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Full Length Article

Dry lubrication of friction on ferroelectric $BiFeO_3$ film

Chunbin Yang[a], Shuyu Xiao[a], Jan-Chi Yang[b,c], Xiaomei Lu[a,d,*], Ying-Hao Chu[b,c], Min Zhou[a], Fengzhen Huang[a,d], Jinsong Zhu[a]

[a] National Laboratory of Solid State Microstructures and Physics School, Nanjing University, 210093 Nanjing, People's Republic of China
[b] Department of Physics, National Cheng Kung University, 70101 Tainan, Taiwan
[c] Department of Materials Science and Engineering, National Chiao Tung University, Hsinchu 30010, Taiwan
[d] Collaborative Innovation Center of Advanced Microstructures, Nanjing University, 210093 Nanjing, People's Republic of China

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ABSTRACT

Epitaxial $BiFeO_3$ film shows excellent ferroelectric properties, which is not only studied extensively in magnetoelectric modulation but also shows good tribological performance. In our experiment, we find that the surface of $BiFeO_3$ film can be more lubricant with the application of bias voltages or stress through the AFM probe with a reduction (up to ~40%) of the friction between the sample and the probe tip. Based on the Prandtl-Tomlinson model, the mechanism of the dry lubrication on $BiFeO_3$ film originates from the reduction of van der Waals force between sample surface and probe tip. The findings here could be advantageously used in promoting the performance and durability of Micro/Nano Electromechanical devices based on ferroelectric thin films.

1. Introduction

With the miniaturization of Micro/Nano Electromechanical System (M/NES), the surface-volume ratio of M/NES devices have increased dramatically, and surface forces such as friction and adhesion have become critical factors in determining the performance and durability of M/NES devices. Therefore the frictional modulation of material surface becomes a critical issue [1]. Among various modulation manners, electronic control of friction has a charming perspective due to its convenience and controllability [2–6]. The tuning of electronic friction tuning was demonstrated in extrinsic silicon semiconductor through the application of forward and reverse bias voltages between the AFM (Atomic Force Microscopy) probe and the sample to control local carrier concentration in p and n regions [7]. The electronic friction tuning also has remarkable effect in polymer materials, and the on-off control of friction between polyelectrolyte-coated micas was realized under the alternating electric field while a liquid lubricating layer is necessary [8]. The molecular dynamics simulation of frictional contact between graphene and AFM probe tip, which, towards the dry friction of solid-solid interface, has revealed that the interaction between graphene and its silicon substrate has an impact on the deformation of graphene surface [9]. Imposing pre-compression produced wrinkles on graphene surface increases the dry friction between graphene and probe tip. Besides, the anisotropic friction of wrinkles suggests that a stronger interaction between graphene and the substrate leads to less deformation of the superficial carbon layer [10,11]. These reports indicate that the dry friction between materials could be tuned through stress engineering [12].

The dry friction between sample surface and AFM probe is closely related to the interaction between them, which is dominated by van der Waals force. Van der Waals force is a kind of electrical attraction among neutral molecules or atoms, consisting of three types [13–15], i.e. orientation force between permanent dipoles, induction force between permanent dipoles and induced dipoles, and dispersion force between instantaneous dipoles. As is well known, the permanent dipoles are constituted by polar molecules with separating centers of positive and negative charges; while induced-dipoles could form with nonpolar molecules being polarized by nearby permanent dipoles; still, instantaneous dipoles could arise in nonpolar molecules when the centers of positive and negative charges transiently separate with the motion of electrons, which might further induce new instantaneous dipoles in adjacent nonpolar molecules.

Normally, orientation force is stronger than induction force and dispersion force is the weakest among the three kinds of forces while the separation distance between interactive molecules is below 100 nm [13–16], which is larger than that between the sample surface and the probe tip in PFM and FFM [16]. The graphene is composed of nonpolar molecules, and the interaction between the graphene and the nonpolar AFM silicon probe is dominated by the dispersion force among instantaneous dipoles. In contrast, ferroelectric materials are constituted of polar molecules with spontaneous permanent dipoles [17], and the dominating interaction between ferroelectric materials and the silicon

* Corresponding author at: National Laboratory of Solid State Microstructures and Physics School, Nanjing University, 210093 Nanjing, People's Republic of China.
E-mail address: xiaomeil@nju.edu.cn (X. Lu).

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probe is the induction force. The polar molecules in ferroelectric materials generate a depolarization field that induces nonpolar molecules in the AFM probe [18,19], forming induced dipoles. Given that the induction force is stronger than dispersion force and the state of permanent dipoles in ferroelectric materials can be easily tuned by electric field or applied stress, we expect that the tribological modulation effect of ferroelectric materials could be greater than that of nonpolar systems. However, up to now, there are few reports regarding tribological properties of ferroelectric materials [20], especially in the thin film form.

Here, we choose $BiFeO_3$ (BFO) ferroelectric thin film as the sample material [21–25]. BFO is a single-phase multiferroic material, which is of rhombohedral perovskite structure (*R3c* space group) at room temperature with lattice parameters: $a = b = c = 5.633$ Å, $\alpha = \beta = \gamma = 59.4°$ [26–28]. Compared with the cubic perovskite structure in high-temperature paraelectric phase, the Bi ions move along the ⟨1 1 1⟩ body diagonal relative to the Fe-O octahedrons and the O octahedrons distort along ⟨1 1 1⟩ diagonal, forming permanent dipoles along ⟨1 1 1⟩ diagonal. Under an external electric field, three types of polar flips (71°, 109° and 180°) may occur. As BFO is an unique material that exhibit both ferroelectric and antiferromagnetic properties at room temperature with Curie temperature and Néel temperature being 830 °C and 370 °C, respectively, it has potential applications in micro/nano integrated devices. In this study, by applying bias voltages and compressive stress to the epitaxial (0 0 1) BFO film, the friction between the film surface and the AFM probe is reduced by up to ~40%, which, according to the Prandtl-Tomlinson model [29–33], can be predominantly attributed to the change of van der Waals interaction. Our work provides novel pathways for tuning dry friction lubrication in M/NES.

2. Experimental

A ~120 nm thick $SrRuO_3$ (SRO) conductive layer, which is used as the bottom electrode for the BFO film, is deposited on the (0 0 1) $SrTiO_3$ (STO) single crystal by pulsed laser deposition (PLD). Then the ~30 nm thick epitaxial BFO film is deposited over the SRO conductive layer (Fig. 1a) and the initial out-of-plane (OP) polarization of the BFO film points downwards [34,35]. The roughness, which is the root mean square of the morphology signal here, of the BFO film is ~0.5 nm calculated by the Nanoscope Analysis software (Fig. 1b). Using the AFM (Bruker, Icon), we apply bias voltages and compressive stress to the BFO thin film through the Pt/Ir-coated conductive silicon probe (SCM-PIT, Bruker), and the SRO conductive layer remains grounded.

The ferroelectric domain distribution of the sample is characterized under the PFM (Piezoresponse Force Microscopy) mode of AFM, with the scan rate, drive amplitude and drive frequency being 1 Hz, 6000 mV and 30 kHz, respectively. For polar scan with a scan rate of 3 Hz, direct current (DC) voltages ranging from ± 5 V to ± 9 V are applied to the AFM probe. The friction between the sample surface and the probe is measured under the FFM (Friction Force Microscopy) mode at a scan rate of 1 Hz. During FFM measurement, the fast scan direction of the probe is perpendicular to the main axis of the cantilever (Fig. 1), while the friction from the sample surface and the probe causes the probe to twist in the transverse direction, and the friction image is just composed of the FFM transverse torsion signal. Generally, the FFM transverse torsion signal is disturbed by the morphology signal, while the crosstalk could be reduced by subtracting the signals of reverse friction from forward friction and the value of friction signal is equal to half of the difference [36–38]. Thus, the small variation of sample morphology has little impact on the friction image in FFM. Besides, for comparing the influence of external fields on friction, we define the reduction of friction as $\Delta F = (F_{Out} - F_{In})/F_{Out}$, where F_{In} is the mean value of friction in the area treated by external fields and F_{Out} is that in the untreated area.

3. Results and discussion

3.1. Negative-biased scan

First, we choose a 3 μm × 3 μm region in the BFO film and conduct polar scan with the AFM probe. The bias voltage of the probe ranges from −5 V to −9 V and the SRO conductive layer remains grounded. Subsequently, piezoresponse and friction detection are performed in a larger 5 μm × 5 μm region (Fig. 2). At −5 V, there is almost no observable change on the sample surface; when the voltage reaches −6 V, we can see from the corresponding PFM OP (out-of-plane) phase image that the OP polarization of the polar-scanned region starts to reverse, while the friction noticeably decreases (Fig. 2a) by approximately 12% (Fig. 2b); when the voltage reaches −7 V, the friction reduce further by ~35% (Fig. 2b). As the voltage continues to decrease, the friction reduction stays almost constant.

3.2. Prandtl-Tomlinson model

Generally, the friction reduction could be attributed to the change of the interaction energy between contacting materials, and the relationship between them can be well understood by the Prandtl-Tomlinson (P-T) model [29]. Compared to more complex models and molecular dynamics simulations [29–32], the P-T model is equally effective but simpler, which, as currently the most influential minimalistic model for nanoscale tribology, can nicely explain the lateral force generated during the AFM probe scanning process.

The P-T model is abstractly described as the movement of a probe in a periodic potential representing the interaction between the sample surface and probe (Fig. 3). For a one-dimensional system, the total energy can be described as follows:

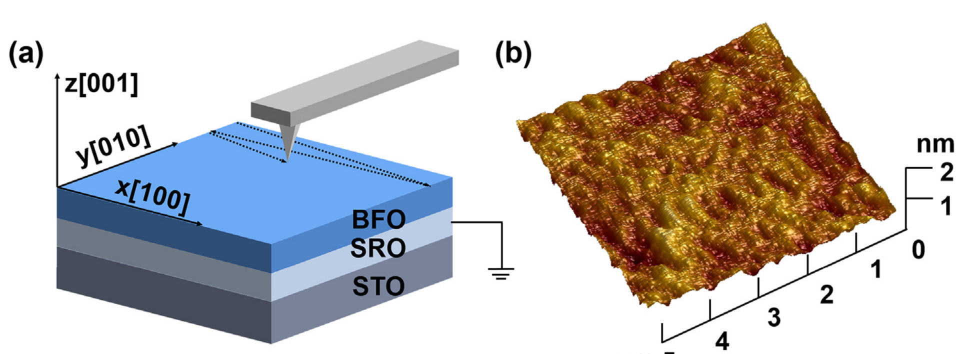

Fig. 1. (a) Schematic of AFM characterization. Bias voltages and compressive stress are applied to the surface of the BFO film through the AFM probe, and the SRO conductive layer remains grounded. (b) The 3D morphology image of the BFO film surface.