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## Induced magneto-electric coupling at ferroelectric/ferromagnetic interface

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We have demonstrated that at room temperature the electric polarization of a ferroelectric polymer, polyvinylidene fluoride (PVDF), in an artificial multiferroic structure can be controlled by applying an external magnetic field. We found that as the applied magnetic field is changed, the switching of electric polarization for the PVDF depends on the external magnetic field. We also noticed that both the electric coercivity and polarization of the PVDF polymer display hysteretic features as the applied magnetic field is changed. We found that the thickness of the PVDF layer has an effect on the magnetoelectric coupling strength in our samples. The coupling sensitivity increases as the PVDF layer thickness increases. It is possible to control the ferroelectric properties of a PVDF film by tuning the external magnetic field. © 2013 American Institute of Physics.

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### INTRODUCTION

Room temperature manipulation of magnetic states by electrical means<sup>1,2</sup> and, conversely, magnetically manipulated electronics<sup>3,4</sup> are among the most promising approaches towards developing novel information and technology devices. Materials with ferroelectric and ferromagnetic properties or magneto-electric (ME) coupling effects<sup>5-7</sup> are the potential candidates for this purpose.<sup>8</sup> Preparation and characterization of multiferroic materials would represent a milestone for multifunctionalized materials.<sup>5,9</sup> Both single phase and composite multiferroic studies have been reported.<sup>10-15</sup> Typically, the ME coupling constant at room temperature for single phase materials is usually quite small. The multiferroic composite heterostructures show relatively large room-temperature extrinsic ME effect. The reported values for the magneto-electric coupling sensitivity for composite ME materials range from 1 V/(cm × Oe)<sup>16</sup> to 6 V/cm Oe,<sup>17</sup> and up to 21 V/(cm × Oe).<sup>18</sup> Compared with bulk ME composites, multiferroic ME films can be combined at the atomic level. Recently, pioneering work on multiferroic ME films has been reported by few groups.<sup>19-24</sup> A more impressive coupling sensitivity of up to 310 V/(cm Oe) was reported for a polyvinylidene-fluoride (PVDF)-metglas laminate composite.<sup>25</sup> The origin of the ME coupling in polymer-based nanoscale heterostructures is not clearly established.

In this experiment, we have synthesized multiferroic heterostructures consisting of ferromagnetic iron layers and ferroelectric PVDF polymer layer. We have observed giant magnetoelectric coupling in our samples, i.e., by applying an external magnetic field, we are able to tune the electric polarization of the ferroelectric PVDF layer. Moreover, as the applied magnetic field is varied, the properties of PVDF

layer show hysteretic features. We have also noticed that changing the thickness of the PVDF layer will affect the magnetoelectric coupling strength between the ferroelectric and ferromagnetic layers. The magnetoelectric coupling sensitivity was also estimated which is in the range of 3700 V/cm Oe for our sample.

### EXPERIMENTAL TECHNIQUE

Heterostructured multilayer samples, schematic shown in Fig. 1, are fabricated using physical vapor deposition for magnetic layers and the Langmuir-Schaefer film fabrication method for PVDF layers as described in the previous experiments.<sup>26</sup> The thickness of each sample is precisely controlled and calibrated. A Radiant Technology Precision ferroelectric measurement system was used to measure the polarization versus electric field hysteresis loops of all the samples. Each sample was placed between the pole pieces of an electromagnet with the external magnetic field applied parallel to the sample surface.

### RESULTS AND DISCUSSIONS

Fig. 2 shows the polarization versus electric field (P-E) hysteresis loops measured under different magnetic fields for a multilayer sample with the structure of Fe(80 nm)/PVDF(70 nm)/Fe(26 nm). The magnetic field was varied in such a way that it changes from -880 Oe to 880 Oe and then from 880 Oe to -880 Oe again. Each P-E loop was recorded under an applied magnetic field. It can be clearly seen that as the magnetic field applied, there is a change on the electric properties of the PVDF polymer. The area under P-E loops shows a strong dependence on the external magnetic field. The energy product (area under each P-E loop) shows a minimum at -880 Oe and then it increases as we increase the external magnetic field. When the magnetic field is applied at -880 Oe, as shown in Fig. 1, the polarization can

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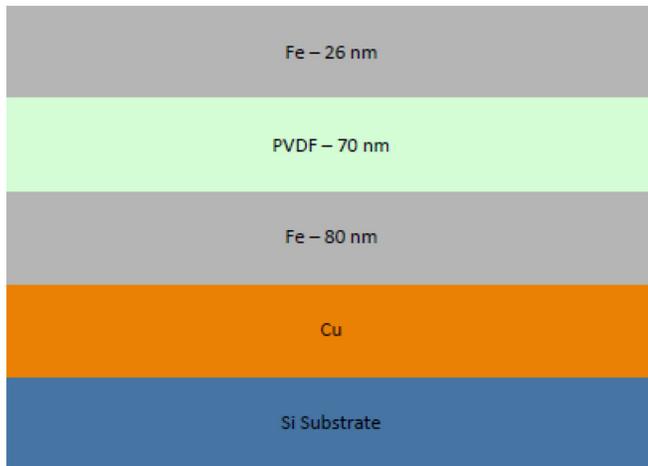


FIG. 1. The schematic graph of the Fe (80 nm)/PVDF (52 nm)/Fe (26 nm) heterostructure.

be saturated at 6.9 MV/m, while the polarization is switched under coercivity of  $-5.7$  MV/m. As the applied magnetic goes to zero, the electric polarization of the PVDF polymer layer saturates at electric field of 8.4 MV/m, and the coercivity is at  $-5.9$  MV/m field. So, the PVDF polymer becomes hard to be saturated and flipped as the magnetic field changed from  $-880$  Oe to 880 Oe. The P-E loop taken at 880 Oe magnetic field shows that the polarization can be saturated at an electric field of 10.7 MV/m and it can be flipped at  $-9.5$  MV/m. When the magnetic field goes back to zero from maximum field, the electric polarization saturates at electric field of 8.9 MV/m and the coercivity is at  $-8.1$  MV/m electric field. The P-E loop taken at this zero magnetic field (labeled as 0 Oe down in Fig. 2) is different from the results taken at the other zero magnetic field which changes from  $-880$  Oe (labeled as 0 Oe up). So, the history of the sample plays a role and hysteresis shows up as indicated in Fig. 2.

In order to characterize the magneto-electric coupling of the sample, we choose an electric bias field of 7.0 MV/m, and plot the polarization versus magnetic field, the result is

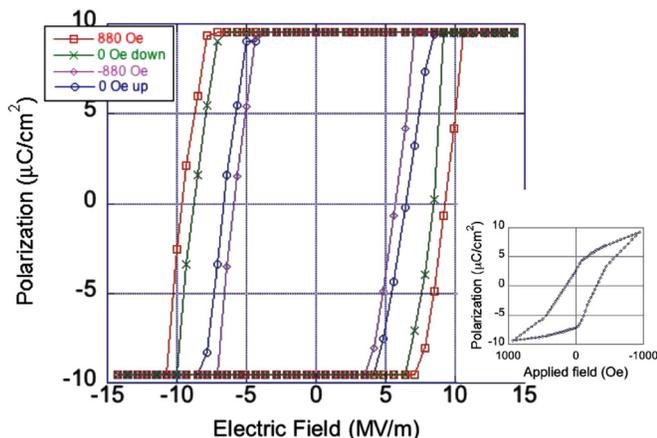


FIG. 2. P-E hysteresis loops of the Fe (80 nm)/PVDF (70 nm)/Fe (26 nm) heterostructure under different applied magnetic fields. The lower inset shows the polarization of the polymer as a function of the applied magnetic field at an electric bias field of 7.0 MV/m.

shown as the inset of Fig. 2. We also plot the electric coercivity and electric saturation field of the PVDF polymer layer under different magnetic fields as shown in Fig. 3. From those data, it can be clearly seen that both the electric polarization and the electric coercivity show hysteretic feature upon external magnetic field. This observed magneto-electric coupling effect originates from the interface between ferroelectric and ferromagnetic. The hysteresis dependence of PVDF upon magnetic field plotted in Fig. 3 is due to the hysteresis of Fe magnetic layers upon magnetic field. Having seen that the magneto-electric coupling is present in our sample, we also studied a series of samples with different PVDF polymer thicknesses while the ferromagnetic iron layers were kept at a constant thickness. In order to compare the magneto-electric coupling strength in each of the sample, the opening width of the hysteresis curve of  $E_C$ -H and  $E_{sat}$ -H graphs is analyzed. This opening width indicates the effect of the magnetic field on the ferroelectric property. The wider the opening of the hysteresis curves plotted in Fig. 3, the stronger the magneto-electric coupling strength. We noticed that as the PVDF thickness increases, the width of the  $E_C$ -H hysteresis loops also increases.<sup>27</sup> This indicates that the magneto-electric coupling strength is getting larger as the PVDF thickness increases. This cannot be explained by the simple strain effect. First, according to strain effect, the magnetostriction stress is kept the same for all our samples by keeping the iron layers unchanged. When this same stress (or pressure) is applied to a thicker PVDF layer, the effect is expected to be smaller which is contradictory to our observed data. Second, magnetostriction does not depend on the polarity of the external magnetic field; however, our data show dependence on the polarity of the magnetic field. We believe it is due to the significant magnetic field in the polymer layer vicinity generated by Fe ferromagnets and this generated magnetic field is hysteretic and it is much larger than the external applied magnetic field due to the ferromagnetism of Fe. The magnetic field generated by magnetized Fe layer can exert a force on rotating dipoles of PVDF and affect the rotation of those electrical dipoles. The direction of this force does depend on the polarity of the magnetic

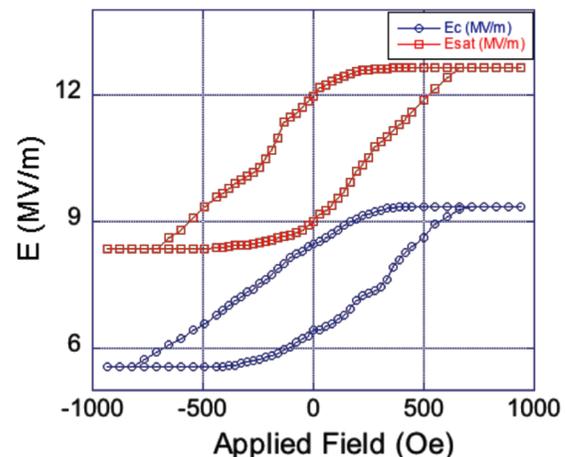


FIG. 3. Electric coercivity and electric saturate polarization as a function of the applied magnetic field, showing that the coercivity and saturation field of the PVDF layer depend on the magnetic field.

field. As a result of that, this additional force can affect and tune the polarization properties of the PVDF layer. The linear magneto-electric coupling constant is often defined as:  $\alpha_E = \Delta E / \Delta H$ . We have estimated the magneto-electric coupling strength by extracting the change in the electric field of the polymer layer with applied magnetic field from the polarization versus magnetic field plot in the region of low applied fields, as inset of Fig. 1. The values of magneto-electric coupling coefficient for our sample are around 3700 V/(cm Oe). This value is much higher than that of the previously reported. We have now shown that a large electrical response for only a small applied magnetic field change is ideal for magnetic sensors.

## CONCLUSIONS

We have been able to create ferromagnetic/ferroelectric heterostructures by sandwiching a layer of PVDF polymer between layers of iron thin films. Our measurements have shown that this heterostructure exhibits the magneto-electric effect. The electric properties of the PVDF polymer can be tuned when a magnetic field is applied to the sample and this tuning effect shows clear hysteresis upon magnetic field. It is also seen that the thickness of the PVDF layer affects the magneto-electric coupling strength.

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