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Magnetic control of dielectric properties of $Ba_{0.6}Sr_{0.4}TiO_3$ in a trilayer system

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Abstract

Dielectric properties of $Ba_{0.6}Sr_{0.4}TiO_3$ (BST) in a layered terfenol-D/BST/terfenol-D system were investigated. Dielectric properties of the BST layer can be adjusted by magnetostrictive strain of terfenol-D layers under applied magnetic fields. Our results show that the small-signal dielectric constant of BST can be tuned ranging from about 2300 to 4000 by a small magnetic field. By adjusting the relative layer thickness ratio, a more broad adjustable dielectric constant and tunability of BST layer can be designed, which may have potential applications in sensitive multifunctional devices and microwave devices.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Dielectric materials with high tunability and dielectric permittivity have attracted much attention because of their potential applications in next generation dynamic random access memories with higher integration densities and voltage-sensitive and frequency-agile microwave electronic devices, such as phase shifters, varactors, tunable filters, resonators and antennas [1, 2]. $Ba_xSr_{1-x}TiO_3$ $(0 < x < 1)$ thin films have high dielectric constants, high tunability, low dielectric-loss tangent and low leakage current [3]; most importantly, it can be integrated on a traditional Si substrate to replace the current silicon oxide and nitride dielectrics, which would have considerable commercial impact.

Both theoretical and experimental works have shown that the dielectric constant of $Ba_xSr_{1-x}TiO_3$ is highly dependent on the temperature, the Ba/Sr ratio and internal or external stresses [4–7]. By changing one or more factors, the dielectric properties can be tuned broadly, especially in the layered or graded composites [8,9]. It is well known that in the vicinity of the paraelectric-to-ferroelectric phase transition temperature, the thermodynamic properties of BST show large anomalies accompanied with large increases in dielectric constant and tunability [10, 11]. Since the phase transition temperature is highly stress dependent, we can use an external tunable stress to tune the dielectric properties of BST. Recently, magnetodielectric multilayers have been of current interest [12,13]. The use of an external magnetic field to turn the stress is extremely attractive for applications because the controllable dielectric materials will provide more freedom for designing agile and multifunctional devices.

Following the route of multiferroic composites which are composed of piezoelectric and magnetostrictive materials, we use the alloy terfenol-D ($Td_xDy_{1-x}Fe_y$ $0.27 \leqslant x \leqslant 0.3$, $1.9 \leqslant y \leqslant 2$) as the source of tunable stress. Terfenol-D is widely used in transducers, actuators, sensors and multiferroic functional devices because of its low magneto-crystalline anisotropy, giant magnetostriction at low magnetic field [14] and little decrease in magnetostriction under cyclic stress [15]. For further application, composites of terfenol-D are exploited to avoid high frequency limitation due to eddy current loss [16]. Under an external magnetic field, terfenol-D will have a shape change, which is a nonlinear function of the external field. By the use of efficient magnetostrictive stress mediated to the $Ba_xSr_{1-x}TiO_3$ layers though the interface, we can tune the dielectric constant of $Ba_xSr_{1-x}TiO_3$ conveniently.

In this paper, we study and design an intriguing laminate structure consisting of a magnetostrictive material (terfenol-D composite) and a dielectric material ($Ba_{0.6}Sr_{0.4}TiO_3$ (BST))

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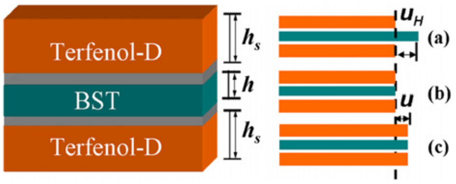

Figure 1. Schematic of the trilayer structure. (*a*) Terfenol-D is constrained under magnetic field and each part is free standing; (*b*) state of $Ba_{0.6}Sr_{0.4}TiO_3$ totally constrained by up and bottom layers of terfenol-D with the same length under magnetic field; (*c*) free state with all the layers reaching a balance.

based on the method of Timosheko [17]. The dielectric properties of BST thin films under different magnetic fields are investigated in the framework of Ginzburg–Landau–Devonshire theory (GLD).

2. Thermodynamic model

As shown in figure 1, the BST thin film with top and bottom $SrRuO_3$ electrodes is sandwiched between polymer-bonded terfenol-D composites. Piezoelectric and magnetostrictive layers with thicknesses of h and h_s, respectively, were bonded together with epoxy. We neglect the elastic effect of the electrodes and the epoxy in this model by assuming that their thicknesses are rather small compared with those of BST and terfenol-D. Under an external magnetic field which is considered in the direction vertical to the film surface, a large magnetostrictive stress can be induced in a free-stand terfenol-D layer. If there is no slippage or fracture in the bonded trilayer, the magnetostrictive strain applies an in-plane constraint on the BST layer and helps BST to have a spontaneous polarization along the z axis according to its phase diagram [18]. Stresses in the trilayer are related to the electrostrictive, magnetostrictive, elastic properties and thickness of each layer. The stress state can be calculated following the method of Timosheko [17]. First, let terfenol-D deform freely under the external magnetic field with the direction vertical to the film. Terfenol-D will have a magnetostrictive strain $u_{33}(H)$ along the same direction, which will give the free-stand terfenol-D layer a compressive strain in the x–y plane, i.e. $u_{11} = u_{22} = u_H$ (figure 1(*a*)). According to the equilibrium $\sigma_{11} = 0$, we can find $u_H = -u_{33}c_{12}/(c_{11}+c_{12})$. u_H will only be the nonlinear function of the external field if we neglect the external stress effect on the magnetostrictive properties of polycrystalline terfenol-D [19]. Second, let the FE layer, which has the same length as the initial state of terfenol-D, shrink with the terfenol-D layers which are in the first state (figure 1(*b*)). If the layers are perfectly bonded, the unconstrained and incompatible in-plane strains in the FE layer will be $\varepsilon_{11} = \varepsilon_{22} = u_H - Q_{12}P^2$. Third, free the layers; the constraint imposed on each layer is relaxed uniformity and achieves displacement compatibility and equilibrium. The built-in or the residual equilibrated uniform stress u can be found in the final configuration. The induced elastic strain in the BST layer and the terfenol-D layer are $u_H - Q_{12}P^2 + u$ and u, respectively.

We consider that both the BST and terfenol-D layers are cubic elastic media with moduli C_{11}, C_{12}; $\bar{C}_{11}$, $\bar{C}_{22}$, respectively. The effective elastic constants are given by $G = C_{11} + C_{12} - 2C_{12}^2/C_{11}$ and $\bar{G} = \bar{C}_{11} + \bar{C}_{12} - 2\bar{C}_{12}^2/\bar{C}_{11}$. The strain distributions in the trilayer system are determined by the equilibrium conditions [20]:

$$\int_{-h_s-h/2}^{-h/2} \bar{G}u\,\mathrm{d}z + \int_{h/2}^{h_s+h/2} \bar{G}u\,\mathrm{d}z + \int_{-h/2}^{h/2} G(u + u_H - Q_{12}P^2)\,\mathrm{d}z = 0. \tag{1}$$

The uniform strain component u from equation (1) can be solved as

$$u = -\frac{Gh\left(u_H - \dfrac{Q_{12}}{h}\displaystyle\int_{-h/2}^{h/2} P^2\,\mathrm{d}z\right)}{2\bar{G}h_s + Gh}. \tag{2}$$

Finally, we can find the total elastic energy F_{ela} of the trilayer:

$$F_{\mathrm{ela}} = \int_{h/2}^{h_s+h/2} \bar{G}\varepsilon(z)\varepsilon(z)\,\mathrm{d}z + \int_0^{h/2} G(u + u_H - Q_{12}P^2)(u + u_H - Q_{12}P^2)\,\mathrm{d}z. \tag{3}$$

The total free energy per unit area of the film layer as the summation of electrical, surface and mechanical components can be written as [21, 22]

$$F = F_0 + F_P + F_{\mathrm{ela}} + F_{\mathrm{ele}} + F_{\mathrm{surf}} = F_0' + \int_{-h/2}^{h/2}\left[\frac{1}{2}A^*P^2 + \frac{B^*}{4}P^4 + \frac{C}{6}P^6 + \frac{1}{2}D_{11}\left(\frac{\mathrm{d}P}{\mathrm{d}z}\right)^2 - E_{\mathrm{ext}}\right]\mathrm{d}z + \frac{1}{2}\int_s D\frac{P^2}{\delta}\,\mathrm{d}s. \tag{4}$$

F_0 is a constant as the reference state and F_0' is the normalized one. F_P is the Landau type energy. F_{ela}, F_{ele} and F_{surf} are contributions of the actions on the reference state caused by mechanical stresses, electric fields and surface, respectively. $A^* = A(T - T_{c0}) - 4GQ_{12}(u_H + u)$, $B^* = B + 4GQ_{12}^2$ are the normalized Landau coefficients considering the applied stress effect. T_{c0} is the Curie temperature and Q_{12} is the electrostrictive constant.

The temporal evolution of the polarization field can be described by the time-dependent GLD equation [23]:

$$\frac{\partial P}{\partial t} = -L\frac{\delta F}{\delta P} = -L\left(A^*P + B^*P^3 + CP^5 - D\frac{\mathrm{d}^2P}{\mathrm{d}z^2} - E_{\mathrm{ext}}\right). \tag{5}$$

The surface items yield the boundary conditions as

$$\frac{\partial P}{\partial z} = \mp\frac{P}{\delta} \qquad \text{for} \quad z = \pm\frac{h}{2}. \tag{6}$$

With the boundary condition, equation (6), equation (5) can be solved numerically to yield the steady-state polarization distribution.

Since the appearance of spontaneous polarization and dielectric constant is highly temperature dependent, it is very important to find the phase transition temperature which can be significantly affected by the external stress induced by the magnetic field. According to linear analysis theory, the phase transition temperature of BST can be found by analysing the critical conditions of the dynamic stabilities of the stationary states [23]. By applying an infinitesimal perturbation Δ to the trivial stationary solution $P_0 = 0$ and neglecting the higher order small qualities, we rewrite equation (5) with only the terms linear in Δ as

$$\frac{\partial \Delta}{\partial t} = -L\left(A^*\Delta - D_{11}\frac{\partial^2 \Delta}{\partial z^2}\right). \tag{7}$$

In the same way, the boundary condition (equation (6)) can be derived only with P replaced by Δ.

The parameters δ in equation (6) give different types of the surface effect. For simplicity, we consider the more common case $\delta > 0$. By using separating variables and applying the boundary conditions, we can find the eigenvalue ω. According to the stability analysis, the supercooling temperature can be found explicitly from the critical condition $\omega = 0$:

$$T_c = T_{c0} + \frac{1}{A}[4GQ(u_H + u) - Dk_c^2], \tag{8}$$

where k_c is the smallest value of k solved from the boundary condition. As shown in the above expression, the phase transition temperature is highly dependent on the external magnetostrictive stress and the induced elastic stress. By adjusting the thickness ratio of the relative layers and the external magnetic field, we can design the phase transition temperature as desired.

The out-of-plane electric field dependent dielectric permittivity can be written as

$$\varepsilon^{-1} = \varepsilon_0(A^* + 3B^*\langle P\rangle^2 + 5C\langle P\rangle^4), \tag{9}$$

where ε_0 is the permittivity of free space. $\langle P\rangle$, the average polarization, which can be described as $\langle P\rangle = \int_0^h P\,\mathrm{d}z/h$, is zero in the paraelectric state and nonzero in the ferroelectric state. The small-signal dielectric constant can be determined with $\langle P\rangle$ solved from the static state in the absence of the external field. The dielectric constant will be very high in the vicinity of the paraelectric-to-ferroelectric phase transition temperature because of the unstable state of polarization in the ferroelectric phase or the non-poled state in the paraelectric state.

The dielectric tunability is a very important factor for dielectric materials. We use the relative tunability ($n_r = 1 - (\varepsilon(E))/(\varepsilon(0))$) to characterize the dependence of the dielectric permittivity on the applied dc electric fields.

3. Results and discussions

Taking the parameters of BST from [24] and of Terfenol-D from [19], we can derive the phase transition temperature using equation (8). As shown in figure 2, the phase transition temperature can be adjusted as broad as almost 30 K by changing the external magnetic field and the thickness ratio of the relative layers. Because the dielectric properties are highly dependent on the phase transition temperature, the results mean that the external magnetic field can adjust the dielectric properties much more agilely and precisely, which give more freedom and convenience for the use of dielectric properties especially in the microwave communication and IR detector.

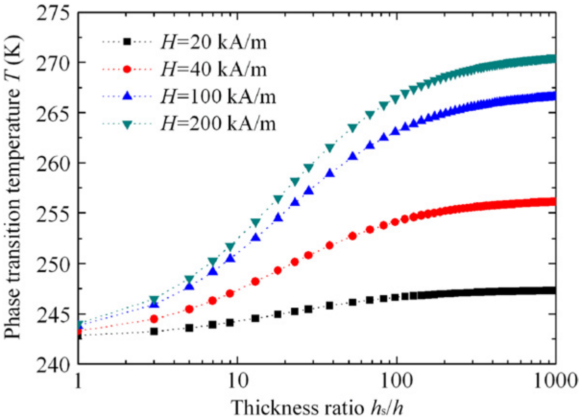

Figure 2. Phase transition temperature of $Ba_{0.6}Sr_{0.4}TiO_3$ versus the thickness ratio of terfenol-D to $Ba_{0.6}Sr_{0.4}TiO_3$ under external magnetic fields.

To investigate the dielectric properties of BST under an external magnetic field, we calculated the dielectric constant with the thickness ratio of 100 versus temperature under different external magnetic fields (figure 3). As shown in figure 3(*a*), the dielectric constant is controllable via the external magnetic field. Because of the relatively small magnetostrictive constraint typically less than 0.5%, the external magnetic field can just tune the dielectric constant in a small range. The results may be used for designing dielectric detection devices in microwave communication. The dielectric constant and relative dielectric tunability of BST in 300 K versus the external electric field are given in figures 3(*b*) and (*c*). The small-signal dielectric constant under a magnetic field of 200 kA m^{-1} is almost 2000 higher than a zero magnetic field. With the increase in the external electric field, the dielectric constant shows little change with the change in the magnetic field. It indicates that the magnetic-field-induced strain is weak compared with the electrostrictive strain under a large electric field. Besides, the thickness ratio can also tune the dielectric constant. The thicker the terfenol-D, the stronger the constraint applied on the BST layer. To reach proper dielectric properties of BST with the determined Ba/Sr ratio under ambient temperature, we should carefully design the relative thickness of each layer to induce a proper constraint or tensile strain.

4. Conclusions

We studied the dielectric properties of BST in a Terfenol-D/BST/Terfenol-D trilayer system. In order to reach a large dielectric constant and dielectric tunability, we use an external magnetic field to turn the stress applied on the BST layer.

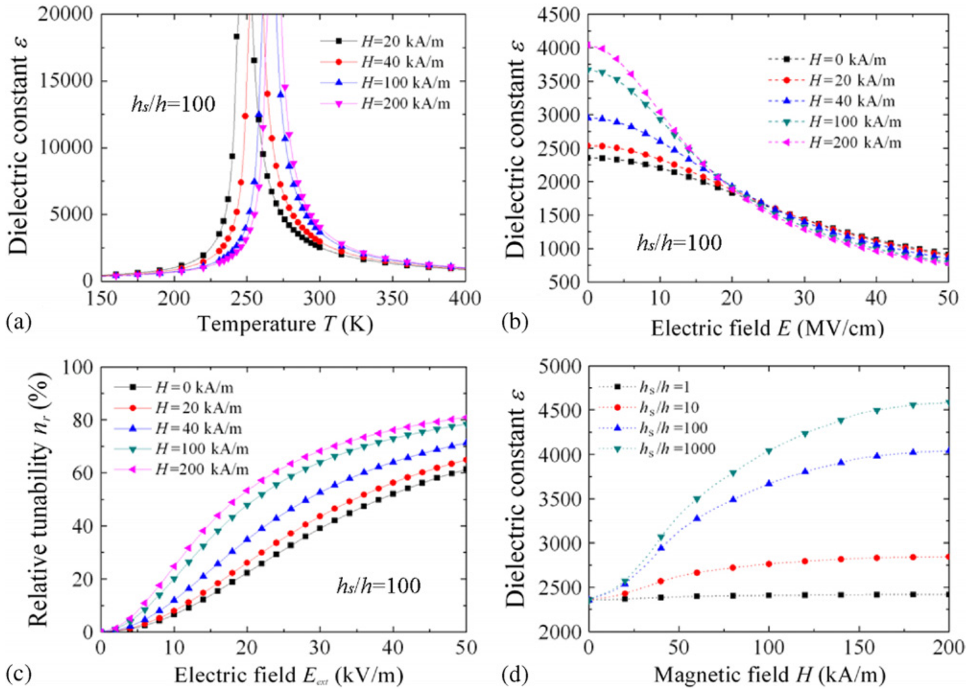

Figure 3. Dielectric properties of $Ba_{0.6}Sr_{0.4}TiO_3$: (*a*) dielectric constant versus the temperature under different external magnetic fields; (*b*) and (*c*) dielectric constant and relative dielectric constant versus the external electric field under different external magnetic fields at 300 K; (*d*) dielectric constant versus the external magnetic field under different thickness ratios at 300 K.

The use of the magnetic field is more convenient than other methods such as the change in the composition, the ratio of the thickness layer and the substrate. Using such a trilayer system, the dielectric constant of BST can be tuned ranging from about 2300 to 4000 by applied magnetic fields of 0–200 kA m^{-1}.

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