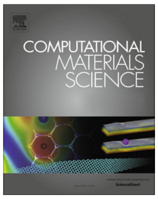
Computational Materials Science 115 (2016) 120–124

Contents lists available at ScienceDirect

Computational Materials Science

journal homepage: www.elsevier.com/locate/commatsci



Interface effect on leakage current of ferroelectric film

W.Q. Zhang, Q. Yang, Y.C. Zhou *, J.X. Cao

Key Laboratory of Low Dimensional Materials and Application Technology of Ministry of Education, School of Materials Science and Engineering, Xiangtan University, Xiangtan, Hunan 411105, China
Key Laboratory of Key Film Materials & Application for Equipment (Hunan Province), School of Materials Science and Engineering, Xiangtan University, Xiangtan, Hunan 411105, China

ARTICLE INFO

Article history:
Received 21 July 2015
Received in revised form 5 November 2015
Accepted 7 December 2015
Available online 2 February 2016

Keywords:
Interface effect
Leakage current
Ferroelectric film

ABSTRACT

The miniaturization trend of ferroelectric memory puts forward higher requirements on the quality of ferroelectric thin films. One of the most important factors which restrict the use of ferroelectric thin films is the leakage current. The electrode and substrate show significant effects on the film leakage, in which the interface has been considered to play an important role. In order to study the interface effect on the leakage current, the transport of electrode/$PbTiO_3$(PTO)/electrode ferroelectric capacitor was calculated by first principle method. Taking consideration of Pt, $LaNiO_3$ electrodes and the PbO- or TiO_2-terminated PTO thin film, we constructed four capacitor structures. The results indicated the current depends not only on the electrode types but also on the interface termination of film. On the whole, PTO thin film with oxide electrode has the greater current than that with metal electrode. Besides, PTO thin film with TiO_2-terminated interface shows a larger leakage current than that with PbO-terminated interface. The interface barriers for the four ferroelectric capacitors were also investigated to analyze the intrinsic mechanism of interface effects. It was founded that there is an increase of leakage current following the decrease of the potential barrier and current of PTO thin film obeys tunneling emission conduction mechanism. It suggests that the interface barrier is the important factor which determines leakage and can be used to tune the electrical properties of ferroelectric films.

1. Introduction

Ferroelectric materials are promising for a variety of technological applications such as actuators and sensors, nonvolatile ferroelectric memories, integrated capacitors, and ferroelectric tunnel junction [1–3], which have aroused great interest in recent years. However, some ferroelectric failures should be solved for the wide application of thin film. The electrical failures of ferroelectric film include fatigue [4–6], imprint [7] and retention loss [8–10]. There are many factors which result in the ferroelectric failures, such as depolarization, lattice defects, interface layer problem and leakage current. At the same time, the miniaturization trend of all devices puts forward higher requirements on the quality of ferroelectric thin films. When the thickness of ferroelectric film approaches nanometer-scale, leakage current becomes a key factor, which restricts the application of ferroelectric film. Leakage currents are supposed to be induced by ion drift and carrier conduction under the depolarization field, leading to the retention loss through the charge compensation of the ferroelectric polarization [11]. Leakage must be kept sufficiently low such that a capacitor does not discharge before it is refreshed. However, the control of leakage of a ferroelectric film is a challenging subject and its mechanism is under intense debate up to date [12–14]. Many classical conduction mechanisms, such as Schottky emissions, Poole–Frenkel (PL) emissions, Fowler–Nordheim (FN) tunneling, space charge limited current (SCLC) have been introduced to interpretate the leakage of ferroelectric film. The SCLC and PF emission are bulk limited conductions related to the trapped charges of the defects states. The Schottky emission and FN tunneling are interface limited conductions which are dominated by the interface barrier. Thus, the Schottky emission and FN tunneling can be recognized as the intrinsic conduction of a defect free dielectric film. Moreover, the current of an actual ferroelectric film is usually dominated by several mechanisms at the same time. A lot of factors, including electrode types [15], preparation temperature [13], the partial pressure of oxygen [16], annealing atmosphere [17,18] and temperature [19,20] have been found to show influences on the leakage current. Many efforts have been taken to tune the leakage current and improve electrical properties of ferroelectric films. Recently,

---

* Corresponding author at: Key Laboratory of Low Dimensional Materials and Application Technology of Ministry of Education, School of Materials Science and Engineering, Xiangtan University, Xiangtan, Hunan 411105, China.

E-mail addresses: zhouyc@xtu.edu.cn (Y.C. Zhou), jxcao@xtu.edu.cn (J.X. Cao).

http://dx.doi.org/10.1016/j.commatsci.2015.12.007

researchers have demonstrated that the leakage current, as well as other ferroelectric properties, such as polarization strength, fatigue and retention, are strongly dependent on the types of substrate and electrode used [21–23]. Especially, driven by the continuing miniaturization of electronic devices, the thickness of ferroelectric films will fall into nanoscale, which makes the interface effect more critical than before.

Many studies have been focused on the relationship between interfacial microstructure and leakage current of ferroelectric film by experimental approaches. Yan et al. [24] found that there are different orientations of $BiFeO_3$ (BFO) thin films deposited on different oxide bottom electrodes and the BFO film with the $LaNiO_3$ bottom electrode shows a lower leakage current in the electric field, <200 kV/cm, than that with the $SrRuO_3$ bottom electrode. Jang et al. [25] have reduced the leakage current of the thin film by introducing an antiferroelectric buffer into the ferroelectric capacitors. Moreover, leakage current and polarization properties of ferroelectric materials can be improved by tuning the miscut angles of substrate, which have been indicated by Shelke et al. [26]. Based on the existing researches, the heterointerface between the electrode and ferroelectric film shows important effect on leakage current. People even found that the interfacial termination of ferroelectric film also give rise to the electrical properties [27]. Though many works have been focused on the leakage current, a systematic study about the interface effect on the leakage of ferroelectric film is still lacking in the literature. Since the heterointerface between electrode and ferroelectric film is a kind of the most important interface structures in ferroelectric devices, in this paper, the leakage current of an ultrathin $PbTiO_3$ (PTO) film sandwiched between metallic or conductive oxide electrodes were systematically studied using the first-principle approaches. And the relationship between the interface barrier and leakage current was also analyzed to study the interface effects on the leakage properties of ferroelectric thin film.

2. Computational details

In the present work, we constructed a series of electrode/ferro electric/electrode capacitor structures. The calculations of atomic structural relaxation and interfacial potential barriers were performed using the Vienna Ab initio Simulation Package (VASP) [28] employing the projector-augmented-wave (PAW) [29] method within the local density approximation (LDA). For each electrode/fer roelectric/electrode system, a 1 × 1 supercell is stacked along the PTO [001] pseudocubic direction. A 500 eV plane-wave cutoff energy and a 6 × 6 × 1 Monkhorst–Pack k-point grid [30] were used for self-consistent calculations. Atomic relaxations were performed until the Hellmann–Feynman force on each atom was less than 5 meV/Å. The computational details are consistent with our previous study [31] and it is found that the lattice constant a and axial ratio c/a for the tetragonal PTO are 3.854 Å and 1.05, respectively, which are in reasonable agreement with the experimental data [32].

As for the calculation of leakage current, we used the ATOMISTIX TOOLKIT package (ATK 2008.10) [33], which combines non-equilibrium Green's function technique with the self-consistent pseudopotential method in the framework of DFT, to explore the electron transport properties of PTO thin film under different electrodes. In the calculation, exchange and correlation effects were accounted for using GGA-PBE and Monkhorst–Pack k-point grid within ab plane is 9 × 9.

3. Results and discussion

Four interface systems have been taken into consideration in order to study the interface affect on the transport current. Pt or $LaNiO_3$ electrodes, which contact with either the PbO- or TiO_2-terminated PTO, were constructed. In the Pt/PTO/Pt capacitors, Pt atoms are above the oxygen atoms on the TiO_2-terminated surface, and above Pb and O atoms on the PbO-terminated surface for the energy stability consideration [34]. While in the LNO/PTO/LNO capacitors, the interfacial atoms are continuously arranged according to the perovskite structure. These 1 × 1 supercells imply the films which are infinite in the x and y directions due to the periodic boundary condition. The supercells for the four types of interface systems can be described by the following general formulas:

(a) $Pt_4/PbO–(TiO_2–PbO)_8/Pt_4$,
(b) $Pt_4/TiO_2–(PbO–TiO_2)_8/Pt_4$,
(c) $NiO_2–(LaO–NiO_2)_2/PbO–(TiO_2–PbO)_8/(NiO_2–LaO)_2–NiO_2$,
(d) $LaO–(NiO_2–LaO)_2/TiO_2–(PbO–TiO_2)_8/(LaO–NiO_2)_2–LaO$.

Note that the thickness of PTO is 8.5 unit cell layers so that each structure studied in this work is symmetrical and has two identical film-electrode interfaces. And open-circuit boundary conditions are imposed with a 15 Å vacuum layer. In order to compare the effect of different interfaces, the in-plane lattices are all set to be the theoretical PTO value (i.e., $a = b = 3.854$ Å). Atomic positions are fully relaxed along the direction perpendicular to the interface. The relaxed structures are shown in Fig. 1, respectively.

In the transport calculation, we expanded the screening regions by repeating the two superficial atomic layers of the electrodes to ensure the accuracy of current. We have checked that the bias voltage potential is smooth enough between the central region and the electrodes. The voltage dependent current for the four capacitor structures were calculated. The tunneling current is calculated with the Landauer–Büttiker formula as follows [35,36]:

$$I = \frac{2e}{h}\int_{-\infty}^{\infty} T(\varepsilon)\{n_F(\varepsilon-\mu_R) - n_F(\varepsilon-\mu_L)\}d\varepsilon \qquad (1)$$

where μ_L and μ_R is the chemical potential of left and right electrodes, respectively, n_F is the Fermi distribution function, and $T(\varepsilon)$ is the transmission probability of electrons passing through the barrier. Fig. 2 shows the current as a function of the applied voltage bias from 0 V to 1 V for the above-mentioned four structures, respectively. It can be seen that the PTO film with TiO_2-terminated interface shows a larger leakage current than that with PbO-terminated interface for both the two types of electrodes. Based on our former study, the TiO_2-terminated PTO/electrode interface is the more possible in a PTO capacitor due to their higher interface adhesion [31]. So we considered that the current of PTO film is more likely to be dominated by the TiO_2-terminated case. From Fig. 2, we concluded that the PTO thin film with oxide electrode has the greater current than that with metal electrode, which is accordance with the experimental results [22,37]. The disadvantages of oxide/PZT/oxide capacitors are their relatively large leakage currents and high susceptibility to dielectric breakdown, which have been indicated by H. Han et al. [22]. One can also find that the leakage currents of PTO thin film in our calculation are quantitatively larger than experimental values [38]. This is because in our calculation, the thickness of PTO thin film is less than 5 nm and there is no defects in PTO film, which is different from the experimental conditions. The currents of $Pt_4/TiO_2–(PbO–TiO_2)_8/Pt_4$ system in our calculation is about three orders of the magnitude less than the ferroelectric junction, $Pt_5/TiO_2–(PbO–TiO_2)_4/Pt_4$, which has been researched by Luo et al. [39]. Generally, current of system not only depends on the electrode types but also on the interface termination of ferroelectric film thin films.

As we have discussed, the leakage current of system can be influenced by interfacial structure, such as different combinations of thin film and electrodes. Consequently, when the ferroelectric thin film is used in memory, we should take the choice of materials