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## Ac conductivity and dielectric spectroscopy studies on tin oxide thin films formed by spray deposition technique

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#### ABSTRACT

Au/tin oxide/n-Si (1 0 0) structure has been created by forming a tin oxide (SnO<sub>2</sub>) on n-type Si by using the spray deposition technique. The ac electrical conductivity ( $\sigma_{ac}$ ) and dielectric properties of the structure have been investigated between 30 kHz and 1 MHz at room temperature. The values of e', e'', tan  $\delta$ ,  $\sigma_{ac}$ , M' and M'' were determined as 1.404, 0.357, 0.253, 1.99 × 10<sup>-7</sup> S/cm, 0.665 and 0.168 for 1 MHz and 6.377, 6.411, 1.005,  $1.07 \times 10^{-7}$  S/cm, 0.077 and 0.078 for 30 kHz at zero bias, respectively. These changes were attributed to variation of the charge carriers from the interface traps located between semiconductor and metal in the band gap. It is concluded that the values of the e', e'' and tan  $\delta$ increase with decreasing frequency while a decrease is seen in  $\sigma_{ac}$  and the real (M') and imaginary (M'') components of the electrical modulus. The M'' parameter of the structure has a relaxation peak as a function of frequency for each examined voltage. The relaxation time of M'' ( $\tau_{M'}$ ) varies from 0.053 ns to 0.018 ns with increasing voltage. The variation of Cole–Cole plots of the sample shows that there is one relaxation.

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#### 1. Introduction

The oxide layers such as SiO<sub>2</sub>, SnO<sub>2</sub> and ZnO fabricated on the semiconductor play a significant role in the description of the characteristic parameters of the MOS devices [1–5]. There are different techniques to fabricate tin oxide (SnO<sub>2</sub>) film such as pulse laser deposition [6], chemical vapor deposition (CVD) [7], spray deposition [3,5,8], sol-gel [9], sputtering [10], etc. Among these techniques, the spray deposition method gives a simple way to contact tin oxide thin films on the Si substrates due to low production costs, low processing temperatures and the possibility of good control of the deposition parameters. SnO<sub>2</sub> is an attractive material for electronic and optoelectronic devices, such as flat panel displays, organic light emitting diodes, and solar cells and heat mirrors. This is mainly due to the coexistence of both high transparency to light in the visible region and high conductivity. It is well known that widely-used SnO<sub>2</sub> thin films are n-type semiconductor oxide because of the presence of intrinsic defects (oxygen vacancies/or metal interstitials) [11]. SnO<sub>2</sub> thin films have a broad band gap of  $E_g$ =3.6 eV at room temperature [12].

The dielectrical and electrical characteristics of metal–semiconductor (MS) contacts [13–21] can be altered by proper oxide semiconductors [3–5], when an oxide layer is added between the semiconductor and metal. The electrical and dielectrical properties of metal-oxide-semiconductor (MOS) interfaces have been widely

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investigated for electronic and optoelectronic device applications [1–5]. The electrical properties of MOS interface strongly depend on the formation of the interfacial layer and the fabrication conditions of the surface [22–24]. The performance and reliability of MOS devices such as Schottky diode is strongly affected by the oxide interface layer between the semiconductor surface and metal. Thus, it is important to determine the dielectrical properties of interface of a Schottky diode [25]. The presence of such an oxide layer can have a powerful effect on the diode characteristics [3,8,25]. Therefore, it is significant to obtain the ac conductivity and dielectric properties of such an oxide based Schottky diode. The forward and reverse bias C-V and G-V measurements give the important information about ac conductivity and the dielectric spectroscopy properties of the Schottky diodes. The G–V and C–V curves in the idealized case are frequency independent [26–28]. However, this idealized case is often disturbed due to the existence of an oxide layer such as tin oxide between interface states at the oxide layer/semiconductor interface and the contact materials [26-28].

In the present work, the ac conductivity and dielectric parameters of Au/tin oxide/n-Si (100) structures fabricated by spray deposition technique were investigated. The important goal of this study was to report estimates of the relaxation behavior of the structure as a function of voltage and frequency.

#### 2. Experimental procedure

The Au/tin oxide/n-Si structure was formed on n-type (P doped) Si wafer with  $(1\ 0\ 0)$  surface orientation, 380  $\mu$ m thickness







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**Fig. 1.** Variation of (a) the real  $(\varepsilon')$  and (b) the imaginary  $(\varepsilon'')$  parts of the dielectric constant with voltage between 30 kHz and 1 MHz.

and  $20\,\Omega\,cm$  resistivity. Firstly, the Si wafer was cleaned in trichloro ethylene, methanol and acetone using ultrasonic agitation for 5 min and then rinsed in de-ionized water of 18  $M\Omega$  and then chemically etched in a sequence of  $H_2SO_4 + H_2O_2$  (1:1) for 10 min,  $NH_3 + H_2O_2$  (1:1) for 10 min, 38%  $HF + H_2O$  (1:15) for 2 min,  $\text{HNO}_3 + \text{HF} + \text{CH}_3\text{COOH}$  (2:1:1) for 4 min, 38%  $\text{HF} + \text{H}_2\text{O}$ (1:15) for 2 min. All the chemical mixtures used are in vol. The metal contact with a thickness of 2000 Å was done by evaporating aluminum (Al) on n-Si wafer, then was annealed at 400 °C for 3 min in high purity nitrogen for creating ohmic contact. A layer of tin oxide (SnO<sub>2</sub>) was deposited on n-Si (100) wafer located onto substrate at 450 °C by spray deposition technique a solution consisting of 27.44 wt% of stannic chloride ( $SnCl_4 \cdot 5H_2O$ ), 32.21 wt% of ethyl alcohol and 40.35 wt% of deionized water. The spray deposition system has been described in detail in Ref. [29]. Tin oxide dots were 4 mm in diameter. The Schottky contacts have been done by evaporation of gold (Au) as dots with a diameter of approximately 2 mm onto all of the n-Si wafer. The thickness of the gold rectifying contact is 2000 Å. All evaporation processes were performed in a vacuum-coating unit at about  $4 \times 10^{-6}$  Torr. The conductance–voltage (*G*–*V*) and capacitance–voltage (*C*–*V*) measurements were obtained between 30 kHz and 1 MHz at room temperature and in dark using HP 4192A LF impedance analyzer (5 Hz–13 MHz).

#### 3. Results and discussion

The real dielectric constant ( $\epsilon'$ ), imaginary dielectric constant ( $\epsilon''$ ), tangent loss (tan  $\delta$ ), ac conductivity ( $\sigma_{ac}$ ) and electric modulus ( $M^* = M' + iM''$ ) are significant parameters in the selection of materials for device application. It has been evaluated the frequency dependence of the conductance–voltage (G–V) and capacitance–voltage (C–V) characteristics for Au/tin oxide/n-Si (1 0 0) structure between 30 kHz and 1 MHz at room temperature. The values of capacitance and conductance obtained as a function of frequency under both reverse and forward bias for the effect of series resistance were corrected. From the values of the corrected C–V and G–V of Au/tin oxide/n–Si (1 0 0) structure between 30 kHz and 1 MHz, the gate voltage dependence of  $\epsilon'$ ,  $\epsilon''$ , tan  $\delta$ ,  $\sigma_{ac}$ , M' and M'' of the structure are obtained. These parameters have been calculated using the following relations [25,30–32]:

$$\epsilon^* = \epsilon' - i\epsilon'' \tag{1}$$

$$\varepsilon' = \frac{C_c d_{ox}}{\varepsilon_0 A} \tag{2}$$

$$\varepsilon'' = \frac{d_{ox}}{A\varepsilon_o} \frac{G_c}{\omega}$$
(3)

$$\tan \delta = \frac{\varepsilon}{\varepsilon'} \tag{4}$$

$$\sigma_{ac} = 2\pi f \varepsilon_0 \varepsilon' \tan \delta \tag{5}$$

$$M^{*} = \frac{1}{\varepsilon^{*}} = \frac{1}{\varepsilon' - i\varepsilon''} = \frac{\varepsilon'}{\varepsilon'^{2} + \varepsilon''^{2}} + i\frac{\varepsilon''}{\varepsilon'^{2} + j\varepsilon''^{2}} = M' + iM'', \quad i = (-1)^{1/2}$$
(6)

where  $C_c$  and  $G_c$  are the corrected capacitance and conductance,  $\varepsilon'$ and  $\varepsilon''$  are the real and the imaginary of the complex dielectric constant, and *i* is the imaginary root of -1, *f* is the frequency of the applied ac field (Hz),  $\omega = 2\pi f$  is the angular frequency, *A* is the surface area of the sample,  $\sigma_{ac}$  is the ac conductivity, *M'* and *M''* are the real and the imaginary electric modulus,  $\varepsilon_o$  is the permittivity of free space charge ( $\varepsilon_o = 8.854 \times 10^{-14}$  F/cm) [15] and  $d_{ox}$  is the interfacial oxide layer thickness. The  $d_{ox}$  calculated from high frequency (1 MHz) capacitance–voltage data in accumulation for oxide layer capacitance ( $C_{ox} = \varepsilon_i \varepsilon_0 A/d_{ox}$ ), where  $C_{ox}$  is determined as 1.061 nF,  $\varepsilon_i = 7\varepsilon_0$  [8] and  $\varepsilon_0$  are the permittivity of the tin oxide layer and free space, has been determined to be about 183.4 nm for tin oxide layer.

Knowledge of the dielectric properties of the structures is of vital importance in various areas of science and engineering. The real part  $\epsilon'$  is attributed to as the dielectric constant and indicates stored energy when the material is exposed to an electric field, while the dielectric loss factor  $\epsilon''$ , which is the imaginary part, effects energy absorption and attenuation. The  $\epsilon'$  values of the structure have been calculated by using Eq. (2) and illustrated in Fig. 1(a). Fig. 1(a) depicts the variation of real part of the dielectric constant ( $\epsilon'$ ) with voltage for Au/tin oxide/n-Si (1 0 0) structure between 30 kHz and 1 MHz at room temperature. It is clear from the figure that the values of  $\epsilon'$  increase with increasing positive

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