Giant Piezoresistance Effect of Asymmetric Ferroelectric Tunnel Junction: a Monte Carlo study

Kangshi Zeng¹, Xinpei Guo², Zhen Xu³ and Tianbai Xiong⁴a

¹School of Science, Xihua University, Chengdu 610039, China
²Monash University, Caulfield East VIC 3145, Australia
³Nanjing Forestry University, Nanjing 210037, China
⁴School of Materials and Energy, Guangdong University of Technology, Guangzhou 510006, China

aCorresponding author: xtb982xtb@163.com

Abstract. Using two-dimensional Ising model and Monte Carlo simulation method, the giant piezoresistance effect (GPE) of ferroelectric tunnel junction (FTJ) is studied. Under different axial pressures, the changes of polarization for different thin ferroelectrics film were simulated. The relationship between pressure and resistance of asymmetric FTJ was numerically investigated. Using this relationship, the giant piezoelectric resistance effect of asymmetric FTJ under different thickness was calculated and discussed. The resistance of FTJ increased at least with an order of magnitude, under the condition of 5 GPa pressure.

1. Introduction

Ferroelectrics have attracted much attention from academia and industry since the discovery of ferroelectricity in Rochelle salt crystal at the early 1920s [1]. With the development of preparation technology, thin ferroelectric films have been successfully synthesized. Their thickness can be in nanoscale, or even in the scale of several unit cells [2-6]. In 2005, Kohlsted proposed the concept of ferroelectric tunnel junction (FTJ) [6]. The FTJ has three parts, in which a nanometer-thick insulating ferroelectric film (barrier part) is sandwiched by two metal electrodes, as shown in Fig. 1(a). Due to the quantum tunneling effect, it shows many interesting electronic properties, such as giant electroresistance [7,8], larger resistance on/off ratio [9]. Hence, FTJ is considered a potential candidate for employing in nonvolatile memories [10], logic devices [11], etc.

Recently, giant piezoresistance effect (GPE) in asymmetry FTJ is proposed [12]. The electroresistance can change by an order of magnitude due to external mechanical loads [13]. Two mechanisms which can give rise to GPE are reported. First, because the different directions of polarization in FTJ exhibit different conductivity, the electroresistance is significantly varied as the polarization of ferroelectric can be reversed with pressure applied. This process is known as reversal mechanism, and it has been confirmed by several experimental studies [14-16]. Second, it has been reported that mechanical loads can induce paraelectric-ferroelectric structural transition in the barrier part. As the potential barrier of ferroelectrics phase is higher than that of paraelectric phase, the electroresistance of FTJ is also changed. This mechanism called phase-transition mechanism is explained by Ginzburg-Landau model [12] and first-principle calculations [13]. Comparing the two mechanisms, the latter is more common, because the barrier part is usually paraelectric at room
temperature when the quantum tunneling effect is evident. Although the phase-transition mechanism has been widely studied, an explanation of this mechanism in the framework of many-body model has not been reported.

Figure 1. (a) is the structure of FTJ, (b) is the schematic representation of two-dimensional Ising model.

In present study, the GPE in asymmetry FTJ is verified by Monte Carlo simulation. The paper is organized as follows. Firstly, the current-voltage relationship for tunnel junction is briefly introduced, and how external mechanical loads influence the conductivity of FTJ is explained. Then, the Ising-type Hamiltonian of ferroelectric and the details of simulation are presented. At last, the results are showed, and a brief discussion is made.

2. Methodology

2.1. Tunneling electroresistance for FTJ

Tunneling effect is a quantum phenomenon that microparticles, e.g., electrons, can pass through a potential barrier, even the height of the barrier is much greater than the total energy of the particles. The transmission probability is related not only to the shape, but also to the thickness of the barrier (along x-direction) [17]:

\[ D(E) = \exp \left\{ -\frac{4\pi}{h} \int_{t}^{m} \frac{2m\Phi(x) - E}{h} dx \right\}. \]  

(1)

where \( \Phi(x) \), \( E \), \( t \), and \( m \) are the barrier function, energy of particle, thickness of barrier, and mass of particle, respectively. \( h \) is the Planck constant. In the case of FTJ, the barrier for electron in short-circuit boundary condition is [6]

\[ \Phi(x) = U + \frac{\delta t - (\delta_1 + \delta_2)}{\epsilon_0 [\epsilon_3 (\delta_1 + \delta_2) + t]} eP, \quad 0 \leq x \leq t, \]  

(2)

where \( \delta_1 \) and \( \delta_2 \) are the screening lengths of the electrodes. If \( \delta_1 = \delta_2 \), the FTJ is symmetric, otherwise FTJ is asymmetric. \( P \) is polarization of ferroelectric, \( U \) is the difference between the bottom of the conduction band for ferroelectric and Fermi energy of electrode. As \( \Phi(x) \) in Eq. (2) is linear, this barrier can be equivalent to a rectangular one. The height of such rectangular barrier is

\[ \Phi = U + \frac{t(\delta_1 - \delta_2)}{2\epsilon_0 [\epsilon_3 (\delta_1 + \delta_2) + t]} eP. \]  

(3)

From Eq. (3), for symmetric FTJ, \( \Phi \) is not related to \( P \). Since the polarization is sensitive to external loadings, for asymmetric FTJ, \( \Phi \) also sensitive to the loadings. That is why GPE usually exists in asymmetric FTJ.

The current-voltage relationship for a tunnel junction for rectangular barrier has the form [17]:

\[ I = \frac{2e}{h} \sqrt{\frac{\pi m}{\hbar^2}} D(E) \]
\[
J = \frac{6.2 \times 10^6}{t^2} \left\{ (\Phi - \frac{V}{2}) \exp[-0.672 \ln(\Phi - \frac{V}{2})^{0.5}] - (\Phi + \frac{V}{2}) \exp[-0.672 \ln(\Phi + \frac{V}{2})^{0.5}] \right\}.
\]

where \( t \) is the thickness of the barrier. The units of \( J \), \( t \), \( \Phi \), and \( V \) are \( \text{A cm} \), \( \text{Å} \), eV, and V, respectively. Then, the electroresistance is

\[
R = \frac{V}{JS}.
\]

When the FTJ is subjected to external loadings (Fig. 1), \( \Phi \) is varied accordingly. For small deformation (strain \( \varepsilon_x \)), \( U \) and \( t \) are linear change

\[
U = U_0 + K \varepsilon_x,
\]

\[
t = t_0 (1 + \varepsilon_x)
\]

In general, \( P \) is also change linearly. However, when the loading induce phase transition, the polarization-strain relationship cannot be expressed analytically. This relation can be numerically derived by means of simulation.

2.2 Hamiltonian for ferroelectrics and details of simulation

For materials with ferroelectric domain structure, the behaviors of ferroelectric domain coupling can be described by a semi-classical model, i.e., Ising model, which is a simplified Heisenberg model. The Hamiltonian has the form:

\[
H = -J \sum_{\langle i,j \rangle} S_i S_j - \mu E \sum_i S_i,
\]

where \( J \) and \( E \) are the exchange coupling constant and external electric field. \( S \) is pseudo spin (-1 or 1), and \( \mu \) is the moment of the pseudo spin. \( \sum_{\langle i,j \rangle} \) denotes the summation over all the neighbor pseudo spin.

For several-unit-cell-thick material, two-dimensional Ising model is adopted, as the schematic representation in Fig. 1(b). When the material is subjected to external loadings, Eq. (7) should be modified as [18,19]:

\[
H = -n J e^{-aF} \sum_{\langle i,j \rangle} S_i S_j - \mu E \sum_i S_i,
\]

where \( n \) is the amount of unit cell along thickness (or \( x \) direction), so that the thickness \( t = n a_0 \), where \( a_0 \) is lattice constant. \( F \) is the loading along \( z \) direction (negative and positive value denote pressure and tension, respectively). According to constitutive relation, it has \( \varepsilon_x = S_w F \), where \( S_w \) is compliance constant.

The polarization of the materials is

\[
P = P_g \langle S \rangle.
\]

where \( P_g \) is the polarization of domain, and \( \langle S \rangle \) denotes thermodynamic statistical mean for pseudo spin.

The Metropolis Monte Carlo algorithm is applied to numerically obtain \( \langle S \rangle \) with certain temperature and loading. For low-voltage conductivity, the electric field related term in Eq. (8) is omitted. The details of the simulation are shown as follows.

(a) Create an 100*100 matrix which represents all the domains (in Matlab), randomly give the value of pseudo spin for each matrix element (-1 or 1), and calculate the Hamiltonian of this system.
(b) Change a value of matrix element to its opposite, and calculate the difference between the Hamiltonian $\Delta H$ of new system (matrix) and former one. The transition probability is

$$p = \begin{cases} e^{\frac{-\Delta H}{kT}}, & \Delta H \geq 0, \\ 1, & \Delta H < 0. \end{cases}$$  \hspace{1cm} (10)$$

where $T$ is thermodynamic temperature.

(c) Create a random number $r$ in interval $[0,1]$. When $rp \leq 1$, the new system is permissible; otherwise, the new system is not allowed.

(d) Repeat steps (a)-(c) for other spin until all the spins have been traversed, and save the new matrix. A Monte Carlo step is completed.

(e) Run the Monte Carlo step 1000 times, output $\langle S \rangle$ for the latest matrix.

After completing above steps, the polarization-strain relationship can be obtained. According to Eqs. (3)-(6), electroresistance-pressure relationship can be numerically derived.

The related parameters (take PbTiO$_3$ for instance) for the simulation and calculation are summarized in Table 1.

Table 1. The related parameters.

| Parameter | Value |
|-----------|-------|
| $\delta_1$ (nm) | 0.07 |
| $\delta_2$ (nm) | 0 |
| $S_{sc}$ ($\mu m^2/N$) | -6.5 |
| $\epsilon_r$ | 200 |
| $a_0$ (nm) | 0.45 |
| $U_0$ (eV) | 0.7 |
| $J_0$ (meV) | 1.5 |
| $a$ ($m^2/N$) | 0.6 |
| $K$ (eV) | -4.5 |
| $P_r$ ($\mu C/cm^2$) | 70 |
| $T$ (K) | 300 |

3. Results and discussion

Figure 2 shows the pressure-polarization relationships for four, three, and two-unit-cells-thick ferroelectrics.

Figure 2 shows the pressure-polarization relationships for different thickness of barrier part (ferroelectric). When pressure is raising from 2 to 3.5GPa for $n=2$, from 1 to 2.5GPa for $n=3$, and from 0.5 to 2GPa for $n=4$, the polarization is increasing rapidly and the phase transition occur. According to Eq. (8), the exchange coupling of two neighbor pseudo spins is proportional to $n$. Thus, the orientations of pseudo spins are easier to be consistent for $n=4$ in order to keep to a minimum Hamiltonian. This result is consistent with Ref. [13].

Based on these pressure-polarization relationships, according to Eq (3), the pressure-barrier height relationships can be derived, as shown in Fig.3. In phase transition region, the barrier height quickly increases when increasing pressure. By contrast, in outside of phase transition region, the barrier height is slightly decreased with increase of pressure. This can be explained as follows. Equation (3) indicates that the change of barrier height is positively correlated with polarization, barrier thickness, and $U$. In phase transition region, because the pressure induces the polarization, the barrier height
rises rapidly. While in outside of phase transition region, the polarization is not sensitive to pressure. It means the barrier height is mainly governed by barrier thickness and $U$. As the pressure thicken the barrier, the thickness effect always increases barrier height. On the other hand, the pressure reduces the energy of bottom of the conduction band, which results in lower $U$. Since the barrier height decreases when increasing pressure in this region, the reduction of barrier height cause by lower $U$ exceeds the increase of barrier height cause by thicken barrier.

\[
R_{GPR} = \frac{R_f - R_0}{R_0},
\]

Figure 3. The pressure-barrier height relationships for four, three, and two-unit-cells-thick ferroelectrics.

Once pressure-barrier height relationships are determined, the pressure-electroresistance relationships can be derived according to Eqs. (4)-(6). To exhibit the effect of giant piezoresistance, a variable called giant piezoresistance ratio (GPR) is defined as

\[
GPR = \frac{R_f - R_0}{R_0},
\]

Where $R_0$ and $R_f$ denote the electroresistances of initial and pressure state, respectively. The pressure-GPR relationships for different thickness of asymmetric FTJs are shown in Fig. 4.

From Fig. 4, the GPE is evident and GPR is very sensitive to the thickness of ferroelectric. When $F=-5\text{GPa}$, the value of GPR is approximately equal to 255% for $n=2$, and 909% for $n=3$. In contrast to them, the GPR for $n=4$ reaches 3568% at $F=-5\text{GPa}$. Moreover, the GPR for thicker FTJ is more sensitive than that for thinner FTJ. This result can be explained as follows. According to Eq. (3), the height of barrier is proportional to the thickness $t$ multiply by polarization $P$. When the polarization increased, the thicker the ferroelectric is, the more increment for height of the barrier would be. Indeed, Fig. 3 has exhibited such trend. So that the electroresistance increased faster, which result in high value of GPR for thicker FTJ.

Although thick FTJ exhibits superior sensitivity to pressure, the electroresistance is extremely high. For $n=4$, $R_0 \approx 2.7 \times 10^7 \Omega$. Therefore, while fabricating the FTJ based device, a proper thickness of FTJ, for which it possesses both high sensitivity and suitable conductivity, should be carefully considered.

Figure 4. The pressure-GPR relationships for asymmetric FTJs, where the ferroelectrics are four, three, and two-unit-cells-thick, respectively.
4. Conclusion
The GPE in asymmetric FTJ has been studied by using two-dimensional Ising model and Metropolis Monte Carlo algorithm. The electric barrier is related to the polarization as well as to the thickness of barrier in asymmetric FTJ. The Monte Carlo simulation is taken to acquire the pressure-polarization and pressure-barrier height relationships for different thickness of FTJs at room temperature. Based on these relationships, the pressure-GPR relationships are numerically expressed. The results show that the thicker FTJ possesses more significant GPE.

References
[1] Valasek, J. "Piezo-electric and allied phenomena in Rochelle salt." Physical review 17 (1921).
[2] Setter N, Damjanovic D, Eng L, et al. "Ferroelectric thin films: Review of materials, properties, and applications." Journal of Applied Physics 100 (2006).
[3] Wang B, Woo C H. "Curie temperature and critical thickness of ferroelectric thin films." Journal of applied physics 97 (2005).
[4] Tybell T, Ahn C H, Triscone J M. "Ferroelectricity in thin perovskite films." Applied physics letters 75 (1999).
[5] Cai M Q, Zheng Y, Ma P W, et al. "Vanishing critical thickness in asymmetric ferroelectric tunnel junctions: First principle simulations." Journal of applied physics 109 (2011).
[6] Kohlstedt H, Pertsev N A, Contreras J R, et al. "Theoretical current-voltage characteristics of ferroelectric tunnel junctions." Physical Review B 72 (2005).
[7] Zhuravlev M Y, Sabirianov R F, Jaswal S S, et al. "Giant electroresistance in ferroelectric tunnel junctions." Physical Review Letters 94 (2005).
[8] Tao, L. L., Wang J. "Ferroelectricity and tunneling electroresistance effect in asymmetric ferroelectric tunnel junctions." Journal of Applied Physics 119 (2016).
[9] Gruverman A, Wu D, Lu H, et al. "Tunneling electroresistance effect in ferroelectric tunnel junctions at the nanoscale." Nano letters 9 (2009).
[10] Chanthbouala A, Crassous A, Garcia V, et al. "Solid-state memories based on ferroelectric tunnel junctions." Nature nanotechnology 7 (2012).
[11] Garcia V, Bibes M. "Ferroelectric tunnel junctions for information storage and processing." Nature communications 5 (2014).
[12] Zheng Y, Woo C H. "Giant piezoelectric resistance in ferroelectric tunnel junctions." Nanotechnology 20 (2009).
[13] Luo X, Wang B, Zheng Y. "Tunable tunneling electroresistance in ferroelectric tunnel junctions by mechanical loads." ACS nano 5 (2011).
[14] Gruverman A, Kolosov O, Hatano J, et al. "Domain structure and polarization reversal in ferroelectrics studied by atomic force microscopy." Journal of Vacuum Science & Technology B: Microelectronics and Nanometer Structures Processing, Measurement, and Phenomena 13 (1995).
[15] Gruverman A, Khokin A, Kingon A, et al. "Asymmetric nanoscale switching in ferroelectric thin films by scanning force microscopy." Applied Physics Letters 78 (2001).
[16] Paruch P, Posadas A B, Dawber M, et al. "Polarization switching using single-walled carbon nanotubes grown on epitaxial ferroelectric thin films." Applied Physics Letters 93 (2008).
[17] Simmons J G. "Generalized formula for the electric tunnel effect between similar electrodes separated by a thin insulating film." Journal of applied physics 34 (1963).
[18] Zhang L, Zhong W L, Kleemann W. "A study of the quantum effect in BaTiO3." Physics Letters A 276 (2000).
[19] Kim H J, Oh S H, Jang H M. "Thermodynamic theory of stress distribution in epitaxial Pb(Zr, Ti)O3 thin films." Applied physics letters 75 (1999).