Optical properties of SrTiO3 thin films by pulsed laser deposition

1 National Laboratory of Solid State Microstructure and Center for Materials Analysis, Nanjing University, Nanjing 210 093, P.R. China
2 Department of Electrical Science and Engineering, Nanjing University, Nanjing 210 093, P.R. China

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ABSTRACT SrTiO3 thin films were prepared on a fused-quartz substrate by pulsed laser deposition (PLD). Dense and homogeneous films with a thickness of 260 nm were prepared. Optical constants (refractive index \( n \) and extinction coefficient \( k \)) were determined from the transmittance spectra using the envelope method. The optical band gap energy of the films was found to be 3.58 eV, higher than the 3.22 eV for bulk SrTiO3, attributable to the film stress exerted by the substrate. The dispersion relation of the refractive index vs. wavelength follows the single electronic oscillator model. The refractive index and the packing density for the PLD-prepared SrTiO3 thin films are higher than those for the SrTiO3 films prepared by physical vapor deposition, sol–gel and RF sputtering.

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1 Introduction

ABO	extsubscript{3}-type perovskite oxides display interesting phase transitions and ferroelectric, dielectric and optoelectronic properties [1–3]. Strontium titanate (SrTiO	extsubscript{3}), as a member of perovskite family, is an important dielectric material with an incipient ferroelectric transition (tetragonal–cubic) at 110 K. Above the transition temperature SrTiO	extsubscript{3} remains in a single stable cubic phase. This material has been investigated for its dielectric, piezoelectric and optoelectronic properties [4–7]. The fundamental band gap for the bulk is at 3.22 eV, attributed to transitions from 2p oxygen states to 3d titanium states [8, 9]. A high dielectric constant, excellent optical properties and a simple cubic structure with stable stoichiometry above the transition temperature of 110 K cause us to pay attention to the thin films, which may have prospective applications in microelectronics and optoelectronic fields. In recent years, a lot of research has been concentrated on optical properties of the films prepared by different techniques: refractive index in BaTiO	extsubscript{3} and SrTiO	extsubscript{3} films by RF sputtering [10], optical constants in SrTiO	extsubscript{3} films by sol–gel [11], chemical vapor deposition [12], excimer laser ablation [13] and hydrothermal and electrochemical processes [14, 15]. In this paper, we report the preparation of SrTiO	extsubscript{3} thin films on fused-quartz substrates by pulsed laser deposition (PLD).

Based on the structural characteristics of the film by X-ray diffraction and scanning electron microscopy, we investigate the fundamental optical constants, dispersion of the refractive index, optical band gap, interband transition and extinction coefficient. We also compare the preparation-technique dependence of the optical properties of the films. As shown, the optical properties of the thin films are strongly dependent on the crystallinity, grain boundaries, nonstoichiometries, precipitates and interface roughness [16].

2 Experimental

SrTiO	extsubscript{3} thin films were deposited on fused-quartz substrates by PLD. SrTiO	extsubscript{3} pellets were prepared by a chemical method to use as a PLD target. Strontium stearate and tetrabutyl titanate were dissolved in molten stearic acid at 70 °C. The resulting transparent solution was cooled, transformed into the dry gel and then heated at 750 °C for 1 h in air to finally form white SrTiO	extsubscript{3} powders. The powders were pressed into pellets and fired at 800 °C for 8 h. The PLD process was performed using a Lambda Physik KrF excimer laser system with 248-nm radiation and 30-ns pulse width. The energy density on the target surface was set at 2.5 J/cm	extsuperscript{2}. SrTiO	extsubscript{3} thin films were deposited at 600 °C for 30 min under 30 Pa of oxygen, followed by annealing at 700 °C under 0.05 MPa of oxygen for 30 min. X-ray diffraction (XRD) measurements were carried out on a Rigaku D/MAX-RA diffractometer with Cu K\(_\alpha\) as incident radiation. Film morphology was analyzed by a scanning electron microscope (SEM). The optical transmittance of the film was measured on a Hitachi U-3410 spectrophotometer in the wavelength range of 200–2000 nm.

3 Results and discussion

The phase structure of the as-prepared SrTiO	extsubscript{3} thin film was examined by XRD. Shown in Fig. 1 is the XRD pattern of the thin film on the fused-quartz substrate. The broadened peak at the lower diffraction angle of 22° is from the quartz substrate, on which a sharp peak from the SrTiO	extsubscript{3} thin film is overlapped. All the diffraction lines are assigned to the cubic perovskite structure. The sharp XRD peaks suggested that the as-prepared thin film was well crystallized after annealing for 30 min. The calculated lattice constants were...
$a = b = c = 3.9046$ Å, which are in good agreement with the data prepared by sol–gel [10]. Figure 2 shows the cross-sectional SEM image where denseness and homogeneity for the film are clearly seen. The thickness of the SrTiO$_3$ film was measured to be about 260 nm.

The optical properties of the as-prepared SrTiO$_3$ thin film on the quartz substrate were investigated by the optical transmittance spectra. Shown in Fig. 3 is the optical transmittance of the film. This spectrum displays a transparent zone with the interference pattern and a strong absorption. The transmittance decreases drastically to zero at about 313 nm. For light with a wavelength longer than 313 nm the SrTiO$_3$ film is transparent. In the region of low absorption, the incident light traverses the film several times and the interference fringes are subsequently produced. The interference fringes and relatively high transmittance indicate the low surface roughness and good homogeneity of the deposited SrTiO$_3$ film.

The optical band gap energy of the SrTiO$_3$ film can be determined from the transmittance spectrum recorded in the ultraviolet region. The transmittance $T$ varies exponentially with the absorption coefficient $\alpha$ near the absorption edge; therefore, $\alpha$ may be determined from the relation [17]

$$T = A \exp(-\alpha d)$$

(1)

where $d$ is the thickness of the film, $k$ is the extinction coefficient and the parameter $A$ is found to be nearly unity at the absorption edge. Assuming a direct transition between the valence and conduction bands, the relation between the absorption coefficient $\alpha$ and the incident photon energy $h\nu$ can be written as

$$(h\nu\alpha)^2 = C(h\nu - E_g)$$

(2)

where $C$ is a constant and $E_g$ is the optical band gap energy [17]. Thus the $E_g$ value can be estimated from the graph of $(h\nu\alpha)^2$ vs. $h\nu$ by extrapolating the linear portion of the graph at higher energies to $(h\nu\alpha)^2 = 0$. The $(h\nu\alpha)^2$ vs. $h\nu$ plot