ELECTRICAL AND DIELECTRIC PROPERTIES OF AMORPHOUS $Ge_1Se_{1.35}Tl_{0.1}$ FILMS

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The temperature dependence of the DC and AC electrical conductivity were measured for $Ge_1Se_{1.35}Tl_{0.1}$ films. The value of DC electrical conduction energy ΔE_σ does not depend on film thickness in the investigated range with mean value of 0.72 eV. The AC conductivity σ_{AC} is related to frequency by the expression $\sigma_{AC} = A\omega^S$, where S is the frequency exponent which decreases linearly with increasing temperature. This can be explained in terms of the pair (bipolaron) correlated barrier hopping model suggested by Elliott. The frequency and temperature dependence of real and imaginary parts of the dielectric constant were studied for $Ge_1Se_{1.35}Tl_{0.1}$ films. The dielectric constant (real part) and the dielectric loss (imaginary part) increase with increasing temperature and decrease with increasing frequency in the investigated range of frequency and temperature. The maximum barrier height W_M can be calculated according to the Giuntini equation at different temperatures. The obtained value of W_M is in good agreement with the theory of hopping of charge carriers over a potential barrier as suggested by Elliott in case of chalcogenide glasses.

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1. Introduction

The nature of electron transport in amorphous materials has attracted great deal of attention. Chalcogenide glasses have been intensively investigated because of their technologically important electrical properties. The addition of thallium to chalcogenide glasses is generally accompanied by a marked change in their structural and physical properties [1–5], and attracted much attention because of their application in acousto-optical devices [6, 7].

Dielectric relaxation studies are important to understand the nature and the origin of dielectric losses which, in turn, may be useful in the determination of the structure and defects in solids. Also, the study of frequency dependent electrical conductivity of amorphous compounds is important to explain the mechanisms of conduction in these substances [8, 9]. This leads to adapting and to elaborating models allowing the electronic properties of several substances to be

described [10, 11]. Generally, such solids are correctly represented by localized states, simulating conducting domains in totally amorphous matrix [10, 12, 13].

A model proposed in the study of DC conductivity [10, 14] takes into account the assumption expressed by Mott et al. [15], according to which, when the sample is placed in an electrical field, electron hopping takes place between localized sites. The charge carriers, moving between these sites, hop from a donor to an acceptor state. In that respect each pair of sites forms a dipole. The described model thus supposes that each dipole has a relaxation time depending on its activation energy; the latter can be essentially attributed to the existence of a potential barrier, over which the carriers must hop [16]. This potential barrier, as proposed by Elliott, is due to the Coulombic interaction between neighboring sites forming a dipole [14, 17].

The paper aims at the study of electrical properties of $Ge_1Se_{1.35}Tl_{0.1}$ in thin film forms. This study includes DC and AC conductivity, determination of the dielectric constant ε', dielectric losses ε'', and the maximum barrier height W_M.

2. Experimental details

Bulk $Ge_1Se_{1.35}Tl_{0.1}$ glass was prepared under vacuum of 10^{-6} Torr by quenching from the melts [2]. Amorphous thin films of different thickness were deposited under vacuum of 10^{-5} Torr (or less), by the thermal evaporation technique, using a coating unit (Edward 306 A), between two evaporated aluminium electrodes (sandwiched) on standard glass substrates. The film thickness was controlled during deposition by using an hf crystal monitor (Edward FTM5). Subsequently, it was measured after deposition by Tolansky's method using multiple-beam Fizeau fringes to ensure an accurate value of the thickness.

The film amorphicity was carefully checked for all investigated compositions by absence of any diffraction lines in their X-ray patterns. The films composition and homogeneity were checked by energy dispersive X-ray spectroscopy (EDX), where very satisfactory results were obtained for the amorphous films deposited at rate of 100 Å/s.

The measurements of film resistance were carried out using a simple electrical circuit provided with a digital electrometer (Keithley type E616A) and a micro digital multimeter (TE 924) to measure the respective potential drop and the current passing through the sample. Also the circuit was provided with a heater, for sample heating, and a calibrated chromel–alumel thermocouple through an automatic temperature central system [18]. The dielectric constant, dielectric loss tangent, and the AC conductivity as a function of temperature and frequency were measured using a programmable automatic RCL meter (PM 6304 Philips). The AC measurements were carried out in the frequency range (50 Hz–100 kHz).

3. Results and discussion

The temperature dependence of the measured total conductivity σ_T observed using an AC bridge was studied in the frequency range (500 Hz–20 kHz) for

two films of thicknesses 300 and 180 nm respectively, and the temperature range (373–443 K). It is clear that σ_T increases with increasing temperature.

The value of AC conductivity σ_{AC} can be deduced from the measurements of the total conductivity $\sigma_{AC} = \sigma_T - \sigma_{DC}$ [19]. The temperature dependence of the DC conductivity, σ_{DC}, of glassy $Ge_1Se_{1.35}Tl_{0.1}$ can be expressed in the form $\sigma_{DC} = \sigma_0 \exp(-\Delta E_\sigma/k_B T)$, where E_σ is the activation energy, σ_0 is the pre-exponential factor and k_B is Boltzmann's constant. The average value of ΔE_σ is found to be 0.72 ± 0.02 eV for investigated films. That is, the mean value of ΔE_σ is approximately independent of film thickness for amorphous $Ge_1Se_{1.35}Tl_{0.1}$ films in the investigated temperature range.

It is well known in chalcogenide glass semiconductors that σ_{AC} is expressed by the empirical equation [20]:

$$\sigma_{AC} = A\omega^S, \tag{1}$$

where A is a constant and ω — the regular frequency of AC field. The exponent S denotes the frequency dependence of σ_{AC}. Therefore, the results of measurements of AC conductivity for the studied composition (thickness 180 nm as an example) at different temperatures and frequencies is shown in Fig. 1. It is clear from Fig. 1 that σ_{AC} increases linearly with the increasing frequency that σ_{AC} increases with the increasing temperature. The values of S in Eq. (1) have been calculated from the slopes of the linear lines in Fig. 1. The temperature dependence of S for amorphous $Ge_1Se_{1.35}Tl_{0.1}$ films is given in Fig. 2. It is clear that S decreases linearly as the temperature increases from 0.48, 0.533 at 373 K to 0.237, 0.30 at 453 K for samples 300 and 180 nm, respectively.

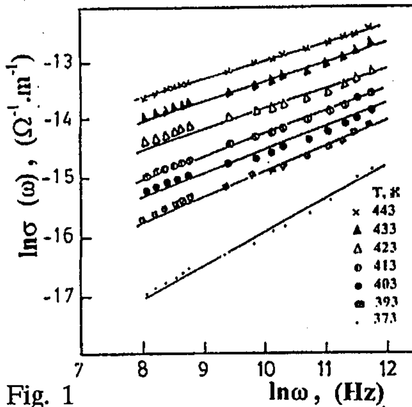

Fig. 1

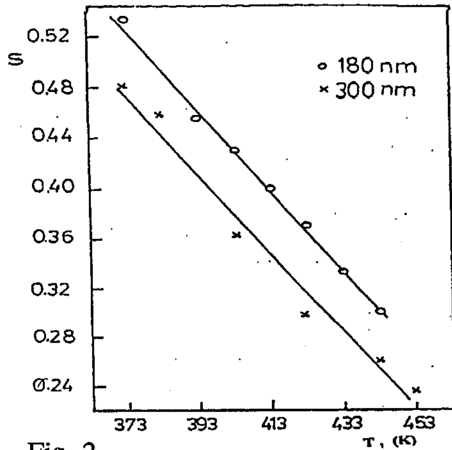

Fig. 2

Fig. 1. Frequency dependence of AC conductivity σ_{AC} for $Ge_1Se_{1.35}Tl_{0.1}$ film of thickness 180 nm.

Fig. 2. Temperature dependence of the frequency exponent (S) for $Ge_1Se_{1.35}Tl_{0.1}$ films of thicknesses 300 and 180 nm.

The observed trend of frequency and temperature dependence of AC conductivity and frequency exponent S can be accounted in terms of the pair (bipolaron) correlated barrier hopping (CBH) model suggested by Elliott [21]. According to CBH model, $\sigma(\omega)$ should have negative temperature dependence of the frequency exponent S. This is in good agreement with the present results. The frequency dependence of AC conductivity, which rises almost linearly with frequency, is most likely due to hopping of two electrons between two pairs of localized state at the Fermi level. The fact that S is temperature dependent indicates that the bipolaron conduction is thermally activated process, which is phonon assisted.

According to the quantum mechanical tunneling (QMT) model proposed by Pollak and Geballe [22] the exponent S is almost equal to 0.8 and increasing slightly while increasing temperature. Therefore, the QMT model is not applicable to the investigated chalcogenide glasses. Thus, the CBH model can successfully explain the obtained frequency dependence of conductivity.

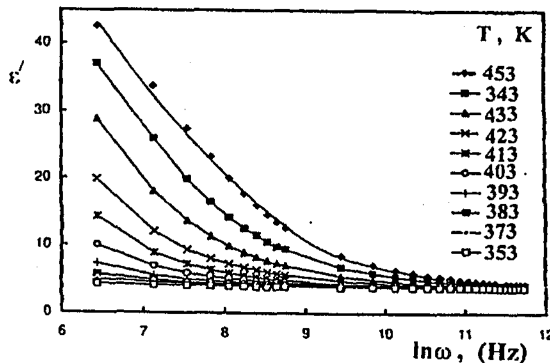

Fig. 3. Frequency dependence of the dielectric constant ε' for $Ge_1Se_{1.35}Tl_{0.1}$ film of thickness 180 nm at different temperatures.

The temperature and frequency dependence of the dielectric constant ε' was studied for two $Ge_1Se_{1.35}Tl_{0.1}$ films of thicknesses 300 and 180 nm in the temperature range (353–453 K) and in the frequency range (100 Hz–20 kHz). This behaviour can be clarified by plotting ε' against $\ln\omega$ for the investigated films at different temperatures as shown in Fig. 3 for sample of thickness 180 nm as representative example. It is clear from the figure that the dielectric constant ε' increases as the temperature increases for all frequencies in the investigated range. First the increase is linear with increasing frequency, and then it becomes non-linear at higher values of temperature. At lower temperature, the variation of ε' vs. frequency are approximately constant, then ε' begins to increase towards the higher values of temperature.

The variation of ε' with frequency and temperature can be attributed to the fact that at low frequencies and high temperatures, ε' is due to the contribution of multicomponent of polarizability (electronic, ionic, orientation, and interfacial polarizability) [23], when the frequency is increased, the dipoles will no longer be able to rotate sufficiently rapidly, so that their oscillations will begin to lag behind those

of the field. As the frequency is further raised the permanent dipoles, if present in the medium, are completely unable to follow the field and the orientational polarization times. Thus the value of ε' is decreased at low temperatures because only the space charge (interfacial) polarization contributes to the polarization.

Also, the temperature and frequency dependence of the imaginary part of dielectric constant ε'' (dielectric loss) was studied for the two samples under test at the same condition. In the investigated amorphous $Ge_1Se_{1.35}Tl_{0.1}$ films, it is found that ε'' follows a power law frequency [24]:

$$\varepsilon'' = A\omega^m, \tag{2}$$

where A is a constant and m is a function of temperature.

Figure 4 presents the relation between $\ln\varepsilon''$ and $\ln\omega$ respectively, for two investigated films at various temperatures. It is clear that the dielectric loss ε'' increases as the temperature increases for all frequencies in the investigated range. The behaviour of ε'' with temperature can be explained by Stevels [24] who divided the relaxation phenomena into three parts: conduction losses, dipole relaxation losses, and deformational losses (vibrational losses). These losses involve the migration of ions over large distances. This motion is the same as that occurring under direct current conditions. The ions jump over the highest barriers in the network. As the ions move, they give some of their energy to the lattice as a heat and the amount of heat lost per cycle is proportional to σ/ω. The conduction loss increases with increasing temperature. At low temperatures, conduction, dipole and vibration losses have minimum value. However, at higher temperatures conduction loss, dipole loss, and vibrational loss all contribute to the dielectric loss.

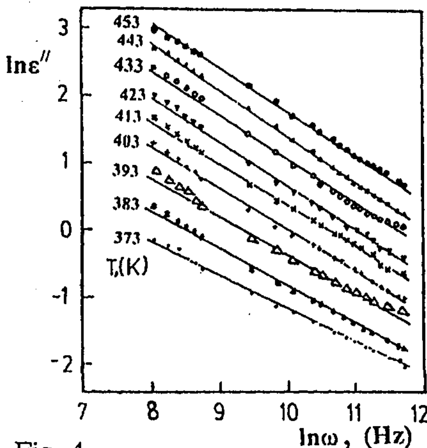

Fig. 4

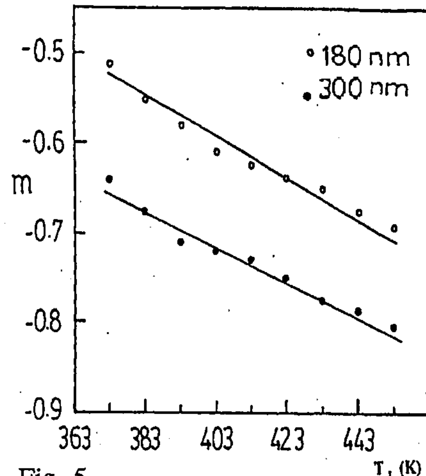

Fig. 5

Fig. 4. Plot of $\ln\varepsilon''$ versus $\ln\omega$ for $Ge_1Se_{1.35}Tl_{0.1}$ film of thicknesses 180 nm at different temperatures.

Fig. 5. Temperature dependence of the experimental values of m for $Ge_1Se_{1.35}Tl_{0.1}$ films of thicknesses 300 and 180 nm.

The frequency behaviour of ε'' can be attributed to the fact that the migration of ions in glass is the main source of the dielectric loss at low frequencies. Accordingly, the dielectric loss at low and moderate frequencies is characterized by high values due to the contribution of ion jump and conduction loss of ion migration, in addition to the ion polarization loss. However, at high frequencies the ion vibrations may be the only source of dielectric loss. The power (m) is calculated from the negative slopes of the obtained straight lines of Fig. 5. Thus values of m are negative for amorphous $Ge_1Se_{1.35}Tl_{0.1}$ films at all temperature values in the investigated range. Figure 5 shows the relation between m and temperature for both investigated films. It is clear that the parameter m decreases linearly with temperature in amorphous $Ge_1Se_{1.35}Tl_{0.1}$ films in the investigated range.

According to Giuntini et al. [25], ε'' at a particular frequency in the temperature range where dielectric dispersion occurs, is given by

$$\varepsilon''(\omega) = (\varepsilon_0 - \varepsilon_\infty)2\pi^2 N(ne^2/\varepsilon_0)^3 k_B T \tau_0^m W_M^{-4} \omega^m, \tag{3}$$

with $m = -4k_BT/W_M$; n is the number of electrons that hop; N is the concentration of localized sites; ε_0 is the static dielectric constant and ε_∞ — the dielectric constant at "infinity high" frequencies; W_M is the energy required to move the electron from one site to the infinite (maximum barrier height).

$$m = -4k_BT/W_M. \tag{4}$$

Values of W_M were calculated from Eq. (3) for the investigated films. The obtained values of W_M are 0.2 ± 0.01 eV and 0.23 ± 0.01 eV for the investigated films of thicknesses 300 and 180 nm, respectively, in the investigated temperature range.

According to Eq. (3), ε'' should follow a power law with frequency, i.e., $\varepsilon'' = A\omega^m$, where m should be negative and linear with T as given by Eq. (4).

The present results are in good agreement with the CBH model suggested by Elliott [21] in case of chalcogenide glasses, i.e., the charge carriers hop over potential barrier.

4. Conclusion

In conclusion, we have studied the frequency and temperature dependence on (i) AC electrical conductivity and (ii) the dielectric constant, for $Ge_1Se_{1.35}Tl_{0.1}$ amorphous semiconductor in temperature range 353–453 K and frequency range (100 Hz–20 kHz).

The AC conductivity σ_{AC} is related to frequency according to the expression $\sigma_{AC} = A\omega^S$, where the exponent S decreases linearly with increasing temperature, which can be explained in terms of bipolar CBH model suggested by Elliott.

The real and imaginary parts of the dielectric constant increase and decrease with increasing temperature and frequency respectively.

The maximum barrier height W_M can be calculated according to the Giuntini equation.

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