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Effects of light on ferroelectric polarization and leakage current

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ABSTRACT

We report the enhancement of polarization in polycrystalline ferroelectric thin films under illumination of light. The $(Pb_{0.6}Li_{0.2}Bi_{0.2})(Zr_{0.2}Ti_{0.8})O_3$ (PLBZT) thin films were fabricated on a $Pt/TiO_2/SiO_2/Si$ (100) substrate by pulsed-laser deposition (PLD) technique. The illumination of weak monochromatic light having a wavelength comparable to the bandgap of PLBZT showed the development of non-equilibrium charge carriers which enhance switchable polarization and displacement current. A Positive-up Negative-down (PUND) analysis also supports the enhancement of switchable polarization under illumination. The fatigue test indicates nearly 20–30% decrease in polarization after a long time write and read cycles. These results may provide an extra degree of freedom to create electrical WRITE and optical READ logic states.

1. Introduction

Ferroelectric (FE) materials retain permanent polarization after removal of an applied electric field. It shows two thermodynamically stable polarization states defined by their remanent polarization ($\pm P_R$). FE polarization states reverse its direction from $+P_R$ to $-P_R$ and vice-versa by an applied external electric field larger than the coercive field. These permanent polarization states have been used as memory logics (i.e., 0 & 1) to design a nonvolatile ferroelectric random access memory (NV-FeRAM) element [1]. In general, the memory cell consists of one transistor and one ferroelectric capacitor (1T-1C), i.e. ferroelectric capacitor stores the data and a transistor used to address the selected cell. The general requirements of NV-FeRAM cells are low operating voltage, high endurance, long retention time, and little fatigue with conventional electrodes. The presence of two stable spontaneous polarization states makes ferroelectric thin films useful in the area of NV-FeRAM, piezoelectric transducers, optoelectronic devices, and other numerous electronic circuits [2,3].

The photo-ferroelectricity has been extensively studied since the discovery of ferroelectricity which is defined as the change in ferroelectric, dielectric, conductivity properties, control of domains and domain walls under illumination of light [4–6]. Extensive research works on photo-ferroelectricity have been carried out for out-of-plane polarization configuration, especially to understand the domain dynamics, domain walls, above-bandgap photovoltaic effects, and polarization manipulation under illumination of weak light. However, a systematic in-depth study is still required at a microscopic level to understand the domain dynamics and their behavior under illumination of light conditions to commercialize the device aspect of optically active ferroelectric thin films [7]. Photo-sensitive ferroelectric materials change their polarization states under illumination of light which may be utilized to fabricate opto-ferroelectric memory elements, photovoltaics, optical sensors and optoelectronics devices [8,9]. Hu et al. recently illustrated manipulation of electro-resistance under illumination of light which may provide a multi-state ferroelectric tunnel junction [10].

We have observed a significant enhancement in polarization under illumination of light and reproducible changes of polarization on PLBZT bulk polycrystalline ceramics [11]. In the presence of light, the electronic polarization changes the magnitude and direction of ionic polarization due to a displacement between central positive cations and oxygen octahedral anions [12]. The light stimulated free charge carriers to alter the displacement current and polarization. The light also modified trap levels, fills holes, and compensates free charge carriers in polycrystalline grains [13]. Many oxides have shown the capability to store information digitally which can also be controlled optically [14].

Among functional oxides, ferroelectric materials exhibit

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spontaneous polarizations and their control by illumination of light. We report the effects of light on the polarization and leakage current behavior of polycrystalline PLBZT thin films grown on platinized silicon substrate.

2. Experimental details

We performed polarization, displacement current, PUND, and electrical measurements under dark and light conditions to realize various logic states. The ferroelectric ceramic target of PLBZT was prepared by a conventional solid-state reaction route. The details of target preparation process, calcination, and sintering temperature are reported elsewhere.[9] In brief, the compositional green powder of PbO, Li_2CO_3, Bi_2O_3, ZrO_2, and TiO_2 (procured from Alfa Aesar, purity ~ 99.9%) precursors oxides was calcined at 800 °C for 10 h. Later a one-inch ceramic target was sintered at 1200 °C in the air for 4 h for pulsed laser ablation. The PLBZT films on Pt (150 nm)/TiO_2 (40 nm)/SiO_2/Si (0.5 mm) substrate were grown using pulsed laser deposition (PLD) technique. The platinized silicon substrates were first cleaned using ultrasonication in acetone for 2 min and later in methanol for 10 min to remove impurities present on the substrate. The polycrystalline PLBZT thin films of average thickness 200 nm were deposited on a platinized silicon substrate using a KrF Excimer laser (wavelength, 248 nm; frequency, 5 Hz) with an average energy density of 1.5 J/cm^3 at 700 °C temperature, and 0.10 mbar oxygen pressure. These films were further annealed in-situ in pure oxygen ambiance at 250 mbar and 700 °C for 30 min. The detailed crystal structure and surface morphology have been carried out using X-ray diffractometer (Bruker AXSD8), and atomic force microscopy (AFM), respectively. Radiant ferroelectric tester is used for polarization and current-voltage study at room temperature [15].

3. Results and discussion

3.1. Crystal structure and surface topography

The XRD peaks as shown in Fig. 1 is indexed successfully using tetragonal crystal structure with *P4mm* space group which matched with bulk polycrystalline ceramics. [9] The possible development of strain in thin films was calculated by using the equation:

$$\varepsilon = \frac{\alpha_{bulk} - \alpha_{film}}{\alpha_{film}} \times 100$$

where ε is the lattice strain, α_{bulk} is the lattice constant of the bulk, and α_{film} is the lattice constant of the film. The lattice parameters of thin films were compared with lattice constants of bulk ceramics, and it was found that there was nearly 1.5% tensile and compressive strain along a-lattice parameter (in-plane) and c-lattice parameter (out-of-plane), respectively. Inset of Fig. 1 shows the formation of spherical grains with average grain size in the range of 15–30 nm with an average surface roughness of 2–4 nm. A dense and granular microstructure can be seen in almost every part of the film under investigation.

The graphical presentation for out-of-plane ferroelectric polarization measurement set-up under dark and light conditions is shown in Fig. 2(a). Semi-transparent gold (Au) electrodes were deposited on the top surface of PLBZT thin films. We have not used transparent ITO thin films for top electrode due to its large contact/surface resistance (~50–100 ohm), high-temperature growth and annealing conditions, and possible diffusion across the electrode-film interface. The electrical measurements were carried out for three different environmental conditions: dark, white light and laser light (wavelength (λ) = 405 nm). The power intensities of the light sources were 60 mW/cm^2 and 30 mW/cm^2, respectively. The set-up for illumination is made in such a way that maximum area can be exposed and generates more free charge carriers. Fig. 2 (b) & (c) illustrate polarization hysteresis loops measured at small probe pulse width (1 ms) under dark, white light and laser light, and their corresponding displacement currents. Well-saturated ferroelectric hysteresis loops were observed for all three conditions. However, we observed an enhancement in the remanent polarization nearly 28% and 100%, and saturation polarization nearly 12% and 50%, under white light and laser, respectively. A significant enhancement in displacement current with sharp displacement current peak near the coercive field was observed under illumination of light; the effects were more prominent with the monochromatic light with an energy comparable to the bandgap of the PLBZT. Similar effects were observed for a range of frequencies from 100 to 500 Hz (see Fig. 3 (a,b,c)). However, the effect of leakage current on polarization and displacement current for <1 kHz probe frequencies was significant. A large opening in most of the P-E loops can be seen in negative polarization switching, revealing that the effect of leakage current in total polarization is significant under illumination. However one cannot ignore the important change in

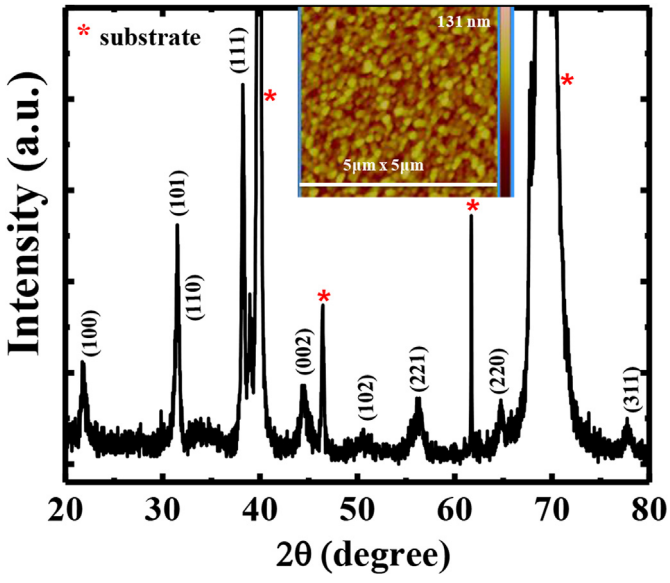

Fig. 1. X-ray diffraction patterns of PLBZT thin (* mark for Pt/Ti/SiO_2/Si substrates). The inset shows surface morphology of a 5 μm × 5 μm thin film with height ~131 nm.

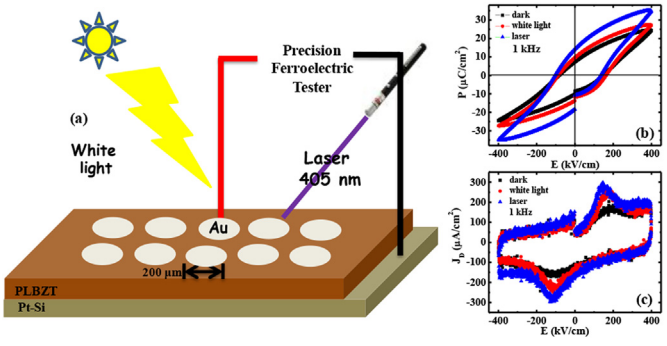

Fig. 2. (a) Shows a schematic illustration of the test-bed device structure for out-of-plane polarization measurements. The condition of generating of charge carriers under illumination of light and a top view of semi-transparent Au electrode deposited on the thin film. (b) Ferroelectric polarization under dark, white light, and laser light, measured at small probe pulse width (1 ms) (c) derived displacement currents for all three conditions.