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초고주파 응용을 위한 (100) 방향으로 성장된 PST / $LaNiO_3$박막의 구조적, 유전적 특성

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Dielectric and Structural properties of highly oriented PST/$LaNiO_3$ Thin Films for Microwave application

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Abstract

Pb0.5,Sr0.5TiO3(PST) thin films were deposited on the LaNiO3 (LNO(100))/Si and Pt/Ti/SiO2/Si substrates by the alkoxide-based sol-gel method. Structural and dielectric properties of PST thin films for the tunable microwave device applications were investigated. The PST films, which were directly grown on the Pt/Ti/SiO2/Si substrates showed the random orientation. For the LNO/Si substrates, the PST thin films exhibited highly (100) orientation. Compared with randomly oriented films, the highly (100)-oriented PST thin films showed better dielectric constant, tunability, and figure of merit (FOM). The dielectric constant, tunability, and FOM of the highly (100)-oriented PST thin film increased with increasing annealing temperature due to the decrease in lattice distortion. The differences in dielectric properties may be attributed to the change in the film stress and the in-plane oriented polar axis depending on the substrate was used. The dielectric constants, dielectric loss and tunability of the PST thin films deposited on the LNO/Si substrates measured at 1 MHz were 483, 0.002, and 60.1%, respectively.

Key Words : Dielectric properties; Sol-gel, Tunable,

1. 서 론

Dielectric thin films are attractive materials for the applications in tunable microwave devices such as electrically tunable mixers, delay lines, filters, capacitors, oscillators, resonators,and phase shifters [1, 2]. Nevertheless, for the applications mentioned above, there are the set of properties to be satisfied: moderate-to-low dielectric constant at microwave frequencies, low dielectric loss factor, large-scale variations of the dielectric constant by direct current (DC) biasing fields and low leakage current density [3].

Dielectric materials such as (Ba,Sr)TiO3 (BST) [4], PST (Pb,Sr)TiO3 [5] and (Pb,Ca)TiO3 (PCT) [6] and there similar titanates *characterized by close values of Curie* temperature androom temperature are well studied for tunable microwave applications. B. Dibenedetto et. al. investigated polycrystalline samples of (PbxSr1-x)TiO3 ceramic system and established a complete seriesof solid solution from PbTiO3 (PTO) (T_c = 485 C, T_c Curie temperature)to SrTiO3 (STO) (T_c = -237 C) [7]. In addition, our previous reported that the lattice constant anddielectric properties of PST thin films depend on Pb/Sr ratio [8]. STO films have a cubic phase at room temperature and they are characterized by a lowercrystallization temperature in comparison with BST films. On the other hand, the dielectric constant is lower than that of BST films. In contrary, tetragonal PTO films showed higher dielectric constant at room temperature. The main peculiarity of PST films is that they have Curie temperature below than room temperature. Therefore, it is possible to expectthat PST films will show the properties of paraelectric materials with a high dielectric constant. This fact makes PST films to be very perspective material for tunable devices applications. The dielectric properties of dielectric materials depend on the structural properties of the films such as phase structures developed during an annealing process, orientations of crystalline phase and the interface structure between an electrode and a film. Many researchers have studied the relationships between electrical and structural properties for the dielectric thin films [9,10].

LaNiO3(LNO) films have attracted attention in few last years using a conducting material for applications in ferroelectric memories [11]. The LNO material hasthe same perovskite-type structure with a lattice constant of 0.38 nm, which matches well ferroelectric and dielectric material such as BST, and PZT perovskite materials. The similarity in crystal structure and lattice constants between the electrode and dielectric thin films produces the better lattice matching and favorable structural, and thus dielectric properties. Since the PST films for tunable microwave device are not widely studied, the main purpose of our work includes to use LNO as the metal oxide bottom electrode and to offerthe benefits of better lattice matching to grow PST thin films with the preferred orientation.

2. 실 험

LNO thin films were prepared by the MOD method and were spin-coated on various substrates. The precursor materials used for the LNO preparation were lanthanum acetate, and nickel acetate. Acetic acid and de-ionized water were used as the solvents. First, the equal moles of lanthanum and nickel acetate were dissolved separately in acetic acid under the constant stirring at room temperature. Then, the stock solutionswere mixed and stirred at room temperature. The total concentration of the synthesized solution was 0.25 Mole. The final solution becametransparent and green colored. The LNO solutions were spun to a Si (100) substrate, and then pre-baked on a hot plate at 400 C for 10 min. The pre-baked film was annealed at 650 C for 1 h in the oxygen atmosphere to obtain crystallization. The total thickness of the LNO films was about 130 nm. The LNOthin films annealed at temperature as high as 600C exhibit an (100)-oriented structure. The resistivity of LNO thin film annealed at 650 C was 3.5 10-5 cm.

PST thin films were deposited on the LNO/Si and Pt(111)/Ti/SiO2/Si substrates by a sol-gel method. Thin films of (PbxSr1-x)O3 (x=0.5) with excess Pb-acetate of 10 mol % were prepared using lead acetate trihydrate, acetate and titanium iso-propoxide as the starting materials. Acetic acid and 2-methoxyethanol were used as the solvent forthe sol-gel method. PST precusor solutions were spin-coated on the LNO(100)/Si and Pt(111)/Ti/SiO2/Si substrates, and then pre-baked on a hot plate at 400 C for 10 minto remove the organic materials. The pre-baked films were annealed at various temperatures in the range 550700 C for 1 h in the oxygen atmosphere to obtain crystallization. The final thickness of PST thin film was about 200 nm. Measurements of dielectric properties were carried out in the metal-insulator-metal (MIM) capacitor cell. For these purposes, top Pt electrodes with diameter of 300 m were deposited on the PST films by dc sputtering method. Capacitance-voltage characteristics as well as dielectric constant and dielectric loss were measured using HP 4192 impedance analyzer. To investigatethe crystallinity, the phase formation parameters and orientation of the PST thin films, X-ray diffraction (XRD) profiles were obtained using CuK radiation source (Rigaku-D/MAX). Surface morphology of PST films was analyzed and quantified by atomic force microscope (AFM).

3. 결과 및 고찰

Figure 1 shows XRD patterns of the PST thin films deposited on the Pt(111)/Ti/SiO2/Si and LNO(100)/Si substrates with various annealing temperatures. All the films show a well-developed perovskite structure with no evidences of secondary phases formation. Both XRD pattern of the PST film annealed at 500 C shows the presence of such peaks as (100), (110), (111), (200) and (211). This fact indicates that the onset of crystallization into the perovskite phase is close to 500 C. Therefore, the annealing temperature of PST thin films is lower than that of BST (~650 C) thin film. The PST thin film deposited on Pt(111)/Ti/SiO2/Si substrate (Fig. 1(a)) exhibit randomly oriented structure, while the film was grown on LNO/Si substrate showed highly (100)-oriented structure. The degree of preferred orientation of each film is roughly estimated with the relative intensity ratio (*hkl*) = I(*200*)/(*I*(200) +*I*(110)), where I is peak intensity of the each film [12]. For the considered in Fig. 1(b), the value increased with increasing annealing temperature. The values of the PST thin films deposited on LNO substrates were 0.86, 0.88, 0.89, and 0.94for the annealing temperatures of 500 C, 550 C, 600 C, and 650 C, respectively. The values derived from XRD suggest that the PST films deposited on LNO/Si substrate were crystallized with (001) preferred orientation. The results mentioned above indicate that the crystallization and growth of the PST thin films are influenced by the substrate was used. Particularly, (100)-oriented LNO thin film as a substrate provides (001) preferred orientation for PST films due to well matching of the lattice parameter between the LNO and PST film.

Figure 2 illustrates the lattice parameters along the surface normal, in-plane, and lattice distortion of the PST/LNO/Si substrate structure measured from the XRD patterns for (001)-orientated PST films. The in-plane lattice parameters of the PST thin films were larger than normal lattice parameters. This resultindicates that the possibility of the in-plane tetragonal lattice distortion, although the corresponding bulk structure is a cubic. Additionally, these lattice parameters were elongated along the in-plane direction in the PST films. As it can be seen in Fig. 2, the variation of the lattice distortion, which is the lattice parameter ratio of surface normal to in-plane direction, decreased with increasing annealing temperature. Generally speaking, there are two stresses such as intrinsic and extrinsic stress in a thin film. The intrinsic stress appears in a bulk as the response for the defect such as dislocation in the film. The extrinsic stress comes from the contact layer between the film and substrate. The reasons are the difference of thermal expansion coefficient between the film and its substrate, lattice mismatch as well as the possibility of chemical reactions. As shown in Fig. 2, the lattice distortions decrease with increasing annealing temperature. Therefore, we suggest that the thermal stress developed by the difference in the thermal expansion coefficient is gradually relaxed during a high annealing temperature.

Figure 3 shows the dielectric constant and dielectric loss of the PST thin films deposited on both types of the substrates mentioned above with various annealing temperatures. The frequency range for these measurements was 100 Hz - 1 MHz with oscillation voltage of 0.1 V. The dielectric constant of the PST thin film with the LNO bottom electrode were higher compared with the PST thin films on the Pt substrates. The increase of the dielectric constant may be explained by the increase of the grain size as well as by the reduced lattice distortion. The dielectric constants for the PST thin film deposited on the LNO bottom electrode and Pt substrates measured at 1 MHz were 483, and 368, respectively.

Figure 4 shows the voltage dependent dielectric properties such as dielectric constant of the PST thin film annealed at 550C on the LNO bottom electrode and Pt bottom electrode substrates measured at 1 MHz. The figure of merit (FOM) is a frequently used parameter to characterize the correlations between the tunability and dielectric loss. This parameter is defined as FOM = [(%) tunability / tan (%)], where dielectric loss is given in percentage scale [10]. FOM value reflects the fact that a tunable microwave circuit cannot take full advantage of high tunability if the loss factor is too high. Ideally, the FOM value is desired to be as high as possible. The tunability was defined as (max-min)/max, where max and min are the maximum and minimum values of permittivity, which were measured at zero electric field and 250 kV/cm electric field, respectively. The tunability and FOM for the PST thin film on the LNO bottom electrode was 60.1%, and 29.5. These values are relatively higher in comparison with the PST thin films on the Pt bottom electrode (51.7%, and 25.8). The enhanced tunabilityand FOM may be related to (100)-oriented PST thin film. We assume that the changes of dielectric properties mentioned above may be attributed to the changes in the film stress and in the in-plane oriented polar axis. This result is in a good agreement with the data reported by P. Padmini et. al [13]. They suggested that a BST film was subjected to tensile stress, a contraction occurred along the caxis resulting in the enhancement of the in-plane oriented polar axis.

4. 결 론

In this work, we have shown that PST films with high tunabilities, low losses and high FOM can be prepared onto the LNO/Si substrate by sol-gel method. The PST films directly grown on the Pt/Ti/SiO2/Si substrates showed randomly orientation. For the LNO/Si substrates, the PST thin films exhibited highly (100) orientation. The highly (100)-oriented PST thin films showed better dielectric constant, tunability, and FOM parameter. Improved dielectric properties have been attributed to the change in the film stress and the in-plane oriented polar axis in the PST thin film.

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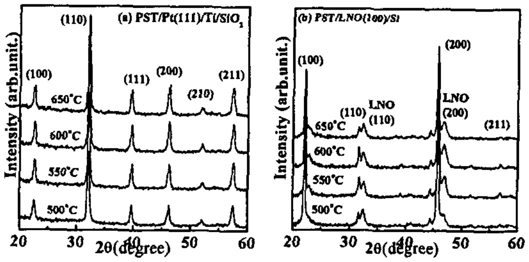
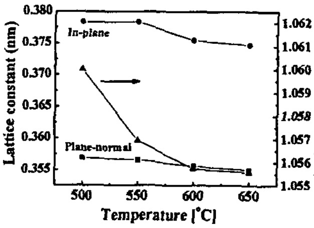


Figure 1. XRD patterns of (a) PST/Pt(111)/Ti/SiO2/Si and (b) PST/LNO/Si for various annealing temperatures.



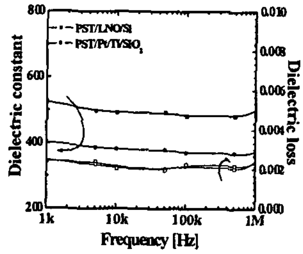


Figure 2. The lattice constant along in-plane and surface-normal directions, and lattice distortion of PST/LNO/Si structure as a function of annealing temperature.

Figure 3. Dielectric constant and dielectric loss of PST thin films deposited on: (a) Pt bottom electrode, and (b) LNO bottom electrode annealed at 550 C as a function of frequency.

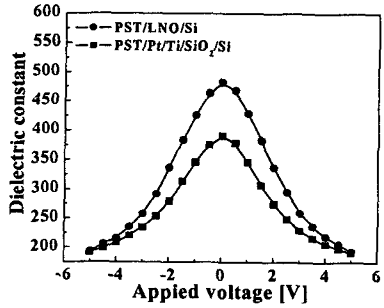


Figure 4. Dielectric constant voltage curves of the PST thin films deposited on: (a) Pt bottom electrode, and (b) LNO bottom electrode annealed at 550 C.