EFFECT OF ANNEALING ON THE AC CONDUCTIVITY AND THE DIELECTRIC PROPERTIES OF In_2Te_3 THIN FILMS

M.A. Afifi, E. Abd El-Wahabb, A.E. Bekheet and H.E. Atyia

Physics Department, Faculty of Education, Ain Shams University, Roxy, Cairo, Egypt

(Received April 26, 2000; revised version July 13, 2000)

In_2Te_3 thin films were prepared by thermal evaporation technique. The composition of the films is checked by energy dispersive X-ray analysis. X-ray analysis showed that the as-deposited In_2Te_3 films as well as films annealed at temperatures $\leq$ 473 K have crystalline structure. The ac conductivity $\sigma_{ac}(\omega)$, the dielectric constant ε_1 and the dielectric loss ε_2 of In_2Te_3 films were studied in the temperature range 303–373 K and in the frequency range 100 Hz–100 kHz. The ac conduction activation energy $\Delta E_\sigma(\omega)$ was found to be 0.065 eV for the as-deposited films. The ac conductivity was found to obey the relation $\sigma_{ac}(\omega) = A\omega^s$, where s is the frequency exponent. The obtained temperature dependence of s is reasonably interpreted by quantum mechanical tunneling model. Both the dielectric constant ε_1 and the dielectric loss ε_2 increased with temperature and decreased with frequency in the investigated range. The frequency and temperature dependencies of $\sigma_{ac}(\omega)$, ε_1, and ε_2 for the annealed samples have the same behavior as that for the as-deposited samples. However, values of $\sigma_{ac}(\omega)$, ε_1, and ε_2 measured at any frequency and temperature increased with annealing temperature up to 473 K. It was found also that $\Delta E_\sigma(\omega)$ decreased with annealing temperature.

PACS numbers: 72.20.–i, 77.55.+f

1. Introduction

Although many recent works have dealt with $A_2^{III}B_3^{VI}$ compounds with A = Ga, In, Tl and B = S, Se, Te, their properties remain rather confusing. These compounds have mainly zinc-blende and wurtzite structures with tetrahedral [1] bonding. They are the simplest types of defective structures in which some of the crystallographically equivalent sites are only partially occupied and that the lattice has vacant sites [2]. The presence of a large number of defects destroys the periodicity of the lattice and distorts the crystal field. Therefore, the semiconducting properties of these materials should be strongly affected by annealing which increases the degree of ordering and hence affects their physical properties.

In_2Te_3 is a member of the above-mentioned compounds, has two phases (α and β) [3–6]. β-phase has a zinc-blende structure [7] which is characterized by a completely random distribution of the metal ions in their sublattice. It can be transformed to the ordered state (α-phase) as obtained by Kosevich et al. [8]. Zahab et al. [9] have shown that an appropriate annealing of In_2Te_3 films leads to a polycrystalline β-phase with a preferred orientation in the [111] direction. Several authors investigated the electrical properties of In_2Te_3 thin films [9–13]. Papers on the ac conductivity and the dielectric properties of In_2Te_3 are rare.

Measurements of ac conductivity of chalcogenide semiconductors have been extensively used to interpret the conduction process in these materials. For the mechanism of the ac conductivity, the quantum mechanical tunneling (QMT) model was proposed by Pollak and Geballe [14] to interpret impurity conduction in n-type silicon.

This work is aimed to investigate the temperature and frequency dependence of the ac conductivity and the dielectric properties of In_2Te_3 thin films of different thicknesses. Also, the effect of annealing at different temperatures on these properties has been investigated. The results are discussed on the base of QMT theory of ac conductivity.

2. Experimental technique

In_2Te_3 was synthesized [13, 15] by direct fusion of stoichiometric amounts of indium and tellurium (purity 99.999%) in an evacuated sealed silica tube (10^{-5} Torr). Thin films with different thicknesses of In_2Te_3 were obtained by thermal evaporation of the investigated composition onto cleaned glass substrates. X-ray diffraction technique was used to investigate the structure of the investigated films, using Philips PM 8203 diffractometer. The chemical composition of the obtained films was checked by energy dispersive X-ray analysis (EDX) using scanning electron microscope (Joel 5400). For dc and ac measurements, films were sandwiched between two Al electrodes. The resistance (R) was measured using an electrometer (Keithley model 616). The dc conductivity σ_{dc} is related to the electrical resistance by the relation

$$\sigma_{\mathrm{dc}} = d/RA, \tag{1}$$

where d is the thickness of the film and A is the cross-sectional area. The dielectric constant ε_1, the dielectric loss ε_2, and the ac conductivity σ_{ac} were measured using programmable automatic RCL meter (Philips PM 6304) which measures the impedance Z, the capacitance C and the loss tangent ($\tan\delta$) directly. The dielectric constant ε_1 (real part of the dielectric constant) was calculated using the relation [16]

$$\varepsilon_1 = C_x d/\varepsilon_0 A, \tag{2}$$

where C_x is the capacitance of the sample, A is the cross-sectional area of the parallel surfaces of the sample and ε_0 is the permittivity of the free space. The dielectric loss ε_2 (imaginary part of the dielectric constant) was calculated using the relation [17]

$$\varepsilon_2 = \varepsilon_1 \tan\delta, \tag{3}$$

where $\delta = 90^\circ - \phi$; ϕ is the phase angle. The ac conductivity $\sigma_{\mathrm{ac}}(\omega)$ is determined by the relation [17]

$$\sigma_{\mathrm{ac}}(\omega) = \sigma_{\mathrm{tot}}(\omega) - \sigma_{\mathrm{dc}},$$

where $\sigma_{\mathrm{tot}}(\omega)$ is the total conductivity which is calculated from the relation

$$\sigma_{\mathrm{tot}}(\omega) = \frac{d}{A}\frac{1}{z}, \tag{4}$$

where Z is the total impedance of the sample.

3. Results and discussion

3.1. Frequency and temperature dependence

The frequency and temperature dependence of the ac conductivity $\sigma_{\mathrm{ac}}(\omega)$, the dielectric constant ε_1, and the dielectric loss ε_2 were studied for In_2Te_3 thin films of different thicknesses 170–300 nm, in the frequency range 100 Hz–100 kHz and in the temperature range 303–373 K to understand the conduction process in the In_2Te_3 system.

The variation of the ac electrical conductivity with the frequency of In_2Te_3 thin film of thickness 170 nm as an example is shown in Fig. 1 for several fixed temperatures. It is clear from the figure that $\sigma_{\mathrm{ac}}(\omega)$ increases linearly with increasing frequency according to the equation [17]

$$\sigma_{\mathrm{ac}}(\omega) = A\omega^s, \tag{5}$$

where s is the frequency exponent, and ω — the angular frequency. Values of s were calculated from the slopes of the linear lines of this figure. It is clear also from the figure that s is independent of the temperature in the investigated range, and it was found that $s = 0.67$.

Figure 2 shows the relation between the ac conductivity $\sigma_{\mathrm{ac}}(\omega)$ and the temperature at different fixed frequencies of In_2Te_3 thin film of thickness 240 nm. It is clear from this figure that $\sigma_{\mathrm{ac}}(\omega)$ increases linearly with the absolute temperature according to the well-known equation

$$\sigma = \sigma_0 \exp\left[-\Delta E_\sigma(\omega)/\mathrm{kT}\right], \tag{6}$$

this suggested that the ac conductivity is a thermally activated process. The ac conduction activation energy $\Delta E_\sigma(\omega)$ is calculated for different frequencies from the slopes of these lines. There is no significant variation in the value of $\Delta E_\sigma(\omega)$ and this means that $\Delta E_\sigma(\omega)$ is frequency independent, its average calculated value was found to be 0.065 eV.

The QMT model [18] predicts a linear temperature dependence of $\sigma_{\mathrm{ac}}(\omega)$ and the exponent s is almost equal to 0.8 and increases slightly or independent of the temperature which is given by [17]

$$S = 1 - 4/\ln(1/\omega\tau_0), \tag{7}$$

where τ_0 is a characteristic relaxation time. This means that our experimental results agree with the QMT model, so the frequency dependence of $\sigma_{\mathrm{ac}}(\omega)$ for In_2Te_3 can be explained in terms of QMT model.

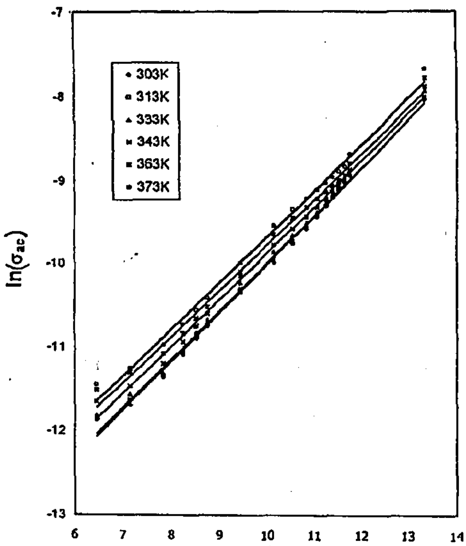

Fig. 1

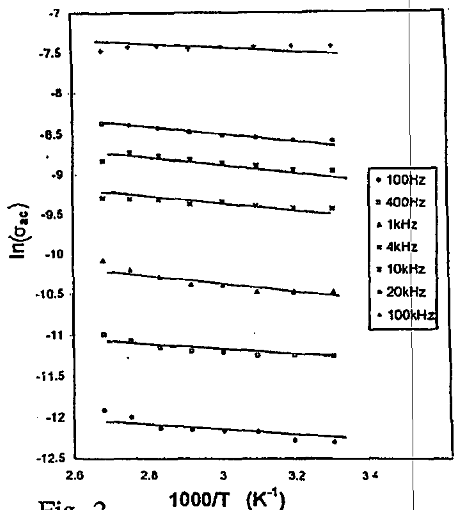

Fig. 2

Fig. 1. Frequency dependence of ac conductivity $\sigma_{ac}(\omega)$ for the as-deposited In_2Te_3 film at different constant temperatures with thickness 170 nm.
Fig. 2. The ac conductivity $\sigma_{ac}(\omega)$ dependence on the temperature at different fixed frequencies of the as-deposited In_2Te_3 film of thickness 240 nm.

According to QMT model, the ac conductivity $\sigma_{ac}(\omega)$ is related to the density of states $N(E_F)$ at the Fermi level by the following equation [19]:

$$\sigma_{ac}(\omega) = (\pi/3)[N(E_F)]^2 kTe^2\alpha^{-5}\ln(\nu_{ph}/\omega)]^4, \qquad (8)$$

where T is the temperature, $N(E_F)$ — the density of defect states at the Fermi energy E_F, ν_{ph} is the phonon frequency and α^{-1} — the spatial extent of the localized wave function. By assuming $\nu_{ph} = 10^{12}$ s^{-1} and $\alpha^{-1} = 10$ Å [9], the density of states is calculated. The calculated values of $N(E_F)$ of In_2Te_3 thin film of thickness 170 nm as an example at different temperatures are listed in Table. It is clear from Table that $N(E_F)$ has values in the order of 10^{19} eV^{-1} cm^{-3} which decreases with the frequency and increases with the temperature.

The variation of the dielectric constant ε_1 with the frequency at different constant temperatures for In_2Te_3 film with thickness 220 nm as an example is shown in Fig. 3. The dielectric constant ε_1 decreases with increasing frequency. This decrease is more sharp at lower frequency and higher temperature and can be attributed to the fact that, at low frequencies, ε_1 for polar material is due to the contribution of multicomponents of polarizability [20, 21] (electronic, ionic, orientational and interfacial polarizability). When the frequency increases, the dipoles will no longer be able to rotate sufficiently rapidly, so ε_1 begins to decrease approaching a constant value at high frequencies. This value is due to interfacial polarization and is typical of nonpolar material.

It is clear also from Fig. 3 that the dielectric constant ε_1 increases with the increase in temperature for all the investigated range of frequency. The increase in

TABLE

The dependence of the density of states $N(E_F)$ on the frequency, working temperature and annealing temperature.

Frequency [kHz]	$N(E_F) \times 10^{19}$ [eV^{-1} cm^{-3}]								
	Measuring temperature [K]						Annealing temperature*		
	303	313	333	343	363	373	As-deposited	423 K	473 K
0.1	5.10	5.30	5.45	5.75	6.00	6.10	4.00	4.96	5.90
0.4	3.95	4.03	4.10	4.20	4.28	4.42	3.90	4.90	5.63
1	3.75	3.77	3.80	3.90	3.97	4.15	3.87	4.82	5.50
4	3.27	3.30	3.32	3.43	3.60	3.72	3.70	4.60	5.06
10	3.08	3.15	3.13	3.17	3.22	3.35	3.45	4.20	4.63
20	2.88	2.91	2.95	3.00	3.04	3.10	3.30	4.03	4.50
100	2.62	2.78	2.70	2.75	2.8	2.92	3.20	3.70	4.00

*$N(E_F)$ of annealed films was calculated at working temperature of 303 K.

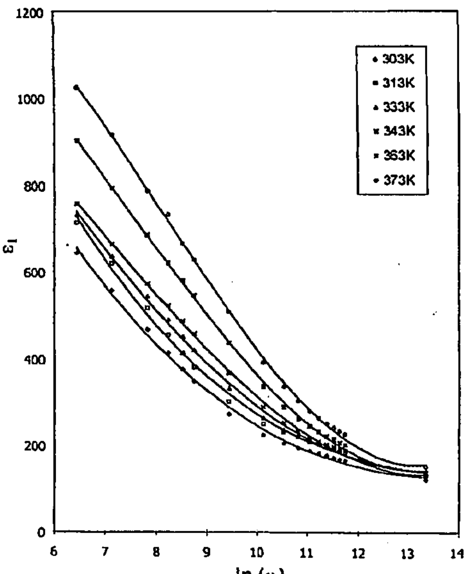

Fig. 3

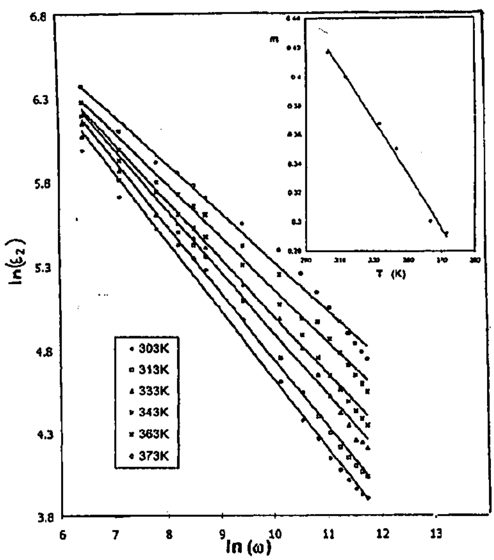

Fig. 4

Fig. 3. The dielectric constant ε_1 dependence on the frequency at different constant temperatures for the as-deposited In_2Te_3 film of thickness 220 nm.
Fig. 4. The dependence $\ln \varepsilon_2$ vs. $\ln \omega$ for the as-deposited In_2Te_3 film of thickness 220 nm at different constant temperatures; the inset shows the temperature dependence of the calculated values of m for the same film.

ε_1 with the temperature is more clear at lower frequencies and can be attributed to the fact that the dipoles in polar material cannot orient themselves at low temperatures and the orientation of dipoles is facilitated when the temperature is raised which increases the value of the dielectric constant ε_1.

The variation in the dielectric loss ε_2 as a function of temperature and frequency is similar to the results obtained for the dielectric constant ε_1, but the decrease in ε_2 with the frequency is higher at low temperature. This is clear from Fig. 4 which represents the relation between the dielectric loss ε_2 and the frequency at different constant temperatures as $\ln(\varepsilon_2)$ vs. $\ln(\omega)$ for a thin film with thickness 220 nm as an example according to the relation [22]

$$\varepsilon_2 = A\omega^m, \tag{9}$$

where A is a constant. The power m was calculated from the negative slopes of the obtained straight lines of Fig. 4 for different temperatures and is represented as a function of temperature in the inset of Fig. 4. It is shown that m decreases linearly with the increase in temperature.

It is also clear from Fig. 4 that the dielectric loss ε_2 increases with temperature. This increase is more clear at higher frequencies and can be explained by Stevels [23] who divided the relaxation phenomena into three parts: conduction losses, dipolar losses and vibrational losses. At low temperatures, conduction losses have minimum value since it is proportional to σ/ω and σ increases with increasing temperature, so the conduction losses increase which increases the value of dielectric loss ε_2.

3.2. Effect of annealing

To illustrate the effect of annealing on the ac electrical conductivity and the dielectric constants $\varepsilon_1, \varepsilon_2$ of In_2Te_3 thin films, several films of In_2Te_3 with different thicknesses were annealed at the temperature 373, 423, and 473 K for four hours. X-ray diffraction patterns indicate that In_2Te_3 thin films annealed at 423 K and then at 473 K have a polycrystalline structure and the degree of crystallinity increases by increasing the annealing temperature as shown in Fig. 5. During the process of annealing, the atomic orientation to the planes of the as-deposited In_2Te_3 becomes easier and besides, more reflecting planes appear indicating polycrystalline growth associating the full crystallinity of the investigated films. Results of careful analysis for this pattern together with JCPDS card no. 33-1488 indicates polycrystalline nature of β-In_2Te_3. The composition of the annealed investigated films was checked by energy dispersive X-ray (EDX) spectroscopy and it was found to be the same as that of the as-deposited films.

For annealed films of different thicknesses, no distinguishable changes in the ac electrical conductivity, dielectric constant ε_1, and dielectric loss ε_2 were observed for films annealed at 373 K in comparison with the as-deposited films. However, the films annealed at 423 and 473 K show significant variation. The variation of ac conductivity with the annealing temperature is shown in Figs. 6 and 7.

Figure 6 shows the frequency dependence of $\sigma_{ac}(\omega)$ for film annealed at 423 K, then at 473 K (t = 300 nm) as an example. It is clear from this figure that at any frequency, $\sigma_{ac}(\omega)$ increases with increasing the annealing temperature and the same results are obtained for all measuring temperatures. The calculated

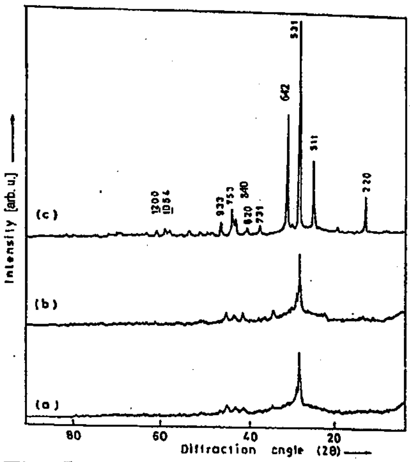

Fig. 5

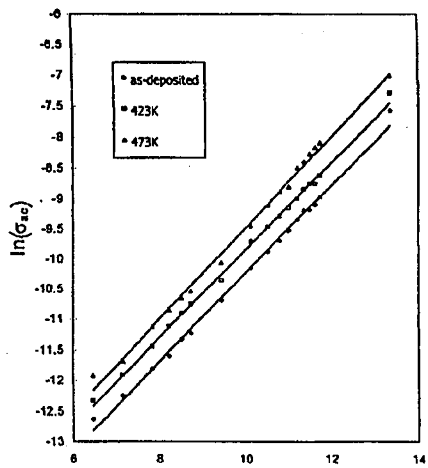

Fig. 6

Fig. 5. X-ray diffraction pattern of In_2Te_3 thin films (*a*) as-deposited, (*b*) annealed at 423 K, (*c*) annealed at 473 K.

Fig. 6. Frequency dependence of ac conductivity $\sigma_{ac}(\omega)$ measured at room temperature for In_2Te_3 thin film of thickness 300 nm annealed at different temperatures.

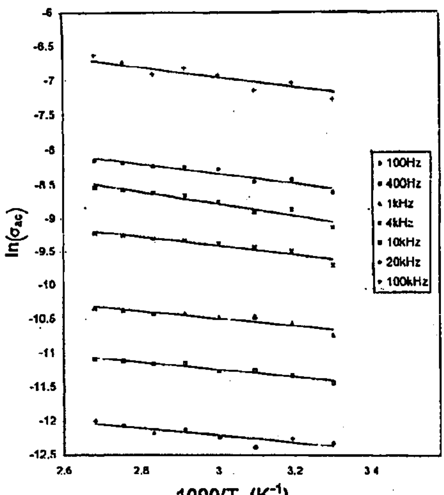

Fig. 7

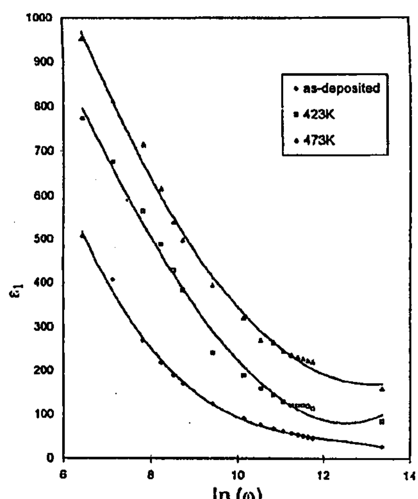

Fig. 8

Fig. 7. Temperature dependence of the ac conductivity for In_2Te_3 film of thickness 300 nm annealed at 423 K at different fixed frequencies.

Fig. 8. Frequency dependence of the dielectric constant ε_1 measured at room temperature for In_2Te_3 film of thickness 300 nm annealed at different temperatures.

values of s show that it is independent of the thickness, measuring and annealing temperature, and have the same values as that of the as-deposited films.

The temperature dependence of $\sigma_{ac}(\omega)$ for In_2Te_3 film with thickness 300 nm annealed at 423 K is shown in Fig. 7 at different constant frequencies. It is clear from this figure that $\sigma_{ac}(w)$ for the annealed films increase linearly with increasing temperature according to Eq. (8). The calculated values of $\Delta E_\sigma(\omega)$ are found to be independent of the thickness and frequency in the investigated range. Its average value is found to be 0.054 eV and 0.043 eV for films annealed at 423 and 473 K respectively and this means that $\Delta E_\sigma(\omega)$ decreases with increasing the annealing temperature and this is due to the increase in the degree of crystallinity to a more ordered state by annealing. It is also clear from Fig. 8 that $\sigma_{ac}(\omega)$ increases with the increase in frequency.

The calculated values of $N(E_F)$ for films annealed at 423 and 473 K are listed also in Table. It is clear that $N(E_F)$ increase with the annealing temperature at any frequency.

The change of the dielectric constant ε_1 and the dielectric loss ε_2 with the annealing temperature is shown in Figs. 8 and 9 which show the frequency dependence of the room temperature ε_1 and ε_2 for In_2Te_3 films with thicknesses 300 and 240 nm respectively annealed at different temperatures. It is clear from these figures that ε_1 and ε_2 increase with annealing temperature at any frequency and decrease as the frequency increases. This is also attributed to the increase in the degree of crystallinity.

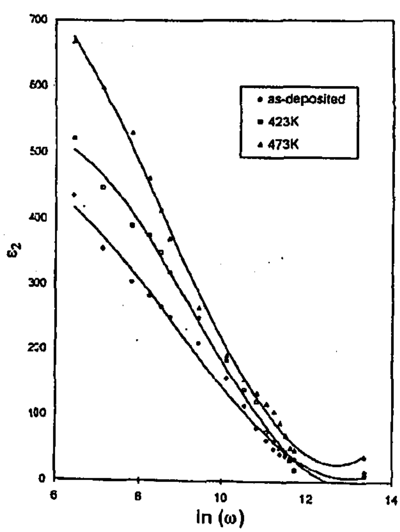

Fig. 9. Frequency dependence of the dielectric loss ε_2 measured at room temperature for In_2Te_3 film of thickness 240 nm annealed at different temperatures.

In conclusion, the data of ac conductivity and dielectric properties for as--deposited and annealed In_2Te_3 films can be explained on the basis of the QMT models which assumed that carrier motion occurs through quantum-mechanical tunneling between localized (defect) states near the Fermi level.

References

[1] K. Kambas, J. Spyridelis, *Mater. Res Bull.* **13**, 653 (1978).

[2] C. Julien, M. Eddrief, K. Kambas, M. Balkanski, *Thin Solid Films* **137**, 27 (1986).

[3] A.I. Zaslavskii, N.F. Kartenko, Z.A. Karachentseva, *Sov. Phys.-Solid State* **13**, 2152 (1972).

[4] A.I. Zaslavskii, V.M. Sergeeva, *Sov. Phys.-Solid State* **2**, 2556 (1961).

[5] G.L. Bleris, T. Karakostas, J. Stoemenos, N.A. Economou, *Phys. Status Solidi A* **34**, 243 (1976).

[6] T. Karakostas, N.A. Economou, *Phys. Status Solidi A* **31**, 89 (1975).

[7] G.L. Bleris, T. Karakostas, N.A. Economou, R. Deridder, *Phys. Status Solidi A* **50**, 579 (1978).

[8] V.M. Kosevich, A.A. Sokal, A.D. Barrinok, *Sov. Phys.-Crystallogr.* **29**, 212 (1984).

[9] A. Zahab, M. Abd-Lefdil, M. Cadene, *Phys. Status Solidi A* **117**, K 103 (1990).

[10] A. Zahab, M. Abd-Lefdil, M. Cadene, *Phys. Status Solidi A* **115**, 491 (1989).

[11] M.A. Afifi, N.A. Hegab, A.E. Bekheet, *Vacuum* **47**, 265 (1996).

[12] N.A. Hegab, A.E. Bekheet, M.A. Afifi, A.E. El-Shazly, *Appl. Phys. A* **66**, 235 (1998).

[13] N.A. Hegab, M.A. Afifi, A.E. El-Shazly, A.E. Bekheet, *J. Mater. Sci.* **33**, 2441 (1998).

[14] M. Pollak, T.H. Geballe, *Phys. Rev. B* **22**, 1742 (1961).

[15] D.P. Singh, K.D. Kundra, *J. Mater. Sci. Lett.* **8**, 524 (1989).

[16] A.K. Shavna, K.L. Bahata, *J. Non.-Cryst. Solids* **109**, 95 (1989).

[17] A. Ghosh, *Phys. Rev. B* **42**, 5665 (1990).

[18] A. Ghosh, *Phys. Rev. B.* **41**, 1479 (1990).

[19] I.G. Austin, N.F. Mott, *Adv. Phys.* **18**, 41 (1969).

[20] B. Tareev, *Physics of Dielectric Materials*, Mir, Moscow 1975.

[21] A.A. Zaky, R. Hawley, *Dielectric Solids*, Routledge & Kegan Paul Ltd., London 1970, p. 16.

[22] J.S. Giuntini, J.V. Zanchetta, *J. Non.-Cryst. Solids* **45**, 57 (1981).

[23] J.M. Stevels, *Handbuch der Physik*, Ed. S. Flügge, Vol. XX, Springer, Berlin 1957, p. 350.