Thickness dependence of dielectric constant in barium strontium titanate thin films

B Panda, G D Nigam & S K Ray

Department of Physics & Meteorology, Indian Institute of Technology, Kharagpur 721 302

Received 3 February 1999

Radio frequency magnetron sputtered $\text{Ba}_{0.8}\text{Sr}_{0.2}\text{TiO}_3$ thin films have been deposited on silicon $\text{Si}/\text{SiO}_2/\text{Ti}/\text{TiN}/\text{Pt}$ substrate. The structural and electrical properties have been investigated using X-ray diffraction and capacitance-voltage characteristics of fabricated capacitors. The growth and orientation of the film have been found to depend upon the type of substrates and deposition temperatures. The effective dielectric constant of BST film in $\text{Si}/\text{SiO}_2/\text{Ti}/\text{TiN}/\text{BST}/\text{Al}$ thin film capacitor is found to increase with the increasing thickness of the film. A space charge layer with low dielectric constant is found to form at the film electrode interface.

1 Introduction

The search for a high permittivity material, that could increase the storage capabilities of next generation dynamic random access memory (DRAM) cell is underway. Ferroelectric perovskite oxide films are very much attractive due to their high dielectric constant, and are being exploited in the VLSI/ULSI applications. The potential utilization of high dielectric constant films can be made for fabricating new generation of memory devices by studying the synthesis, characterization and determining the process composition microstructure-property relationship of ferroelectric thin films. Although, lot of studies have been undertaken to make a good progress in achieving the above goals, there exists a considerable scope for further research in this challenging field. In recent years, barium strontium titanate (BST) thin films are found to be very attractive because of their very high dielectric constant. The present investigation is concerned with the studies on the structural and thickness dependence of dielectric constant of non-lead based barium strontium titanate (BST) thin films prepared by rf magnetron sputtering.

2 Experimental Details

A 13.56 MHz, 0-500 W, rf magnetron sputtering system (EDWARDS ESM 100) with 7.5 cm diameter cathode was used for deposition of $\text{Ba}_{0.8}\text{Sr}_{0.2}\text{TiO}_3$ (BST) films. These films were deposited at different chamber pressures maintaining an argon to oxygen ratio 90:10 on silicon and $\text{Si}/\text{SiO}_2/\text{Ti}/\text{TiN}/\text{Pt}$ substrates. TiN(2000Å) on SiO$_2$/Si substrate was used as an intermediate barrier layer. Pt(300Å) electrode was deposited on $\text{Si}/\text{SiO}_2/\text{Ti}/\text{TiN}$ substrates by e-beam evaporation at a substrate temperature of 450°C. Here Ti layer is used as an adhesive layer. Sputter deposition of BST film was carried out at substrate temperatures of 480°, 530° and 580°C respectively. The influence of bottom electrode, substrate temperature and deposition temperature on film orientation and phase formation was investigated using X-ray diffraction analysis. Dielectric properties of BST films were evaluated from capacitance voltage ($C-V$) characteristics of fabricated metal-insulator-semiconductor (MIS) and metal-insulator-metal (MIM) capacitors. All the measurements were made using HP 4061-A computerized semiconductor test system consisting of multi-frequency LCR meter (HP-4275-A), a pico-ammeter/dc voltage source (HP 4140-B) and a switching subsystem.

3 Results and Discussion

Structural characteristics of BST films were studied from X-ray diffraction (XRD) pattern of the deposited films using CuK$_\alpha$ radiation. X-ray diffraction spectra of as grown BST films deposited on silicon (100) face at various substrate temperatures are shown in Fig. 1. As can be seen from the figures, the orientation and crystallinity of the films are strongly dependent on the deposition temperature. Since sputtering from a multi-component target like $\text{Ba}_{0.8}\text{Sr}_{0.2}\text{TiO}_3$ causes fragmentation into $\text{BaO}$, $\text{SrO}$ and $\text{TiO}_2$ components, we investigated the lowest temperature required for the formation of BST film during in situ deposition. It was...
observed from X-ray diffraction that a perovskite phase of BST was formed at a temperature greater than 480°C. In situ deposition and heating at 580°C resulted in the growth of oriented films. The crystallinity of the films increased with increasing temperature. Higher degree of preferred orientation of the BST film is observed on Si/SiO₂/SiN/Pt structure (Fig. 2) as compared to Si substrate.

It is clear from Figs 1 and 2 that the film deposited on silicon is polycrystalline at a deposition temperature around 500°C and gradually orients itself along <200> direction at a higher substrate temperature (580°C), while in the case of platinum the orientation takes place at a lower temperature (530°C). Lattice mismatch is very large between silicon (a = 5.43 Å) and BST (a = 3.93 Å), while the lattice constant of platinum (a = 3.93 Å) is very close to that of BST. The thick films at higher substrate temperatures preferentially orient along <200> direction which is considered to be related to the surface energy and the structure factor of the perovskite phase formation. The effective dielectric constant of BST film used in the multilayered capacitor, Si/SiO₂/Ti/TiN/Pt/BST/Al, increases with the increase of BST film thickness and ultimately tends to a saturation (Fig. 3).

The local field in the film originating from the space charge layers at the interfaces, located at both the top and the bottom ends of the film and the electrode, is the route cause of this thickness dependence. If \( C_{\text{BST}} \) be the capacitance due to BST film and \( C_{\text{i}} \) be the interfacial capacitance, the effective capacitance is given by:

\[
C = \left( \frac{C_{\text{BST}}}{C_{\text{BST}} + C_{\text{i}}} \right)
\]

If \( C_{\text{BST}} \ll C_{\text{i}} \), i.e., BST film thickness is very high, the effective capacitance will be due to the capacitance of the BST film only. So, at higher thickness the effective capacitance \( \varepsilon_{\text{eff}}/A/C \), i.e., the effective dielectric constant will tend towards the actual dielectric constant of the BST film. Fig. 4 shows the \( \varepsilon_{\text{eff}}/A/C \) as the function of film thickness \( d \) where \( A \) is the electrode area, \( C \) is the measured capacitance and \( \varepsilon_{0} \) is the dielectric constant of the free space. The positive value of \( \varepsilon_{\text{eff}}/A/C \) at zero film thickness indicates the assumption of the formation of an interfacial space charge layer.

4 Conclusions

BST films have been deposited on Si/SiO₂/Ti/TiN/Pt/BST (80/20) substrate by rf magnetron sputtering. The crystallinity of BST films has been
Fig. 4 — $\varepsilon_{A/C}$ as a function of film thickness for BST film Si/SiO$_2$/Ti/TiN/Pt/BST (80/20); frequency = 1 MHz, substrate temperature = 530°C

found to increase with substrate temperature. Films deposited on platinum coated substrates orient preferentially along $<200>$ direction at a lower temperature compared to that on silicon. The effective dielectric constant of the film is found to increase with the increasing thickness of film. The formation of an interfacial space charge layer is assumed to be the cause behind the thickness dependent dielectric constant of the film.

References