Tunable Photonic Bandgap Devices Based on Pb-La-Zr-Ti-O/Sr-Ti-O Two Dimensional Multi-layers on GaAs Substrates

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Abstract - Multiple-layer perovskite $Pb_{0.91}La_{0.09}Zr_{0.65}Ti_{0.35}O_3/SrTiO_3$ (PLZT/STO) heterostructures have been grown by pulsed laser deposition (PLD) on (001) MgO/GaAs substrates. The perovskite layers are single phase with 35nm RMS roughness on the top surface. Electrically induced birefringence in the PLZT layers, resulted in both optical phase and optical amplitude modulation. The application of an electric field across the PLZT layers resulted in optical modulation at both the primary and first harmonic frequencies, and direct evidence of the non-linear electro-optic effect.

A. Introduction

The family of Pb-La-Zr-Ti-O (PLZT) perovskite oxide ceramics has been the focus of considerable research efforts due to their many unique properties [1,2,3] including ferroelectric and electro-optic effects. While other perovskite oxide materials have recently supplanted PLZT as the material-of-choice for many ferroelectric, piezoelectric and electrostrictive applications [1,2] PLZT ceramics continue to receive attention as a promising electro-optic oxide material. The large quadratic electro-optic coefficient, high optical transparency and extensive knowledge of basic material properties along with an established processing technology for PLZT continue to make it an attractive electro-optic material [2,3]. Electro-optical devices have been successfully fabricated from bulk PLZT [1,2].

Recent advances in pulsed laser deposition (PLD), sol gel processes and molecular beam epitaxy, along with the application of in-situ monitoring techniques to oxide film growth, has advanced the understanding of oxide epitaxy [2,3]. These developments promise to improve the quality and complexity of available thin film oxide heterostructures and devices. Considering the expected improvements in the epitaxial growth of perovskite, multilayers can be deposited where one of the layers can act as a chemical diffusion barrier between semiconductor and perovskite oxide materials

Multi-layer PLZT/BST structures are the simplest type of photonic crystal which can act as a perfect mirror for light with a frequency within a sharply defined gap. If such a structure has multiple defects, then light modes may also be localized. The PLZT/BST multi-layer structure is a one dimensional photonic crystal which refers to the fact that the dielectric is periodic only in one direction. Likewise, two dimensional photonic crystals have been fabricated in the present investigations which are periodic in two directions. These structures are based on the largest difference in the index of refraction between any two adjacent regions and require image reversal lithography in order to achieve a completely planar geometry.

B. Oxide growth and physical characterization

Perovskite heterostructures of LSCO/PLZT/LSCO grown on GaAs substrates, using an MgO buffer layer, have been previously reported for ferroelectric applications [1]. Similar techniques and deposition parameters were used to produce the oxide structures in the present investigation. All oxide layers were grown on (001) GaAs substrates using the pulsed laser ablation technique. A 20ns KrF excimer laser pulse was focused to an energy density of approximately 1-2J/cm^2 at the target surface. The target materials were polycrystalline PLZT, LSCO, STO and BST and single-crystal MgO. The oxygen ambient during ablation was maintained at 1x10^{-1} Torr for perovskite growth and 1x10^{-3} Torr for MgO growth. Substrate temperature was maintained at 650^{o}C for all perovskite layers and 500^{o}C for all MgO layers.

During growth optimization experiments, x-ray diffraction analysis was carried out to determine the resultant phase and crystallographic orientation. Optimized samples consisted of perovskite-phase PLZT, STO, BST and LSCO, on (001) oriented MgO buffer layer all grown on (100) GaAs semi-insulating substrates. The optimized heterostrutures exhibited undetectable levels of pyrochlore formation. LSCO oriented in the (001) direction was successfully deposited on the (001) MgO buffer layer, which resulted in (001) orientation for all further perovskite layers.

A transmission electron micrograph of a typical GaAs / MgO / LSCO/ PLZT / BST / PLZT heterostructure is shown in Figure 1, indicating columnar grains throughout the structure. X-ray diffraction data indicated that the layers in this sample are (001) oriented. This micrograph also indicates a significant amount of interfacial roughness (up to 30nm), which increases with increasing distance from the GaAs substrate. This effect was present in all samples examined, and was also confirmed with atomic force microscopy measurements.

B.1 Optical characterization

Electro-optic modulation measurements were performed on GaAs/MgO and GaAs/MgO/STO/PLZT/STO/PLZT oxide heterostructures. Interdigitated surface electrodes of Cr/Cu/Au were deposited and fabricated on the surface of these structures with semiconductor processing techniques. The electrode finger spacing was 50µm and each electrode finger had a 50µm width. The large aspect ratio of electrode spacing to film thickness assured that the applied electric fields were parallel to the film layers.

The modulation measurements were carried out using techniques described below. The 760nm laser beam was polarized and passed through a non-polarizing beam splitter. The light was focused onto the samples, between two electrode fingers, with a microscope objective. The application of a sinusoidal, 2 KHz electric field to the sample, at a 45^o angle to the incident light polarization vector, produced modulation in the reflected light. The reflected light was diverted by the non-polarizing beamsplitter into a polarizing beam splitter, oriented with the polarizing axes oriented at a 45^o angle to the incident light, and the two orthogonal polarizations were measured with photodiodes operating in photoconductive mode. An instrumentation amplifier subtracted the two photodiode signals. The photodiode differential signal was measured with a spectrum analyzer, and total peak intensity was normalized to the DC light level.

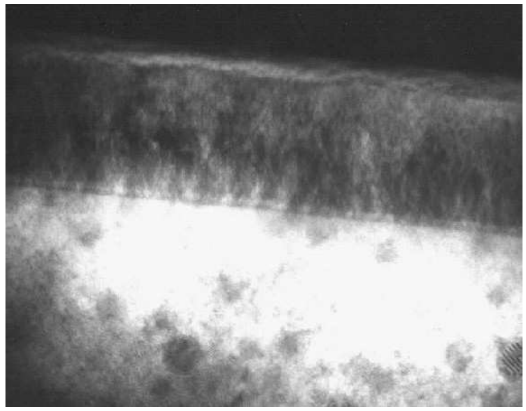

Figure 1. Transmission electron micrograph of GaAs/MgO/LSCO/PLZT/BST where the MgO layer is 25nm thick and the LSCO is 200nm thick.

The electro-optical measurements on single PLZT thin films, performed normally to the film surface, include a waveplate with fast axis parallel to the incident laser light: this experimental setup, including the waveplate, produces a measurement that is sensitive to both amplitude and phase differences between orthogonal polarizations incident on the sample. The amplitude and phase modulation as a function of electric field for a GaAs / MgO (70nm) structure was measured on all samples grown. The data shows that for a sinusoidal electric field, measurable modulation occurs at both primary and first harmonic frequencies. Modulation at higher harmonics was detected, but the total modulation harmonics above the first was less than 1% of the total modulation signal, and is not reported here. Similar investigations on the multi-layered structures were also carried out and show both amplitude and phase modulation for GaAs / MgO (40nm) / STO (70nm) / PLZT (95nm) / STO (70nm) / PLZT (95nm) structures under identical modulation conditions as that for single layer structures. The amplitude modulation has increased by a factor of 3 over that of the GaAs / MgO sample, while the phase modulation data is nearly identical. Modulation at the first harmonic frequency has also been observed. Likewise, the harmonic content in electro-optical modulation data reflects the nature of the electro-optic effect itself. The reflectivity of a dielectric interface between a dielectric and an electro-optic material is changed by the applied electric field. A simple calculation will show, to a first order approximation, that a single dielectric interface with a linear electro-optic material on one side will produce modulation only at the primary modulation frequency. A quadratic electro-optic effect will result in modulation at both primary and first harmonic frequencies, with negligible higher harmonics. The harmonic content in the modulation measurements may be explained by the reported electro- optic coefficients of GaAs and PLZT. The measurements of GaAs at a wavelength of 670nm (above the bandgap) indicate a linear electro-optic coefficient of 2.4e-11 m/V and a quadratic electro-optic coefficient of 1.4e-16 $m^2/V^{2..}$. Previous investigations of (9/65/35) PLZT indicate a quadratic electro-optic coefficient of approximately 1e-16 m^2/V^2, depending strongly on the thin film microstructure [1, 3]. Any room temperature electro-optic effects in MgO or STO are many orders of magnitude weaker than GaAs or PLZT, and may be considered as dielectrics for this study. Therefore, the electro-optic modulation in single layer structures likely arises from the GaAs substrate, and the enhanced modulation in multilayer structures is due to the added (9/65/35) PLZT layers.

The similarity in phase modulation data for both types of structures is due to several structural characteristics. The index of refraction difference between PLZT and STO is only 0.1, resulting in low interface reflectivities. In addition the PLZT and STO layer thickness were not optimized for interference at 670nm. In addition, the two PLZT layers in this structure was not sufficient to create significant optical resonance. Thus, the optical path length of the light through the PLZT is limited to several layer thickner, and the electrically-modulated phase shifts produced by the PLZT layers were not distinguishable from those produced by modulation of the GaAs/MgO interface characteristics.

The addition of two PLZT / STO interfaces substantially increased amplitude modulation. The addition of STO/PLZT/STO/PLZT to a GaAs/MgO structure increased the number of modulated interfaces from one to four. Since GaAs and (9/65/35) PLZT possess similar electro-optic coefficients at 670nm, it may be expected that an increase in modulated interfaces should produce a corresponding increase in amplitude modulation. The ratio of maximum amplitude modulation in multiplayer structures to that of single layer structures is 3.5. Note that the above interpretation of the amplitude modulation is valid only if there is not a significant contribution from the harmonic content of the signal. Additional PLZT layers should further increase amplitude modulation but not phase modulation.

C. Conclusions

Double-layer perovskite $Pb_{0.91}La_{0.09}Zr_{0.65}Ti_{0.35}O_3/SrTiO_3$ (PLZT) heterostructures have been grown by pulsed laser deposition (PLD) onto (001) MgO/GaAs substrates. The perovskite layers are single phase with 30nm rms roughness on the top surface. Electrically induced birefringence in the PLZT layers, applied through surface interdigitated electrodes, resulted in both optical phase and optical amplitude modulation. The application of a purely sinusoidal electric field across the PLZT layers resulted in optical modulation at both the primary and first harmonic frequencies, and direct evidence of the non-linear electro-optic effect.

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