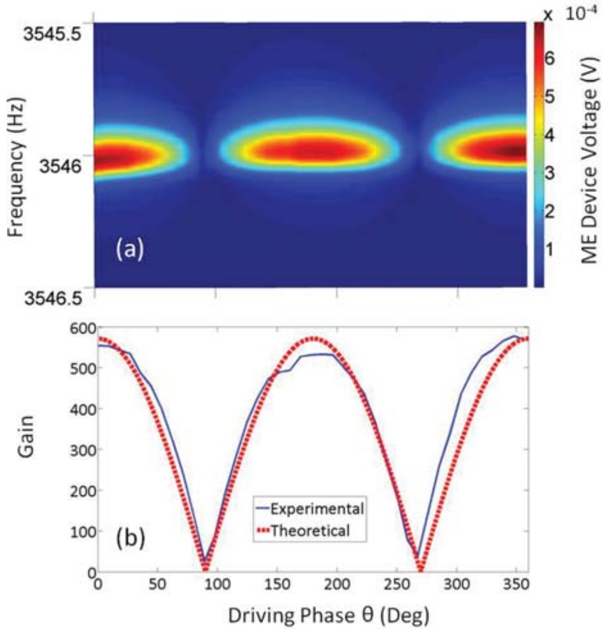

FIG. 2. ME device voltage as a function of driving parameters. (a) ME signal vs. driving phase with pumping phase set to zero. (b) Parametric gain for (a) at the resonance frequency. The driving magnetic field amplitude was approximately 257 nT, and the pumping voltage amplitude was approximately 1.16 V.

same graph, which shows that there is a very good agreement between theory and experiment. These plots point to the importance of properly tuning the phase to achieve the maximum gain.

Fig. 3 shows the response of the ME signal to the pumping voltage, when different driving fields were used. Each data point in this figure is recorded from the maximum ME signal by sweeping the frequency of the driving field upward when the parametric phases are optimized. At low pumping voltages ($V_p < 0.1$ V), the parametric amplification has little effect on the ME voltage. Thus, the ME voltages of the four driving field values reflect the amplitudes of the driving fields. As the pumping voltage is increased up to a threshold voltage, the ME voltages of all four driving field cases increase up to a point where the Duffing nonlinearity takes over, limiting the oscillation amplitude of the cantilver.[17] Assisted by the parametric amplification, the ME voltages were boosted by as much as 3 orders of magnitude. Because all curves have the same Duffing nonlinearity limit, the curve with the lowest driving field (252 nT) can achieve the highest gain (1200) among the four curves. The higher the gain is, the higher the ME coefficient is.

In Fig. 4, We explore the highest gain/ME coefficient that can be measured with parametric amplification. We carried out measurements with an AC driving field of 257 nT and 282 pT for comparison (Fig. 4). As we increase the pumping voltage from 0.8 V to 1.2 V, the gain starts to increase as the pumping voltage gets close to the threshold voltage, V_t. The curve with driving field of 282 pT (Fig. 4, red circles) was able to reach a high ME coefficient of 2 MV/(cm Oe) because it has a lower driving field. However, the red circles data lack the low pumping voltage information because the unamplified signal was lower than the noise floor. The signal became detectable assisted by parametric amplification when the pumping voltage was increased to near the threshold.

One limitation of parametric amplification in the current device and its measurement scheme is due to the drift in the threshold voltage. Because of temperature fluctuation and the instability of the piezoelectric response of the PZT film, we find that the threshold voltage drifts by about ± 5 mV from its average value. This precludes us from exercising ultrahigh parametric gain by getting the pumping voltage arbitrarily close to the threshold voltage.

The other limiting factor for the parametric gain in our devices is the non-linearity of the device. In particular, for large oscillation amplitudes, the cantilevers exhibit a cubic nonlinearity in the equation of motion (Duffing oscillator behavior), as is typical for microcantilever devices.[19,20] In the present devices, the effect of this non-linearity is suppression of the gain (and correspondingly, the ME voltage) in comparison with what one would expect for a linear device. This limiting effect first appears at an output ME voltage amplitude of approximately 0.1 mV and increases in degree until the ME voltage amplitude is saturated at

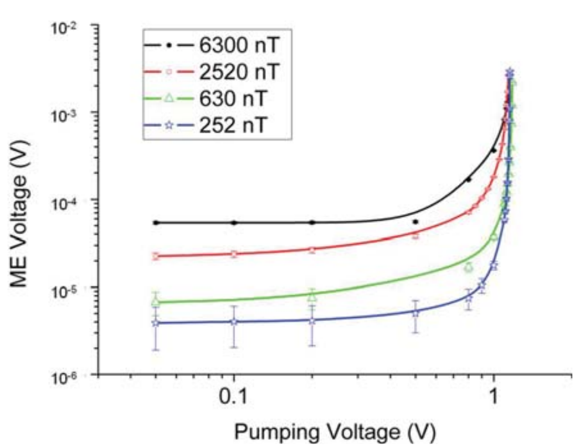

FIG. 3. The ME signals as a function of the pump voltage for different driving fields (252 nT, 630 nT, 2.52 μT, and 6.30 μT). Lines are not theoretical fitted lines but just guidelines. The error bars stand for the noise.

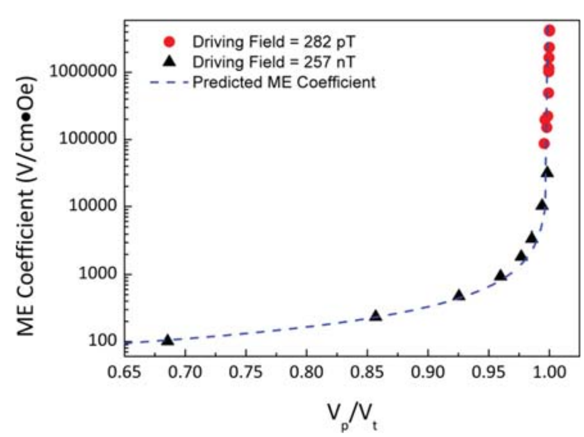

FIG. 4. Parametric amplification theory and experiment: The experimental ME coefficient (black triangles) and the theoretical ME coefficient curve (blue dashed line) as a function of pumping voltage for a driving field of 257 nT. The theory curve is plotted from Eq. (7) with $V_t = 1.166$ V. Red circles show data at the minimum driving magnetic field (282 pT) with the current measurement setup.

approximately 0.4 mV. Thus, in order to observe the maximum parametric gain from our device, we need to keep the cantilever oscillation below certain amplitude.

With these limitations, the smallest AC magnetic field, which can be consistently amplified and detected at the fundamental frequency of 3550 Hz, is about 282 pT, and the highest ME coefficient that has been recorded is 2 MV/(cm Oe) using the amplification. To the best of our knowledge, this is the highest ME coefficient that has been reported in the literature till date. Taking into account the equivalent noise bandwidth of our measurement setup, this field corresponds to $1\,\mathrm{nT}/\sqrt{\mathrm{Hz}}$. In addition to the amplification, enhancement in quality factor from around 3000 to 35 000 was observed. This enhancement in quality factor and sharpening of the resonant peak in frequency space can be useful for detection techniques where high Q resonance is desired, for example, in atomic force microscopy. In the future, by introducing measures to stabilize the threshold voltage, we hope to be able to reliably perform pumping at the top region of the curve in Fig. 4. In doing so, we expect to be able to enhance the field sensitivity by up to another 2 orders of magnitude: this corresponds to $\sim$3 pT or $10\,\mathrm{pT}/\sqrt{\mathrm{Hz}}$.

In conclusion, we have demonstrated parametric amplification of a multiferroic cantilever device operated in the magnetic field sensing mode. A magnetic field was detected by a magnetostrictive FeGa layer and the pumping signal was provided through a piezoelectric PZT layer. A parametric gain of 60 000, a ME coefficient of 2 MV/(cm Oe), and a quality factor of 35 000 were obtained with the parametric amplification method. The magnetic field sensitivity of $1\,\mathrm{nT}/\sqrt{\mathrm{Hz}}$ was achieved with parametric amplification with the present measurement setup. The parametric amplification offers an additional dimension that can be exploited for sensing using ME devices. In addition to stabilizing the parametric instability threshold voltage, lowering of the noise floor of the device by making device arrays or devices with higher resonance frequencies may offer other avenues for enhancing the sensitivity. High-frequency resonant detection with parametric amplification may also open the door for novel antennae applications.

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