

# Relaxor-like dielectric behavior of pulsed-laser-deposited $\text{Pb}_{0.5}\text{Sr}_{0.5}\text{TiO}_3$ films

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## Abstract

$\text{Pb}_{0.5}\text{Sr}_{0.5}\text{TiO}_3$  (PST) ferroelectric films were deposited onto Pt/Ti/SiO<sub>2</sub>/Si substrates by pulsed laser deposition. The state of the films can be described as a mixed state, with both ferroelectric and relaxor-like features. The films exhibited high dielectric constant and tunability at room temperature. At 10 kHz, the dielectric constants of the 200-nm- and 400-nm-thick films are 771 and 971, with the tunability of 60.2% and 70.9%, respectively. The temperature-dependent dielectric properties were studied and the relaxor-like behavior was observed in both the thinner and thicker PST films, which can be established in terms of diffuse phase transition characteristics and Vögel–Fulcher relationship. In addition, the contribution of the film–electrode interface layer to the dielectric properties was evaluated and the true dielectric properties of the films were reconstructed. Consequently, the relaxor-like character of the PST films was mainly ascribed to the effect of the film–electrode interfaces.

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## 1. Introduction

Ferroelectric materials have been applied to many electronic and optical devices, utilizing their excellent dielectric, piezoelectric and optical properties. Some of them exhibit relaxor behavior characterized by a broad maximum in the temperature dependence of dielectric constant and a strong frequency dispersion of the dielectric constant at temperature around and below the transition temperature. The dielectric constant and electrostrictive strain in most relaxors are high, making them attractive for the applications in capacitor and microelectromechanical systems [1,2].

Modern microdevices require high-performance materials in the form of films with thickness in the range of a few

hundred and even tens of nanometers. In recent years, there has been an increasing research activity to search appropriate thin film materials with a high dielectric constant and/or high dielectric constant tunability by d.c. voltage, for dynamic random access memories (DRAM) and microwave dielectric devices applications. Solid solution films of barium strontium titanate (BST) are actively being studied for the above purposes. However, a high growth temperature near 750 °C and large leakage current densities are some of the major obstacles for BST to be used in DRAM and tunable dielectric devices. Pure strontium titanate (ST) film with a cubic phase at room temperature shows lower crystallization temperature in comparison with BST films. However, its dielectric constant and tunability are lower than those of BST films. It has been reported that addition of lead titanate (PT) into ST films results not only in higher dielectric constant and tunability but also in a lower deposition temperature [3,4].

Nomura and Sawada investigated a series of lead strontium titanate ceramic samples and established that PT

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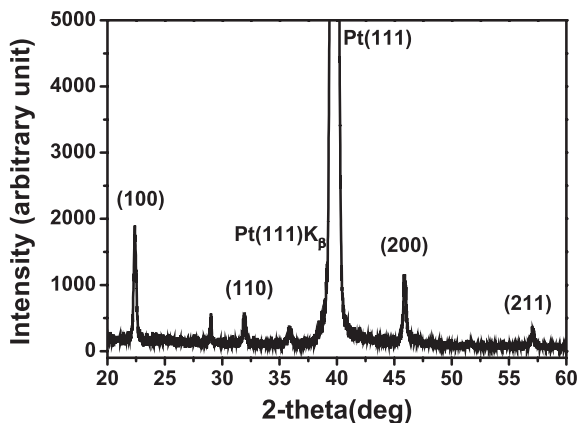


Fig. 1. XRD patterns of the 400-nm-thick PST films deposited on Pt/Ti/SiO<sub>2</sub>/Si substrates.

forms continuous ranges of solid solution with ST, and the Curie temperature decreases linearly with the increase of Sr content [5]. Subrahmanyam and Goo [6] have studied the diffuse phase transitions in lead strontium titanate ceramics and found that their diffuseness increases with increasing Sr content at first and has a maximum around the mid-composition range and then decreases. However this characteristic has only been reported for bulk lead strontium titanate compositions and there are few published data on the relaxor behavior of lead strontium titanate films. The present work is to study and understand the relaxor-like behavior of the thin films for the mid-compositional lead strontium titanate, Pb<sub>0.5</sub>Sr<sub>0.5</sub>TiO<sub>3</sub> (henceforth called as PST).

## 2. Experimental procedure

PST films were grown on Pt/Ti/SiO<sub>2</sub>/Si substrates by a 248-nm KrF excimer laser with the repetition rate of 5 Hz. The laser beam was focused on a PST ceramic target through a quartz window with the energy density of 2 J/cm<sup>2</sup>. A 10-mol% excess PbO was added into the powder mixture during the preparation of PST targets in order to compensate for the Pb loss. During the deposition, the substrate temperature was 650 °C and oxygen pressure was 20 Pa. For carrying out the electrical measurements, Pt dots of 0.28 mm in diameter were deposited onto the PST films at room temperature through a shadow mask by a radio-frequency sputtering technique. Films with thicknesses near 200 nm and 400 nm were selected for this study.

The structure of the films was analyzed by a Rigaku D/MAX 3C X-ray diffractometer with CuK<sub>α</sub> radiation at 40 kV. The surface morphology of the films was investigated using a Hitachi S-5750 scanning electron microscopy with an operating voltage of 20 kV. The thickness of the films was measured with an ET350 Talysurf profilometer (Kosaka Laboratory Ltd). Capacitance–Voltage (C–V) was measured with an impedance analyzer HP4294A at a 10 kHz

frequency. The temperature-dependence of the dielectric properties was measured by a HP4284 LCR meter from 40 °C to 220 °C in a computer-controlled Delta 9023 oven. Hysteresis loops of the films were recorded at room temperature using a Radiant RT6000S type ferroelectric tester in the virtual ground mode.

## 3. Results and discussion

The films with different thickness but prepared at the same conditions show similar microstructures. The typical microstructure of the PST film (400 nm in thickness) is shown in Fig. 1, which indicates a polycrystalline perovskite structure. The film presents textures with (100) and (200) as main components and lower contributions along (110) and (211). Fig. 2 illustrates the surface morphology of PST films on platinum substrate and shows a continuous and fine-grained microstructure without obvious cracks. Some of the big grains are present on the surfaces, which are actually the agglomeration of several small grains.

Fig. 3 shows the frequency dependence of the dielectric constant of the PST films with different thickness. It can be seen that the dielectric constant of the 400-nm-thick PST film was obviously higher than that of the 200-nm-thick film. At 10 kHz, the dielectric constants of the 200-nm- and 400-nm-thick films are 771 and 971, respectively, while their dielectric losses are 0.039 and 0.052, respectively. From Fig. 3 it can also be found that the dielectric constant

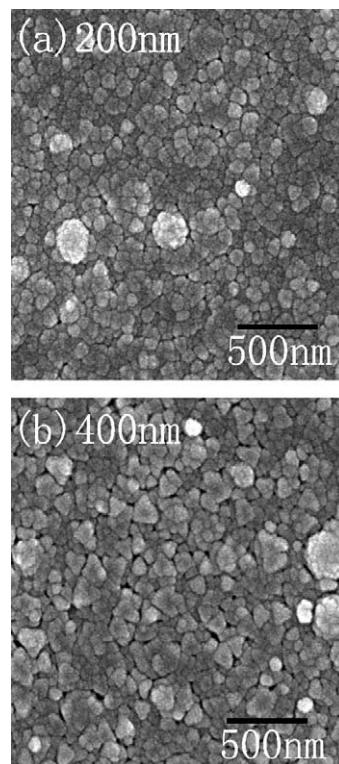


Fig. 2. Surface morphologies of PST films with thickness of (a) 200 nm and (b) 400 nm.

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