

Frequency dependence of dielectric parameters of structure with $\text{Bi}_{3.25}\text{La}_{0.75}\text{Ti}_3\text{O}_{12}$ thin film prepared by sol-gel method

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In this study, the frequency dependence of dielectric parameters of $\text{Bi}_{3.25}\text{La}_{0.75}\text{Ti}_3\text{O}_{12}$ (BLT) thin film prepared on Al-coated p-type silicon substrate by sol-gel method were investigated using admittance ($Y=G+i\omega C$) measurements. These measurements were carried out in the frequency range of 10 kHz-1 MHz. The frequency dependence of the capacitance (C) and conductance (G/ω) indicates the existence of interface states. Dielectric constant (ϵ'), loss (ϵ'') and loss tangent ($\tan \delta$), ac conductivity (σ_{ac}) and complex electric modulus (M) values were calculated from impedance measurements. It has been found that the ϵ' and ϵ'' decrease while the σ_{ac} increases with the increasing frequency. Experimental results show that the values of dielectric parameters are a strong function of frequency.

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1. Introduction

Ferroelectric materials have attracted much attention due to potential applications in multilayer ceramic capacitors and dynamic random access memories. Among numerous ferroelectrics, lanthanum bismuth titanate (BLT) thin film has useful properties for optoelectronic devices, ferroelectric, piezoelectric, and memory applications. For the memory applications, electrical properties on the BLT thin films should be investigated thoroughly. High electric fields in the thin film structure cause nonlinear electrical behaviors; especially, leakage current-voltage characteristics become non-ohmic [1-6].

Dielectric spectroscopy (sometimes called impedance spectroscopy) measures the dielectric properties of a medium as a function of frequency. Dielectric spectroscopy (DS) is based on the interaction of an external field with the dipole moment of the sample. The DS is particularly characterized by the measurement and analysis of some or all of the four impedance-related functions impedance (Z), admittance (Y), electric modulus (M), and dielectric permittivity (ϵ). Furthermore, from an impedance spectrum, both the structure of its equivalent circuit and the parameter values can be extracted [7-11]. The dielectric properties of many materials depend on frequency, temperature, moisture content, bulk density and chemical composition.

In our previous study [6], we fabricated the $\text{Al}/\text{Bi}_{3.25}\text{La}_{0.75}\text{Ti}_3\text{O}_{12}/\text{p-Si}$ structure and its electrical and photoresponse properties were investigated under different illumination intensity and frequency. The aim of the present study is to investigate the effect of frequency on dielectric parameters of the fabricated structure. The

frequency dependence of dielectric parameters such as dielectric constant (ϵ'), loss (ϵ''), loss tangent ($\tan \delta$), ac conductivity (σ_{ac}) and electric modulus obtained from impedance measurements were discussed in detail.

2. Experimental details

$\text{Al}/\text{Bi}_{3.25}\text{La}_{0.75}\text{Ti}_3\text{O}_{12}/\text{p-Si}$ structure was fabricated on p-type silicon (boron doped) crystal with $\langle 111 \rangle$ surface orientation, having thickness of 600 μm and 5-10 $\Omega\cdot\text{cm}$ resistivity. The detail fabrication procedures of the fabricated diode were given in Ref. [6]. The capacitance and conductance measurements of the fabricated structure were performed using Keithley 4200 semiconductor characterization system in the frequency range of 10 kHz-1 MHz and at room temperature.

3. Results and discussion

3.1. Frequency dependence of capacitance and conductance

The variation of the capacitance (C) and conductance (G/ω) with frequency is given in Figs. 1(a) and (b), respectively. As seen in these figures, the value of C and G/ω decreases with increase in the frequency. The frequency dependence of the capacitance can also arise due to the presence of deep lying impurities in the depletion region of semiconductor. Deep impurities are the energy levels of intrinsic lattice defects or impurity atoms that have energy near the center of the band gap. As the

frequency is increased, the capacitance decreases to the same limit, as the charges on the defects no longer have time to rearrange in response to the applied voltage. In other words, at high frequencies such that the carrier life time τ is much larger than $1/\omega$, the charges at the interface states cannot follow the ac signal. Therefore, the contribution of interface states capacitance to the total capacitance can be neglected [12-18].

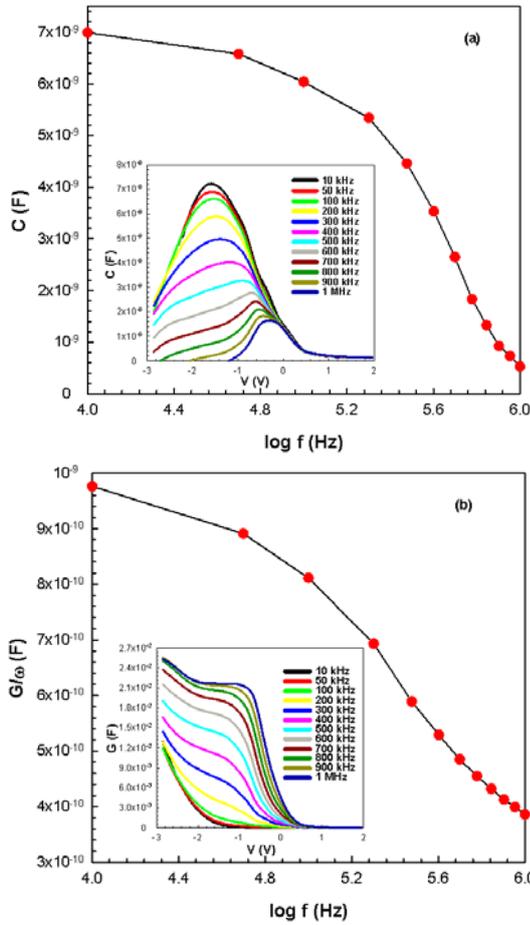


Fig. 1. The variation of (a) capacitance (C) and (b) conductance (G/ω) with log frequency of the structure

3.2. Frequency dependence of dielectric parameters

Dielectric spectroscopy sometimes called as impedance spectroscopy is a method used to determine the dielectric characteristics of non-conducting or semiconducting materials in relation to their structure and also of electronic or sensor devices. Dielectric spectroscopy measures the dielectric permittivity as a function of frequency and temperature. The dielectric permittivity (ϵ^*) is expressed in the complex form: [7-9]

$$\epsilon^*(\omega) = \epsilon'(\omega) - i\epsilon''(\omega) \quad (1)$$

where ϵ' and ϵ'' are the real and the imaginary parts of complex dielectric permittivity (ϵ^*), and $i = \sqrt{-1}$ the imaginary factor. In the ϵ^* formalism, in the case of admittance measurements, the following relation holds:

$$\epsilon^* = \frac{Y^*}{j\omega C_o} = \frac{C_m}{C_o} - j \frac{G_m}{\omega C_o} \quad (2)$$

where Y^* , C_m and G_m are the measured admittance, capacitance and conductance of the dielectric and ω is the angular frequency ($\omega = 2\pi f$) of the applied electric field [19-21]. Also, C_o is capacitance of an empty capacitor. $C_o = \epsilon_o (A/d)$; where A is the rectifier contact area in cm^2 , d is the interfacial insulator layer thickness and ϵ_o is the permittivity of free space charge ($\epsilon_o = 8.85 \times 10^{-14}$ F/cm).

The real part of the complex permittivity, the dielectric constant (ϵ'), at the various frequencies is calculated using the measured capacitance values at the strong accumulation region from the relation [7,8,22-24],

$$\epsilon'(\omega) = \frac{C_m}{C_o} \quad (3)$$

The imaginary part of the complex permittivity, the dielectric loss (ϵ''), at the various frequencies is calculated using the measured conductance values from the relation,

$$\epsilon''(\omega) = \frac{G_m}{\omega C_o} \quad (4)$$

The dissipation factor or loss tangent ($\tan\delta$) is defined as the ratio of the imaginary part of the dielectric constant to the real part and can be expressed as follows,

$$\tan \delta = \frac{\epsilon''(\omega)}{\epsilon'(\omega)} \quad (5)$$

The ac conductivity (σ_{ac}) of all samples has been calculated from the dielectric losses according to the relation

$$\sigma_{ac} = \omega C \tan \delta (d/A) = \epsilon_o \omega \epsilon'' \quad (6)$$

The electric modulus approach began when the reciprocal complex permittivity was discussed as an electrical analogue to the mechanical shear modulus [21]. From the physical point of view, the electrical modulus corresponds to the relaxation of the electric field in the material when the electric displacement remains constant. The complex electric modulus $M^*(\omega)$ was introduced to describe the dielectric response of non-conducting materials and can be expressed as follows [25-27],

$$M^*(\omega) = \frac{1}{\epsilon^*} = M'(\omega) + jM''(\omega) \quad (7)$$

$$M'(\omega) = \frac{\epsilon'(\omega)}{\epsilon'(\omega)^2 + \epsilon''(\omega)^2}$$

and

$$M''(\omega) = \frac{\epsilon''(\omega)}{\epsilon'(\omega)^2 + \epsilon''(\omega)^2} \quad (8)$$

where M' and M'' are the real and the imaginary of complex electric modulus.

Figs. 2a-c show the frequency dependence of dielectric constant (ε'), loss (ε'') and loss tangent (tan δ) of the structure with BLT thin film. As seen in Fig. 2(a) and (b), with increasing frequency, the value of the ε' and ε''

decreases. At low frequencies, where the electron hopping can match the frequency of the applied field, the value of ε' is maximum. When the frequency of the applied field is increased, the electron exchange cannot follow the alternating field and so the ε' decreases. In other words, especially at high frequencies, the interfacial dipoles have less time to orient themselves in the direction of the alternating field [28-32]. Also, the fast rising trend of ε'' at low frequencies may be due to the polarization mechanism associated with the thermally activated conduction of mobile ions and/or other defects [33]. As seen in Fig. 2(c), the value of dielectric loss tangent (tanδ) remains almost constant up to about 200 kHz, and then it increases with increase in the frequency. The loss tangent is associated with the electrical conductivity.

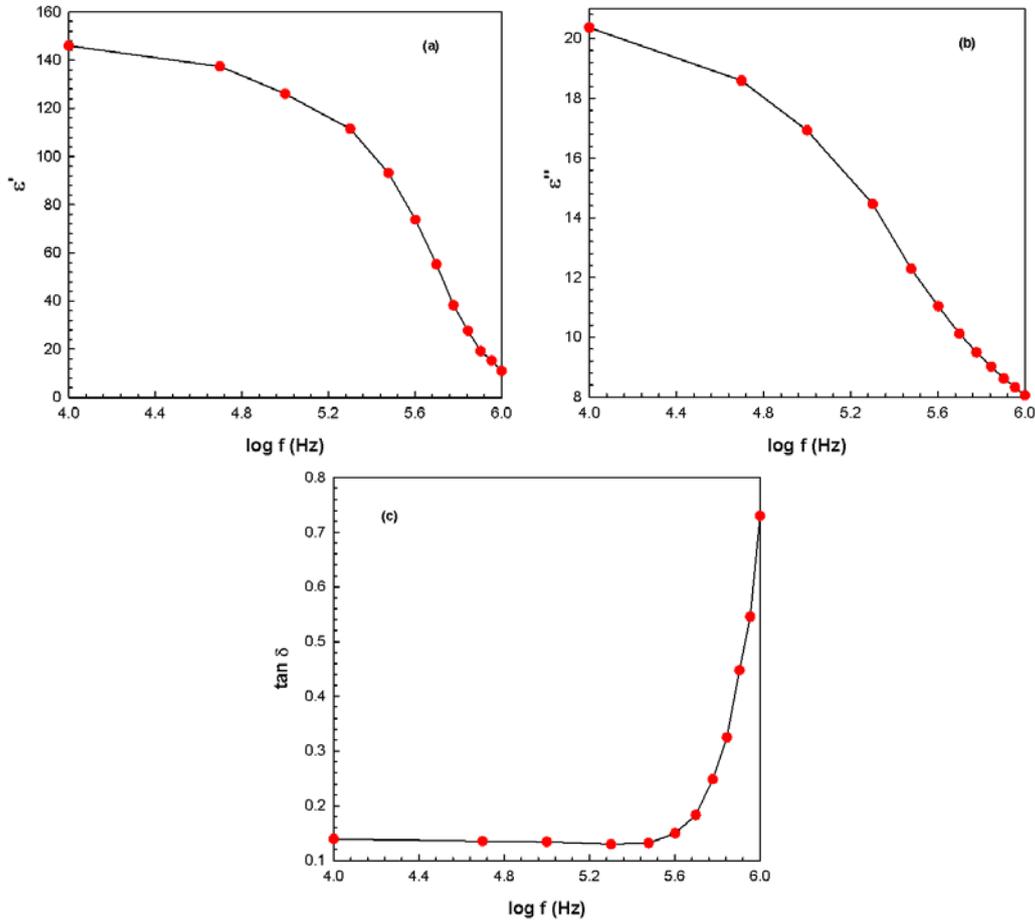


Fig. 2. Frequency dependence of (a) ε' (b) ε'' and (c) tan δ

Fig. 3 shows the frequency dependence of ac electrical conductivity (σ_{ac}). As seen in Fig. 3, the σ_{ac} increases with increase in frequency. The increase in ac conductivity may be due to the increase in electron hopping.

In other words, an increase in frequency improves the electron hopping frequency between the charge carriers resulting thereby in an increase in conductivity. Also, the frequency dependence of ac conductivity can be attributed to the relaxation phenomena arising from mobile charge carriers [34-38].

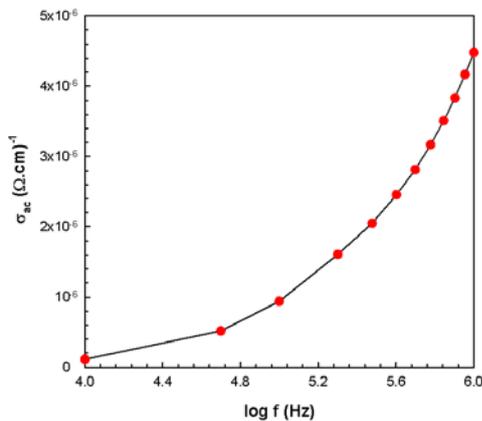


Fig. 3. Frequency dependence of ac electrical conductivity

Fig. 4(a) and (b) show the frequency dependence of real (M') and imaginary (M'') parts of the complex electric modulus. As seen in Fig. 4(a), the value of M' increases with increasing frequency. The increase of M' may possibly be related to a lack of restoring force governing the mobility of charge carriers under the action of an induced electric field [39-45]. As seen in Fig. 4(b), the value of M'' remains almost constant up to about 200 kHz, and then it increases with increase in frequency. Such behavior of M'' may be due to the large value of capacitance associated with the electrode polarization effect.

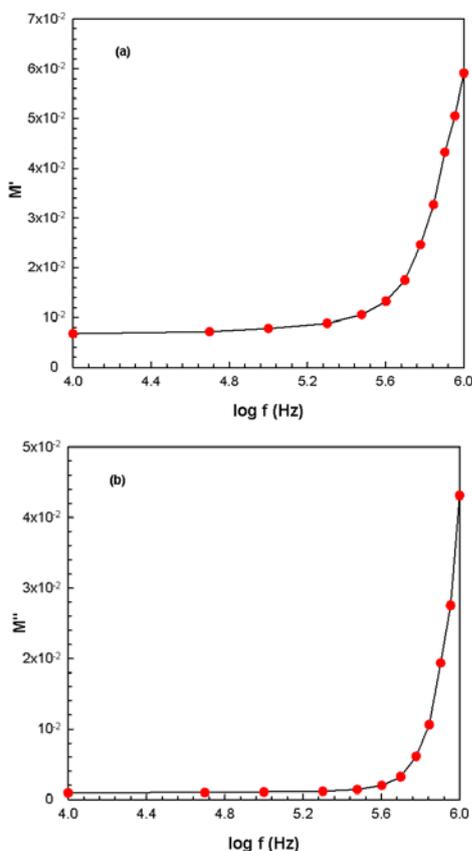


Fig. 4. Frequency dependence of (a) real (M') and (b) imaginary (M'') parts of complex electric modulus

4. Conclusion

In this study, the dielectric parameters of the structure with BLT thin film were obtained from the measured capacitance and conductance data. The dielectric constant (ϵ') and loss (ϵ'') were found to be decrease with increasing frequency. This behavior are explained by the fact that as the frequency is increased, the interfacial dipoles have less time to orient themselves in the direction of the alternating field. The ac conductivity (σ_{ac}) increases with increasing frequency. The increase in σ_{ac} can be due to the increase in charge carries hopping. Electric modulus formalism was also analyzed to obtain experimental dielectric data. The value of M' and M'' increases with increasing frequency. It is concluded that the electric and dielectric properties of the fabricated structure are strongly dependent on frequency.

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