A four-state memory cell based on magnetoelectric composite

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A four-state memory can store four states in each memory cell. We designed a four-state memory cell using Co/PZT magnetoelectric composite and observed a broad magnetoelectric hysteretic output loop on applying magnetic field. Based on magnetoelectric hysteresis, we developed a read method by applying a bias magnetic field on the memory cell. Results gave clearly four-state signals of 15.8, −4.4, 5.5 and −11.3 μV, which demonstrated the feasibility of our design.

multiferroic, magnetoelectric composite, four state memory, device, read

To meet the increasing need of multimedia storage, many efforts have been made to develop storage technique with higher storage speed and higher storage density. Besides the traditional two-state (0 and 1) memory, multi-state memory, which can store more than two states in a memory cell, can improve the storage density and may become the next generation storage technique. Materials for multi-state memory media have multi-stable physical states and these states can be sensed by certain physical properties, such as resistance of GST in multi phases¹ or magnetoresistance in multilayer tunnel junction with multi spin states². Such multi-state memory materials are rare in nature and need a further search.

Multiferroic material with ferroelectric and ferromagnetic coexisting is a candidate for nonvolatile memory because of its four polarization states (±P, ±M). According to the chemical composition, there are two kinds of multiferroic materials, multiferroic compound and multiferroic composite (also called magnetoelectric composite). Because of the limitation in the crystal symmetry, multiferroic compounds with single phase are rare in nature. And the Néel’s or Curie’s temperatures of most of them (except BiFeO₃) are far lower than room temperature, which becomes the obstacle of their industrial application. The other kind of multiferroic materials, multiferroic composites are made by artificial combination of ferroelectric phase and ferromagnetic phase from macro-scale to nano-scale. This artificial multiferroicity can be easily achieved at room temperature and has large potential applications on electric and magnetic devices. For multi-state memory application, the multiferroicity is the fundamental for information storage, while the coupling effect such as magnetoelectric effect³ and magneto-dielectric effect⁴ is the mechanism for the read and write procedure. For example, the M-E coupling effect could be used to read out the state of electric polarization instead of the damage read operation in FERAM, which can extend memory’s service life.

The single phase multiferroic compounds have drawn much attention to memory applications. Recently, strong

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Received October 30, 2007; accepted January 20, 2008
doi: 10.1007/s11434-008-0275-8

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Supported by the Hi-Tech Research and Development Program of China (Grant No. 2006AA03Z101) and National Natural Science Foundation of China (Grant No. 50571084)
coupling between magnetic polarization and electric polarization was observed in single crystal TbMn$_2$O$_5$ [5], making it possible to reverse the electric polarization by applying the magnetic field. This strong coupling effect was expected to be a new write mechanism of multiferroic memory cell. Most recently, a magnetic tunnel junction with multiferroic La$_{0.1}$Bi$_{0.9}$MnO$_3$ barrier demonstrated a breakthrough of four-state multiferroic memory [6]. The resistance of the tunnel junction obviously exhibited different values under negative and positive bias electric field, and more importantly its magnetoresistance hysteresis confirmed the junction could ‘remember’ magnetic polarization. Namely, this four-state memory cell could be written by applying electric field and magnetic field, and read out by measuring its magnetoresistance. However, this multiferroic film should be operated at a very low temperature as 3 K [6], which restricted its industrial application. Furthermore, in some single phase multiferroic compounds, the strong intrinsic coupling makes the four polarization states not truly independent, resulting in only two independent states [7]. Therefore there are still a lot of problems in multi-state memory cell made by multiferroic compound.

Besides compounds, multiferroic composite, this artificial multiferroicity can also generate multi states for memory application. In multiferroic composite, the coupling effect between ferroelectric composition and ferromagnetic composition is a mechanical media coupling effect, called magnetoelectric effect. That is, on applying a magnetic field, the induced magnetostrictive stress/strain in ferromagnetic phase passes to the ferroelectric phase through the phase interface, and then an electric polarization is induced by piezoelectric effect. Hence, this extrinsic coupling is much weaker than the intrinsic one in multiferroic compound, and not strong enough to reverse the polarization in composite, so that the four states in multiferroic composite are really independent from each other. Based on magnetoelectric effect, many device prototypes had been developed, such as magnetic sensor [8], miniature transformer [9] and electric gyrations [10], but there has been few researches on four-state memory. In this work, we make a four-state memory cell using multiferroic composite, and present a write-read procedure similar to previous La$_{0.1}$Bi$_{0.9}$MnO$_3$ tunnel junction. This work will demonstrate the feasibility of four-state memory cell based on magnetoelectric composite.

1 Experimental

For cobalt and PZT are widely used in the magnetic recording memory and ferroelectric memory respectively, a bi-layered magnetoelectric composite was made by gluing a Co layer with a PZT layer together, as shown in Figure 1. The dimensions of the Co and PZT layer were 11 mm×5.5 mm×0.5 mm and 11 mm×5.5 mm×1.0 mm, respectively. The PZT with silver electrodes was poled by an electric field of 2 kV/mm. Then the Co and PZT layers were polished and glued by the epoxy.

![Figure 1](image1.png)

The memory cell consisted of a piece of Co/PZT composite and a driving coil. The coil with about 40 rounds was surrounding the composite, as shown in Figure 1. The memory cell had two pairs of wires. One was the output terminal which was connected with the Co/PZT composite. The other was the driving terminal which was connected with the coil. A lock-in amplifier (EG&G Model 5210) was used to drive the coil generating a small disturbing magnetic field at 10 kHz and correspondingly read the amplitude and phase of the magnetoelectric output in Co/PZT composite. Here, the phase was used to define the ME signal’s sign [8,11].

The polarization directions of the ferromagnetic/ferroelectric composite can be controlled by the applied high magnetic field and high electric field, respectively. The combination of remnant ferroelectric polarization and magnetic polarization in the memory cell exhibits four physical states as shown in Figure 2, denoted as I, II, III and IV.

![Figure 2](image2.png)
2 Results and discussion

On the applied magnetic field, a broad hysteresis loop of the magnetoelectric output in this memory cell was observed as shown in Figure 3. Similar to the M-H hysteresis loop, we name this loop as magnetoelectric (ME) loop. The coercive field of the ME loop is about 200 Oe (1 Oe = 79.58 A/m), which means the cobalt layer is hard enough to record the direction of writing magnetic field. In Figure 3, each ME loop has center symmetry, like M-H hysteresis loop. And the ME loop also reveals mirror symmetry between loop (II, I) (the solid line in Figure 3) and loop (IV, III) (the dot line in Figure 3), which means when the electric polarization reverses, the sign of the ME signal will reverse too. The maximum amplitude of loop (III, IV) is a little smaller than that of loop (II, I), which may result from the mechanical restriction of cobalt layer during the reversion of electric polarization in PZT layer.

The four states in Figure 2 correspond to the four junctions of the ME loops and the axis of $H=0$ in Figure 3. When $H=0$, the ME voltage output of case I and case III are almost equal of about 15 $\mu$V, while case II and case IV are almost equal of about −12 $\mu$V. That is because, as shown in Figure 2, both the remnant magnetization and the remnant electric polarization of case I are opposite to those of case III, so the ME output of case I and case III are equivalent because of the symmetry of the magnetoelectric effect. Similarly, the ME output of case II and case IV are equivalent too. Therefore it seems that only two independent states can be sensed by magnetoelectric effect. However, if we apply a small external bias magnetic field (less than coercive field) on the memory cell, the cobalt’s remnant magnetization in case II and case III will be somewhat reduced (see Figure 4), and the symmetry of the four states will be broken, which makes the four states distinguishable. After applying the positive bias magnetic field, the value of ME output of the four states will change along different directions as shown in Figure 3. For case I and case III, case I changes along the initial part of ME loop, so the amplitude of output is almost constant; and case III changes along the demagnetization part of ME loop, so the output of case III significantly decreases. Therefore these two formerly equivalent states become distinguishable. So does it for case IV and case II.

To further illustrate the working mechanism of this four-state memory cell, a writing-reading operation was performed. First, a write magnetic field of 1.5 kOe and a write electric field of 2 kV/mm were applied on the memory cell, respectively. After writing, the remnant polarization ‘remembered’ the direction of the write field. Then, a read bias magnetic field $H_{\text{read}}$ of about 150 Oe (the dash dot line as shown in Figure 3) was applied and the output signal of memory cell was measured by the lock-in amplifier. Results were shown in Table 1.

The measured results of lock-in amplifier include amplitude $R$ and phase $\theta$, as shown in Table 1. After defining the sign of $R$ by phase $\theta$, the output ME signal of 15.8 $\mu$V, −4.4 $\mu$V, 5.5 $\mu$V and −11.3 $\mu$V show clearly
four states as expected in Figure 4, which are in good agreement with the dots in Figure 3. For case I and case II, they have the same electric polarization but opposite magnetization, so the difference of their phases is about 180°, i.e. their signs are opposite. The amplitudes of case II and case III are reduced by the bias magnetic field, for their remnant magnetization were reduced as shown in Figure 4. Although the bias magnetic field is necessary to distinguish the four states, its reduction of magnetization will weaken the recording ability. And if the bias magnetic field is too large to reverse the magnetization error code of memory will occur. So to get a stable four-states recording, the magnetoelectric loop of memory cell should have enough broad hysteresis and enough decline. Magnetic composition with high remnant magnetization and high coercive field is needed to guarantee a high performance ME memory cell.

3 Conclusion

A four-state memory cell prototype has been made using multiferroic composite Co/PZT. A read procedure based on the magnetoelectric effect under bias magnetic field is developed and the reading results show clearly four states, which are in good agreement with their previous writing states. This work demonstrates the feasibility of four-state memory cell made by multiferroic composite. Further studies can be carried out on multiferroic composites with various compositions and structures. Furthermore, multiferroic composite films in nano-scale have attracted much attention recently[12], but their application on memory is few, which also needs our further studies.