

AC conductivity and dielectric properties of $\text{Se}_{70}\text{Ge}_{30-x}\text{M}_x$ { $x=0\&5$ and $\text{M}=\text{Ag}, \text{Cd}$ or Pb } amorphous films

E. G. EL-METWALLY*, M. FADEL, A. M. SHAKRA, M. A. AFIFI

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

The temperature and frequency dependences of ac conductivity $\sigma_{ac}(\omega)$, dielectric constant $\epsilon'(\omega)$ and dielectric loss $\epsilon''(\omega)$ are studied for $\text{Se}_{70}\text{Ge}_{30}$, $\text{Se}_{70}\text{Ge}_{25}\text{Ag}_5$, $\text{Se}_{70}\text{Ge}_{25}\text{Cd}_5$ and $\text{Se}_{70}\text{Ge}_{25}\text{Pb}_5$ amorphous films, in the temperature range 313-453 K and frequency range 0.1-100 kHz. The obtained data reveals that $\sigma_{ac}(\omega)$ for all investigated compositions is temperature dependent, and obeys $A\omega^S$ law. The temperature dependences of $\sigma_{ac}(\omega)$ and the frequency exponent S are explained on the basis of correlated barrier hopping CBH model. Analysis of the results reveals that the electronic conduction for all investigated compositions takes place via bipolaron hopping at temperatures range 313K – 363K, and single polaron hopping at temperatures range [373K – 453K]. Also, it has been observed that $\epsilon'(\omega)$ and $\epsilon''(\omega)$ exhibit strong temperature and frequency dependencies for all investigated compositions.

(Received April 18, 2008; accepted June 4, 2008)

Keywords: Se-Ge system doped by Ag, Cd, Pb, Amorphous films, a.c. conductivity

1. Introduction

Measurement of ac conductivity is a powerful tool for obtaining information about defect states in amorphous semiconductors [1,2]. The effect of impurities on the transport and structural properties has been an important issue since the discovery of these glasses. The addition of a metallic impurity to amorphous chalcogenides enhances their conductivity and produces a significant decrease in the activation energy for conduction, making them more suitable for device applications. For example, chalcogenide glasses containing silver find applications in photolithography, optical imaging, information storage and micro lithography [3]. Some metallic impurities [Pb,Bi] have been found to change the conduction from p to n-type in some Ge-based chalcogenide glasses [4]. However several workers studied the structural and electrical properties of SeGeM { $\text{M}=\text{Ag}, \text{Cd}$ or Pb } [5-12].

Ac conductivity $\sigma_{ac}(\omega)$ is usually expressed as $\sigma_{ac}(\omega) = A\omega^S$, where ω is the angular frequency, A is a constant and S is the frequency exponent. A model for the frequency dependent conductivity was first proposed by Pollak and Geballe(1961) [13]. They assumed that conduction in doped semiconductors arises from tunneling between neighbouring donor impurity states; this has been called the quantum mechanical tunneling QMT model. According to QMT model, the exponent S is either temperature independent or an increasing function of temperature. Elliot(1977) [1] suggested a model which is based on the concept of charged defect states in which, hopping was considered to be over the barrier with states

in the localized band, and called the correlated barrier hopping CBH model. In this model, the electron pair hops from doubly occupied D^+ states to a nearby D^- center over the barrier separating both sites, the barrier height moreover being correlated with intersite separation via the coulombic interaction between centers [14].

Dielectric relaxation studies are important to understand the nature and the origin of dielectric losses, which, in turn, may be useful in determination the structure and defects in solids. The dielectric properties were also considered as important complementary efforts contributing to a better understanding of the mechanism of transport in such materials.

In the present work, we have reported the temperature, frequency and thickness dependence of Ac conductivity $\sigma_{ac}(\omega)$, dielectric constant ϵ' and dielectric loss ϵ'' for $\text{Se}_{70}\text{Ge}_{30}$, $\text{Se}_{70}\text{Ge}_{25}\text{Ag}_5$, $\text{Se}_{70}\text{Ge}_{25}\text{Cd}_5$ and $\text{Se}_{70}\text{Ge}_{25}\text{Pb}_5$ amorphous films.

2. Experimental details

$\text{Se}_{70}\text{Ge}_{30}$, $\text{Se}_{70}\text{Ge}_{25}\text{Ag}_5$, $\text{Se}_{70}\text{Ge}_{25}\text{Cd}_5$ and $\text{Se}_{70}\text{Ge}_{25}\text{Pb}_5$ compositions were synthesized as follows: The elementary constituents of each composition of purity 99.999% were weighed in accordance with their atomic percentage and loaded in a silica tube, which was then sealed under vacuum (10^{-5} Torr). The content of each tube was heated gradually in an oscillatory furnace to 500 K (m.p. of Se) and kept constant for 2 h, then it was raised to 600 K (m.p of Pb or Cd) and kept constant for 2 h and finally it was raised to 1273 K (m.p. of Ge and Ag) and kept constant for

20 h. Long times of synthesis and oscillation of the tube are necessary for the homogeneity of the synthesized compositions. The tube is then quenched in icy water to obtain the compositions in the glassy state.

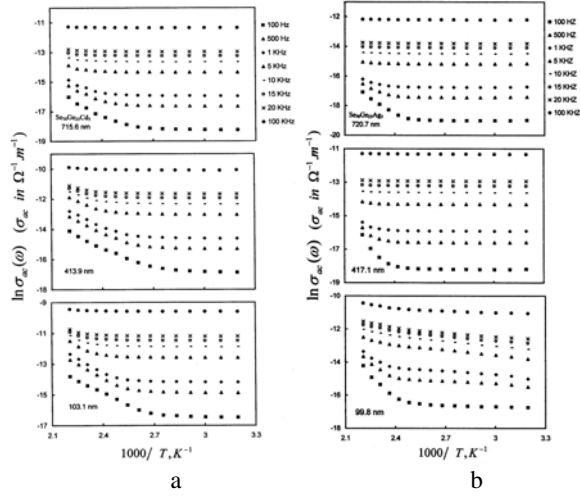


Fig. 1(a) Temperature dependence of ac conductivity $\sigma_{ac}(\omega)$ for $\text{Se}_{70}\text{Ge}_{25}\text{Ag}_5$ films of thicknesses 720.7, 417.1 and 99.8 nm at different frequencies.(b) Temperature dependence of ac conductivity $\sigma_{ac}(\omega)$ for $\text{Se}_{70}\text{Ge}_{25}\text{Cd}_5$ films of thicknesses 715.6, 413.9 and 103.1 nm at different frequencies.

Thin films of the investigated compositions were obtained from bulk samples by a thermal evaporation technique under vacuum on glass substrates. The substrates was fixed onto a rotatable holder (up to 240 rpm) to obtain homogeneous deposited films at a distance of 25 cm above the evaporator. The thickness of film samples was measured during deposition using a thickness monitor (Edward, FTM) and confirmed after deposition by Tolansky's interferometric method [15]. The chemical composition of the investigated samples was checked by energy dispersive X-ray analysis (EDX) using a Joel 5400 scanning electron microscope. The structural identification of the investigated compositions in powder and thin film forms were confirmed by both X-ray diffraction (XRD) and differential thermal analysis (DTA). Thin film samples were sandwiched between two aluminium electrodes as lower and upper electrodes for Ac electrical measurements. All measurements were carried out below the glass transition temperature T_g of each composition.

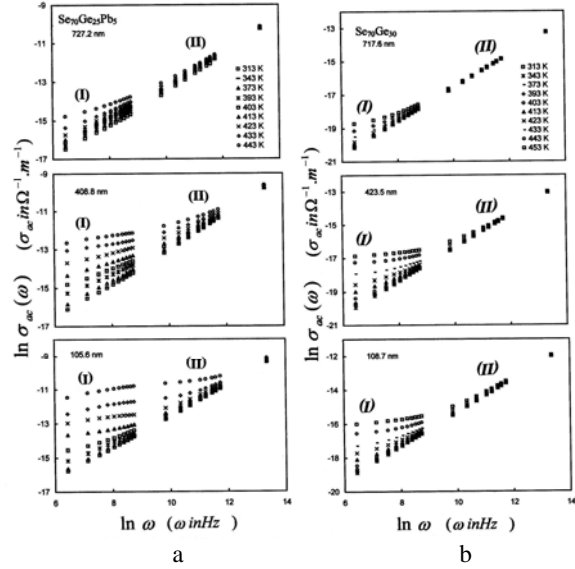


Fig. 2(a) Frequency dependence of ac conductivity $\sigma_{ac}(\omega)$ for $\text{Se}_{70}\text{Ge}_{30}$ films of thicknesses 717.6, 423.5 and 108.7 nm at different temperatures.(b) Frequency dependence of ac conductivity $\sigma_{ac}(\omega)$ for $\text{Se}_{70}\text{Ge}_{25}\text{Pb}_5$ films of thicknesses 727.2, 408.8 and 105.6 nm at different temperatures.

A programmable automatic RCL meter (PM 6304 Philips) was used to measure the electrical impedance Z , the capacitance C and the dielectric loss tangent $\tan \delta$ directly as a function of temperature and frequency. The total conductivity $\sigma_{tot}(\omega)$, $\epsilon'(\omega)$ and $\epsilon''(\omega)$ were calculated for all investigated thin film samples according to :-

$$\sigma_T(\omega) = \frac{t}{ZA} \quad (1)$$

$$\epsilon' = \frac{C}{\epsilon_0} \cdot \frac{t}{A} \quad (2)$$

$$\epsilon'' = \epsilon' \tan \delta \quad (3)$$

where t is the film thickness, A is the cross sectional area of the film and ϵ_0 is the dielectric permittivity of free space.

3. Results and discussion

3.1. AC conductivity of $\text{Se}_{70}\text{Ge}_{30}$, $\text{Se}_{70}\text{Ge}_{25}\text{Ag}_5$, $\text{Se}_{70}\text{Ge}_{25}\text{Cd}_5$ and $\text{Se}_{70}\text{Ge}_{25}\text{Pb}_5$ films.

3.1.1. Temperature and frequency dependence of $\sigma_{ac}(\omega)$.

Temperature dependence of $\sigma_{ac}(\omega)$ [$\sigma_{ac}(\omega) = \sigma_T(\omega) - \sigma_{dc}$] was studied for $\text{Se}_{70}\text{Ge}_{30}$, $\text{Se}_{70}\text{Ge}_{25}\text{Ag}_5$, $\text{Se}_{70}\text{Ge}_{25}\text{Cd}_5$ and $\text{Se}_{70}\text{Ge}_{25}\text{Pb}_5$ films in the

temperature range (313–453 K) for different thicknesses in the range (99.8–727.2 nm), at different frequencies in the range (100 Hz–100 KHz). The obtained results are illustrated as $\ln \sigma_{ac}(\omega)$ against $1000/T$ at different frequencies for $\text{Se}_{70}\text{Ge}_{30}\text{Ag}_5$ and $\text{Se}_{70}\text{Ge}_{25}\text{Cd}_5$ films of different thicknesses as representative examples in Figs.1(a&b) respectively. It is clear from these figures that $\sigma_{ac}(\omega)$ increases slightly with increasing temperature to about 363 K, then it increases linearly with temperature for all investigated compositions. The observed slight increase in $\sigma_{ac}(\omega)$ is found to shift towards higher temperature values as the frequency increased.

The observed variation of $\sigma_{ac}(\omega)$ in temperature range 313K – 363K may be due to conduction takes place via bipolaron hopping between non-randomly distributed defect sites (D^+ and D^-). At temperatures range 373 – 453K, the predominant contributor to conductivity is single polaron hopping between D^0 and D^+ or D^- [16]. It must be noticed here that the obtained results agree with those obtained before [17,18].

A common feature to almost all amorphous semiconductors is a frequency dependent ac conductivity $\sigma_{ac}(\omega)$, that increases, approximately linearly with frequency at least in the frequency range $10 \text{ s}^{-1} < \omega < 10^8 \text{ s}^{-1}$, i.e.[19] :

$$\sigma_{ac}(\omega) = A^{\wedge} \omega^S \quad (4)$$

The frequency dependence of $\sigma_{ac}(\omega)$ is plotted as $\ln \sigma_{ac}(\omega)$ versus $\ln \omega$ at different temperatures for $\text{Se}_{70}\text{Ge}_{30}$ and $\text{Se}_{70}\text{Ge}_{25}\text{Pb}_5$ films of different thicknesses in Figs. 2 (a &b) as representative examples. As observed from these figures, each obtained relation consists of two straight lines in both lower (100 Hz - 1 kHz) and higher (1 kHz - 100 kHz) frequency ranges I and II respectively. In both ranges I and II, $\sigma_{ac}(\omega)$ increases linearly with increasing frequency according to Eq.(4). Values of S and A^{\wedge} were obtained from the slopes of the linear lines of the two parts (I&II), and by extrapolating the obtained lines of $\ln \sigma_{ac}(\omega)$ versus $\ln \omega$ to the y-axis, for all investigated film compositions with different thicknesses. It is observed that the calculated values of S for compositions of different thicknesses, at different temperatures in the studied range are thickness independent, thus the mean value of S was calculated.

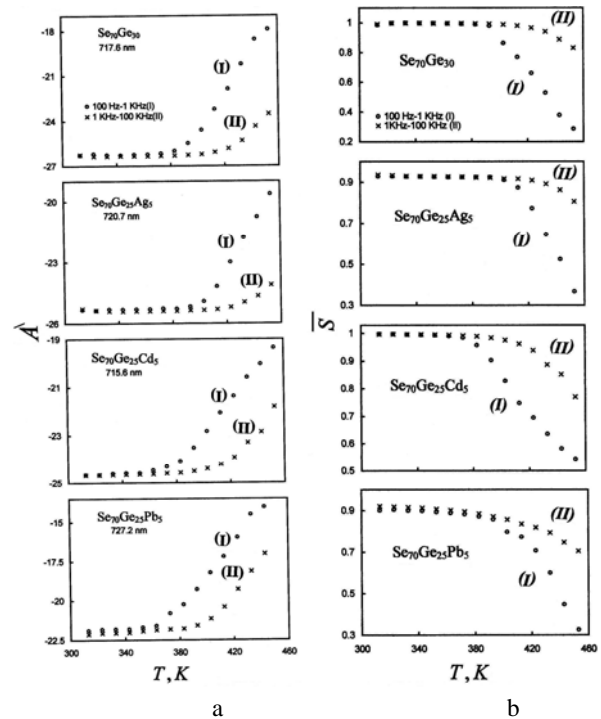


Fig. 3(a) Temperature dependence of the mean value of the frequency exponent \bar{S} for $\text{Se}_{70}\text{Ge}_{30}$, $\text{Se}_{70}\text{Ge}_{25}\text{Ag}_5$, $\text{Se}_{70}\text{Ge}_{25}\text{Cd}_5$ and $\text{Se}_{70}\text{Ge}_{25}\text{Pb}_5$ films. (b) Temperature dependence of A^{\wedge} for $\text{Se}_{70}\text{Ge}_{30}$, $\text{Se}_{70}\text{Ge}_{25}\text{Ag}_5$, $\text{Se}_{70}\text{Ge}_{25}\text{Cd}_5$ and $\text{Se}_{70}\text{Ge}_{25}\text{Pb}_5$ films of nearly the same thicknesses.

Fig.3(a) illustrates the temperature dependence of the mean value of the frequency exponent \bar{S} for the four investigated compositions. Fig.3(b) shows the temperature dependence of A^{\wedge} for the investigated film compositions of approximately the same thickness. It is clear from these figures that, in the temperature range [313-363 K], \bar{S} lies very close to unity, and both \bar{S} and A^{\wedge} are temperature independent. In the temperature range [373-453 K], both \bar{S} and A^{\wedge} depend on temperature, where \bar{S} decreases with increasing temperature while A^{\wedge} increases with temperature. This behaviour is inconsistent with QMT model [20], which doesn't predict a temperature dependence of the frequency exponent \bar{S} but it has a constant value (≈ 0.81). In correlated barrier hopping (CBH) model [1,21,22], the general behaviour of ac conductivity ($\sigma_{ac}(\omega) = A^{\wedge} \omega^S$) for chalcogenide glasses can be subdivided into two categories [21]. In the first both \bar{S} and A^{\wedge} are dependent on temperature, the former tending to unity at low temperatures and both are related to the band gap of the material [1,22]. This behaviour can be explained by the CBH between randomly close pairs of centers forming Non-intimate valence alternation pairs (NVAP's). In the second, \bar{S} lies very close to unity and independent of temperature; A^{\wedge} is also more or less temperature independent. This behaviour can be explained by the CBH between centers forming intimate valence alternation pairs (IVAP's) [23,24]. Thus, the obtained data for the temperature dependence of \bar{S} and A^{\wedge} for the

investigated compositions can be explained on the basis of the CBH model between centers forming (IVAP's) in the temperature range 313-373 K and (NVAP's) in the temperature range 383-453 K. This means that the present experimental results agree well with CBH model.

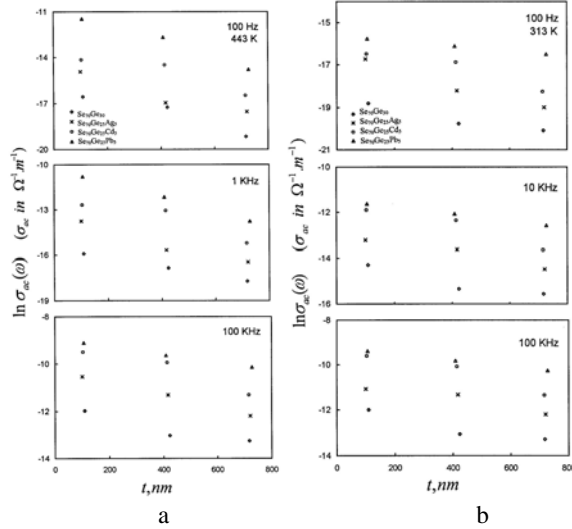


Fig. 4(a) Thickness dependence of ac conductivity $\sigma_{ac}(\omega)$ for $\text{Se}_{70}\text{Ge}_{30}$, $\text{Se}_{70}\text{Ge}_{25}\text{Ag}_5$, $\text{Se}_{70}\text{Ge}_{25}\text{Cd}_5$ and $\text{Se}_{70}\text{Ge}_{25}\text{Pb}_5$ films at room temperature (313K) and different frequencies. (b) Thickness dependence of ac conductivity $\sigma_{ac}(\omega)$ for $\text{Se}_{70}\text{Ge}_{30}$, $\text{Se}_{70}\text{Ge}_{25}\text{Ag}_5$, $\text{Se}_{70}\text{Ge}_{25}\text{Cd}_5$ and $\text{Se}_{70}\text{Ge}_{25}\text{Pb}_5$ films at 443 K and different frequencies.

3.1.2. Thickness dependence of $\sigma_{ac}(\omega)$.

The thickness dependence of $\sigma_{ac}(\omega)$ was studied for the investigated film compositions at room temperature 313 K as well as at the elevated temperature 443 K as representative examples at different frequencies as shown in Figs. 4(a&b). It is clear from these figures that $\sigma_{ac}(\omega)$ decreases with increasing film thickness for all investigated compositions in the investigated ranges of temperature and frequency. Moreover, it is clear also that the addition of the third element Ag, Cd or Pb to $\text{Se}_{70}\text{Ge}_{30}$ increases $\sigma_{ac}(\omega)$ in the investigated range of thickness. The observed increase with Pb is higher than that with Cd which in turn is higher than that with Ag. However, the addition of third element Ag, Cd or Pb to $\text{Se}_{70}\text{Ge}_{30}$ leads to a decrease in the bond energy between the host element Se and the metal additives [25] as given in Table 1.

Table 1. Values of the bond energy of Se-Ge, Se-Ag, Se-Cd and Se-Pb.

Molecule	Se-Ge	Se-Ag	Se-Cd	Se-Pb
Bond energy (eV)	5.08 [26]	3.53 [7]	3.21 [26]	2.71 [27]

3.2. Dielectric properties of $\text{Se}_{70}\text{Ge}_{30}$, $\text{Se}_{70}\text{Ge}_{25}\text{Ag}_5$, $\text{Se}_{70}\text{Ge}_{25}\text{Cd}_5$ and $\text{Se}_{70}\text{Ge}_{25}\text{Pb}_5$ films

3.2.1. Temperature and frequency dependence of the dielectric constant ϵ' .

Temperature and frequency dependence of $\epsilon'(\omega)$ were studied for $\text{Se}_{70}\text{Ge}_{30}$, $\text{Se}_{70}\text{Ge}_{25}\text{Ag}_5$, $\text{Se}_{70}\text{Ge}_{25}\text{Cd}_5$ and $\text{Se}_{70}\text{Ge}_{25}\text{Pb}_5$ films of different thicknesses in the above mentioned investigated ranges of temperature and frequency. Figs.5(a&b) show the temperature dependence of $\epsilon'(\omega)$ at different frequencies for three different thicknesses of $\text{Se}_{70}\text{Ge}_{30}$ and $\text{Se}_{70}\text{Ge}_{25}\text{Ag}_5$ film compositions as representative examples. It is clear from these figures that $\epsilon'(\omega)$ remains almost constant with temperature in the range [313-363 K]. Beyond this range, $\epsilon'(\omega)$ increases with increasing temperature. The increase is higher at lower frequencies. Therefore, $\epsilon'(\omega)$ exhibits strong temperature dependence at higher temperatures and lower frequencies. This type of behaviour has also been reported by various workers in chalcogenide glasses [28-33]. The increase of $\epsilon'(\omega)$ with temperature can be attributed to the fact that dipoles in polar materials can not be oriented at low temperatures (remain frozen), while at high temperatures dipoles can rotate freely as suggested by Srivastava et al. [34], which increases orientational polarization, and hence increases $\epsilon'(\omega)$.

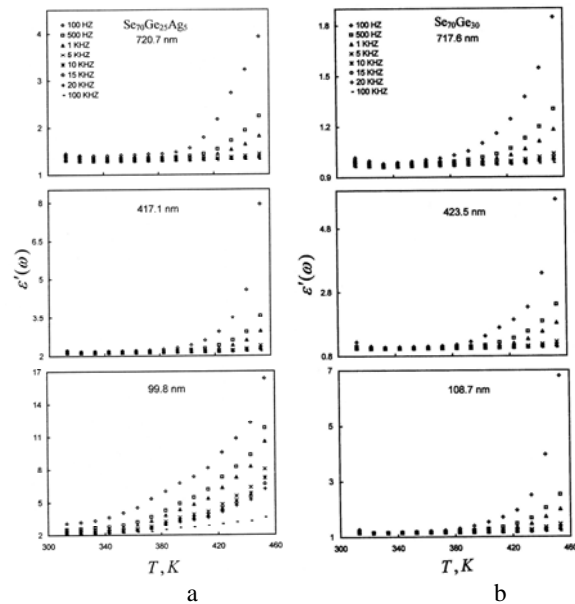


Fig. 5(a) Temperature dependence of the dielectric constant $\epsilon'(\omega)$ for $\text{Se}_{70}\text{Ge}_{30}$ films of thicknesses 717.6, 423.5 and 108.7 nm at different frequencies. (b) Temperature dependence of the dielectric constant $\epsilon'(\omega)$ for $\text{Se}_{70}\text{Ge}_{25}\text{Ag}_5$ films of thicknesses 720.7, 417.1 and 99.8 nm at different frequencies.

Figs. 6(a&b) show the frequency dependence of $\epsilon'(\omega)$ at different temperatures respectively for

$\text{Se}_{70}\text{Ge}_{25}\text{Cd}_5$ and $\text{Se}_{70}\text{Ge}_{25}\text{Pb}_5$ films for three different thicknesses as representative examples. It is clear from these figures that $\epsilon'(\omega)$ decreases with increasing frequency at higher temperatures. This variation is small at lower temperatures. This behavior is similar to that obtained for other amorphous semiconductors [29,30,35]. The decrease of $\epsilon'(\omega)$ with frequency can be attributed to the fact that at low frequencies, the dielectric constant $\epsilon'(\omega)$ for polar materials is due to the contribution of multicomponent of polarizability, deformational polarization (electronic and ionic polarization) and relaxation polarization (orientational and interfacial polarization) [36]. As the frequency is increased, the dipoles can not be able to rotate sufficiently rapidly, so that their oscillations begin to lag behind those of the field. As the frequency is further increased the dipoles will be completely unable to follow the field and the orientation polarization stopped, so $\epsilon'(\omega)$ decreases approaching a constant value at a higher frequency due to the interfacial or space charge polarization.

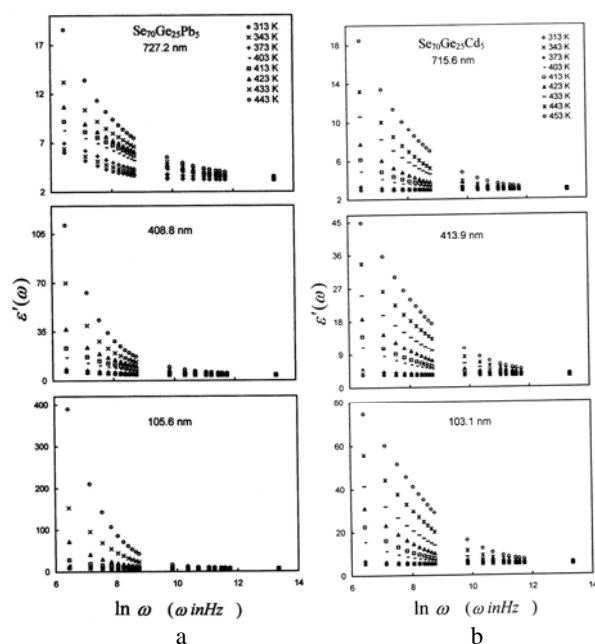


Fig. 6(a) Frequency dependence of the dielectric constant $\epsilon'(\omega)$ for $\text{Se}_{70}\text{Ge}_{25}\text{Cd}_5$ films of thicknesses 715.6, 413.9 and 103.1 nm at different temperatures. (b) Frequency dependence of the dielectric constant $\epsilon'(\omega)$ for $\text{Se}_{70}\text{Ge}_{25}\text{Pb}_5$ films of thicknesses 727.2, 408.8 and 105.6 nm at different temperatures.

3.2.2. Thickness dependence of $\epsilon'(\omega)$.

Thickness dependence of $\epsilon'(\omega)$ was studied at different frequencies at room temperature 313 K as well as the elevated temperature 393 K as representative examples for all investigated film compositions. The obtained results

are illustrated in Figs. 7(a&b). It is clear from these figures that $\epsilon'(\omega)$ decreases with increasing film thickness in the investigated ranges of temperature and frequency. From these figures, it is observed that the addition of third element Ag, Cd or Pb to $\text{Se}_{70}\text{Ge}_{30}$, increases $\epsilon'(\omega)$ in the investigated range of thickness. The observed increase with Pb is higher than that of Cd which in turn higher than that of Ag. This may be due to the formation of charged defect centers by metal additives and the decrease in bond energy between Se (the host element) and metal additives Ag, Cd or Pb as given in Table 1.

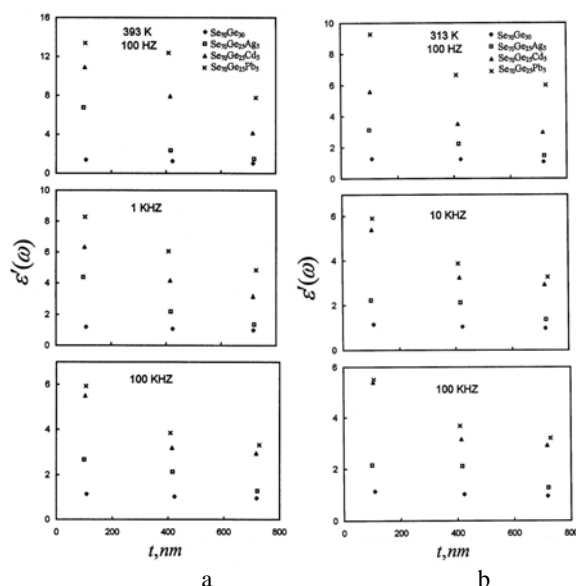


Fig. 7(a) Thickness dependence of dielectric constant $\epsilon'(\omega)$ for $\text{Se}_{70}\text{Ge}_{30}$, $\text{Se}_{70}\text{Ge}_{25}\text{Ag}_5$, $\text{Se}_{70}\text{Ge}_{25}\text{Cd}_5$ and $\text{Se}_{70}\text{Ge}_{25}\text{Pb}_5$ films at room temperature (313K) and different frequencies. (b) Thickness dependence of dielectric constant $\epsilon'(\omega)$ for $\text{Se}_{70}\text{Ge}_{30}$, $\text{Se}_{70}\text{Ge}_{25}\text{Ag}_5$, $\text{Se}_{70}\text{Ge}_{25}\text{Cd}_5$ and $\text{Se}_{70}\text{Ge}_{25}\text{Pb}_5$ films at 393 K and different frequencies.

3.2.3. Temperature and frequency dependence of $\epsilon''(\omega)$.

Dielectric loss $\epsilon''(\omega)$ for films of the investigated compositions was studied as a function of temperature and frequency. Figs.8(a&b) depict the temperature dependence of $\epsilon''(\omega)$ at different frequencies respectively for $\text{Se}_{70}\text{Ge}_{25}\text{Ag}_5$ and $\text{Se}_{70}\text{Ge}_{25}\text{Pb}_5$ films of three different thicknesses as representative examples. It is clear from these figures that $\epsilon''(\omega)$ remains almost constant with temperature till 363 K. After which, $\epsilon''(\omega)$ increases with increasing temperature. The increase is being more at low frequencies. Thus, $\epsilon''(\omega)$ exhibit strong temperature dependence at high temperatures and low frequencies.

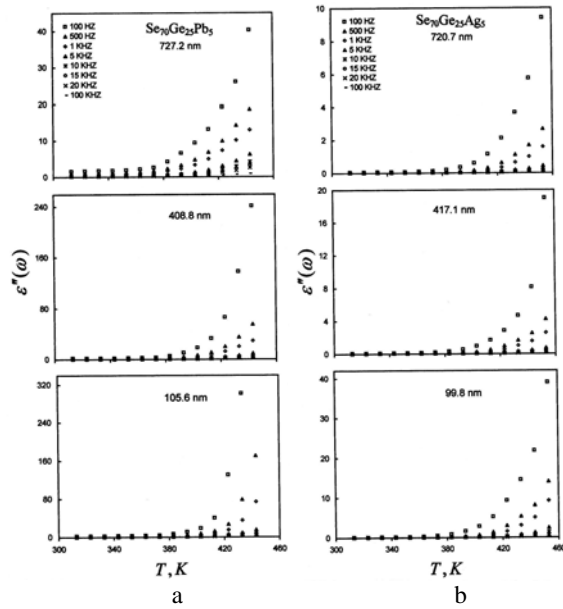


Fig. 8(a) Temperature dependence of the dielectric loss $\varepsilon''(\omega)$ for $\text{Se}_{70}\text{Ge}_{25}\text{Ag}_5$ films of thicknesses 720.7, 417.1 and 99.8 nm at different frequencies. (b) Temperature dependence of the dielectric loss $\varepsilon''(\omega)$ for $\text{Se}_{70}\text{Ge}_{25}\text{Pb}_5$ films of thicknesses 727.2, 408.8 and 105.6 nm at different frequencies.

Study of dielectric relaxation is important to understand the origin of dielectric losses in materials. Stevels [37] divided relaxation phenomenon into three parts: conduction, dipole and vibrational losses. These losses involve the migration of ions over large distances and ions jump over the highest barriers in the network. As ions move, they give some of their energy to the lattice as heat. The amount of heat loss per cycle is proportional to $\sigma_{ac}(\omega) / \omega$. It is thus obvious that values of conduction losses at low temperatures have minimum value. As the temperature increases, $\sigma_{ac}(\omega)$ increases and so the conduction losses increase.

Frequency dependence of dielectric loss $\varepsilon''(\omega)$ at different temperatures for $\text{Se}_{70}\text{Ge}_{30}$ and $\text{Se}_{70}\text{Ge}_{25}\text{Cd}_5$ films with different thicknesses as representative examples can be clarified by plotting $\varepsilon''(\omega)$ against $\ln \omega$, as given in Figs. 9(a&b). It is clear from these figures that $\varepsilon''(\omega)$ decreases with increasing frequency. The variation of $\varepsilon''(\omega)$ with frequency is small at low temperatures but it increases obviously at high temperatures. The decrease of $\varepsilon''(\omega)$ with frequency can be attributed to the fact that at low frequencies, high value of $\varepsilon''(\omega)$ is due to the migration of ions in the material. At high frequencies ion vibrations may be the only source of dielectric loss.

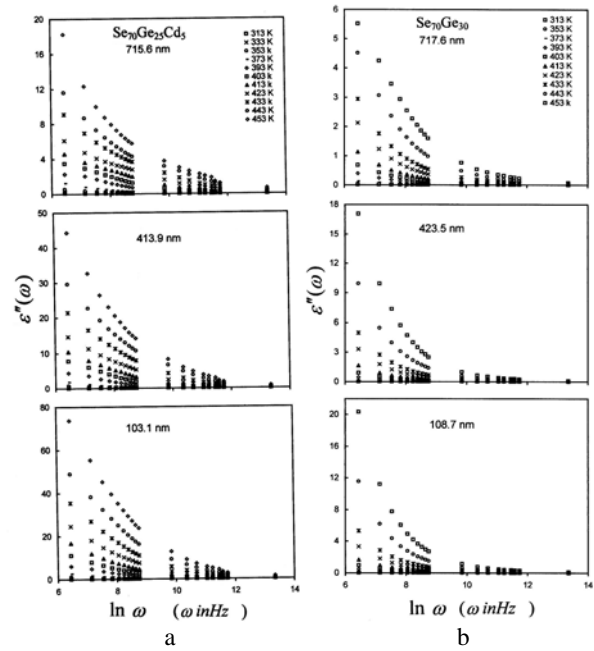


Fig. 9(a) Frequency dependence of the dielectric loss $\varepsilon''(\omega)$ for $\text{Se}_{70}\text{Ge}_{30}$ films of thicknesses 717.6, 423.5 and 108.7 nm at different temperatures. (b) Frequency dependence of the dielectric loss $\varepsilon''(\omega)$ for $\text{Se}_{70}\text{Ge}_{25}\text{Cd}_5$ films of thicknesses 715.6, 413.9 and 103.1 nm at different temperatures.

According to Mott et al. [38,39] when a chalcogenide sample is placed in an electric field, electron hops take place between localized sites. Charge carriers, moving between these sites hop from a donor to an acceptor state. In this respect each pair of sites forms a dipole. So, it can be shown that dielectric properties of chalcogenide glasses can be interpreted, by considering a set of dipoles, as long as the temperature is high enough [34], which is experimentally confirmed; below a certain temperature the dielectric permittivity (loss) does not depend on temperature. It is supposed that each dipole has a relaxation time depending on its activation energy [40], which can be essentially attributed to the existence of a potential barrier W_M , over which carriers must hop [41]. This potential barrier, as proposed by Elliott [1,24], is due to coulombic interaction between neighbouring sites forming a dipole.

The frequency dependence of $\varepsilon''(\omega)$ is represented as $\ln \varepsilon''(\omega)$ versus $\ln \omega$ as given in Fig. (10), for $\text{Se}_{70}\text{Ge}_{30}$ and $\text{Se}_{70}\text{Ge}_{25}\text{Cd}_5$ compositions of nearly the same thicknesses as representative examples, since the observed behaviour is the same for the studied compositions. It is clear from these figures that the obtained curves are straight lines at various temperatures.

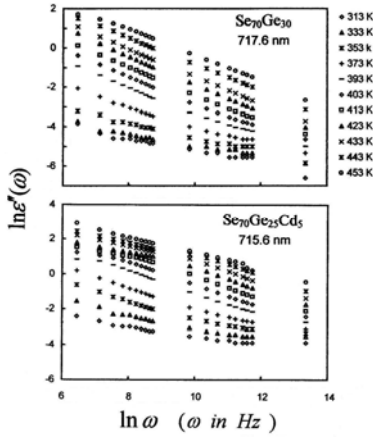


Fig. 10. Plot of $\ln \varepsilon''(\omega)$ versus $\ln \omega$ for $\text{Se}_{70}\text{Ge}_{30}$ and $\text{Se}_{70}\text{Ge}_{25}\text{Cd}_5$ films of nearly the same thickness at different temperatures.

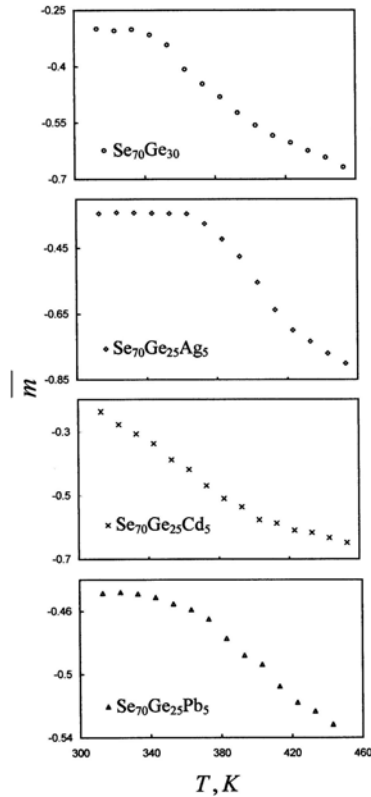


Fig. 11 Temperature dependence of the experimental mean values of \bar{m} for $\text{Se}_{70}\text{Ge}_{30}$, $\text{Se}_{70}\text{Ge}_{25}\text{Ag}_5$, $\text{Se}_{70}\text{Ge}_{25}\text{Cd}_5$ and $\text{Se}_{70}\text{Ge}_{25}\text{Pb}_5$ films.

According to Guintini et al. [42], $\varepsilon''(\omega)$ at a particular frequency in the temperature range where dielectric dispersion occurs, is given by :

$$\varepsilon''(\omega) = (\varepsilon_0 - \varepsilon_\infty) 2\pi^2 N (ne / \varepsilon_0)^2 k T \tau_0^m W_M^{-4} \omega^m \quad (5)$$

which can be written as

$$\varepsilon''(\omega) = B \omega^m \quad (6)$$

Here, n is the number of electrons that hop, N is the concentration of localized sites, ε_0 and ε_∞ are the static and optical dielectric constants respectively and W_M is the maximum barrier height (the energy required to move the electron from a site to the infinity). The power m of Eq.(6) was calculated from the negative slopes of the obtained straight lines of Fig. (10) at different temperatures. The obtained values of m are displayed as a function of temperature in Fig.(11) for all investigated compositions. It is clear from these figures that m decreases with increasing temperature in the investigated range. The obtained results satisfies the empirical law [43]: $\sigma_{ac}(\omega) = \omega \varepsilon''(\omega) = \omega B \omega^m = A \omega^S$. Since S is temperature dependent [1] {Fig.3(a)}, m should, consequently, depend on T .

3.2.4. Thickness dependence of the dielectric loss $\varepsilon''(\omega)$.

The thickness dependence of $\varepsilon''(\omega)$ was studied for the investigated film samples in the studied ranges of frequency and temperature. The thickness dependence of the investigated samples at 100 KHz is displayed in Fig.(12) as a representative example at room temperature (313 K) as well as two other elevated temperatures 353 & 393 K.

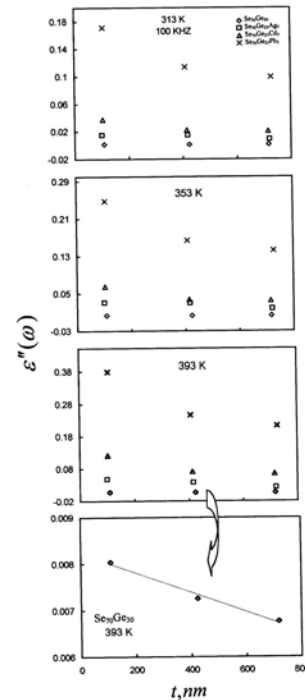


Fig. 12 Thickness dependence of dielectric loss $\varepsilon''(\omega)$ for $\text{Se}_{70}\text{Ge}_{30}$, $\text{Se}_{70}\text{Ge}_{25}\text{Ag}_5$, $\text{Se}_{70}\text{Ge}_{25}\text{Cd}_5$ and $\text{Se}_{70}\text{Ge}_{25}\text{Pb}_5$ films at 100 kHz and different temperatures.

It is clear that $\varepsilon''(\omega)$ decreases with increasing thickness for all investigated compositions. The obtained relation for $\text{Se}_{70}\text{Ge}_{30}$ composition is magnified for clarification.

Moreover, Fig.(12) shows that metal additives Ag, Cd or Pb increase $\varepsilon''(\omega)$ in the investigated range of thickness. The observed increase with Pb is higher than that of Cd which in turn higher than that of Ag. The effect of metal additives on $\varepsilon''(\omega)$ can be confirmed using the relation $\sigma_{ac}(\omega) = \omega\varepsilon''(\omega)$ [43], which implies that $\varepsilon''(\omega)$ increases with $\sigma_{ac}(\omega)$. Since $\sigma_{ac}(\omega)$ increases with metal additives, so $\varepsilon''(\omega)$ increases also with metal additives.

4. Conclusions

Amorphous $\text{Se}_{70}\text{Ge}_{30}$, $\text{Se}_{70}\text{Ge}_{25}\text{Ag}_5$, $\text{Se}_{70}\text{Ge}_{25}\text{Cd}_5$ and $\text{Se}_{70}\text{Ge}_{25}\text{Pb}_5$ thin films were prepared by thermal evaporation technique. The ac conductivity, dielectric constant and dielectric loss of the studied films are found to be temperature and frequency dependent.

The temperature dependence of $\sigma_{ac}(\omega)$ and frequency exponent S are explained on the basis of correlated barrier hopping (CBH) model.

The temperature and frequency dependence of the dielectric constant $\varepsilon'(\omega)$ can be explained in terms of the change of multicomponent of polarization: deformational and relaxation polarization with temperature and frequency.

The temperature and frequency dependence of the dielectric loss $\varepsilon''(\omega)$ can be explained in terms of the change of different losses (conduction, dipole relaxation and deformational) of the relaxation phenomenon with temperature and frequency.

References

- [1] S. R. Elliott, Phil. Mag. **36**, 1291 (1977).
- [2] A. R. Long, Adv. Phys., **31**, 553 (1982).
- [3] S. Mahadevan, A. Giridhar, J.Non-Cryst.Solids, **197**, 219 (1996).
- [4] N. Tohge, T.Minami, M. Tanaka, J.Non-Cryst.Solids **37**, 23 (1980).
- [5] M. F. A. Alias, M. N. Makadsi, Z. M.Al-Ajeli, Turk.J.phys., **27**, 133 (2003).
- [6] K. L. Bhatia, P. Singh, N.Kishore, S. K. Malik, Phil.Mag.B **72**, 417 (1995)
- [7] S. S. Fouad, A. E. Bekheet, A.M. Farid, Physica B., **322**, 163 (2002).
- [8] R. J. Dejus, S. Susman, K. J. Volin, D. G. Montague D. L. Price, J.Non-Cryst.Solids, **143**, 162 (1992).
- [9] K. Sedeeq, Indian J. of Pure & Applied Physics., **36**, 454 (1998).
- [10] T. Kawaguchi, S. Maruno, S. R. Elliot, J.Appl. Phys. **79**, 9096 (1996).
- [11] S. L. Sharma, D. R. Sharma, Phys.Stat.Sol. (a). **127**, k109 (1991).
- [12] S. L. Sharma, D. R. Sharma, Semicon. Sci. Technol. **8**, 344 (1993).
- [13] M. Pollak, T. H. Geballe, Phys. Rev. **122**, 1742 (1961).
- [14] G. E. Pike, Phys. Rev. B, **6**, 1572 (1972).
- [15] S. Tolansky, Introduction to Interferometry, Longman, London (1955).
- [16] K. Shimakawa, Phil. Mag. B, **46**, 123 (1982).
- [17] K. L. Bhatia, P. Singh, N. Kishore, S. K. Malik, Phil. Mag. B, **72**, 417 (1995).
- [18] F. Salman, Turk. J. Phys., **28**, 41 (2004).
- [19] S. R. Elliott, Adv. Phys., **36**, 135 (1987).
- [20] I. G. Austin, N.F. Mott, Adv. Phys., **18**, 41 (1969).
- [21] S. R. Elliott, Phil. Mag. B, **37**, 553 (1978).
- [22] S. R. Elliott, Phil. Mag. B, **37**, 135 (1978).
- [23] M. Kastner, H. Fritzche, Phil. Mag. B **37**, 199 (1978).
- [24] S. R. Elliott, Solid State Comm., **27**, 749 (1978).
- [25] M. A. Afifi, M. Fadel, E. G. El-Metwally, A. M. Shakra, Vacuum, **77**, 259 (2005).
- [26] J. A. Dean, "Lange's Hand Book of Chemistry" Fourteenth Edition, University of Tennessee, Knoxville (1992).
- [27] R. C.Weast "Hand Book of Chemistry and Physics" 50th Edition (1969-1970).
- [28] M. M. El-Nahass, A. F. El-Deeb, H. E. A. El-Sayed, A. M. Hassanien, Physica B **388**, 26 (2007).
- [29] Satish Kumar, M. Husain, M. Zulfeqar, Physica B **371**, 193 (2006).
- [30] N. A. Hegab, A. E. Bekheet, M. A. Afifi, L. A. Wahab, H. A. Shehata, J. Ovonic Research **3**, 71 (2007).
- [31] M. Ilyas, M. Zulfeqar, Z. H. Khan, M. Husain, Physica B **254**, 57 (1998).
- [32] R. Arora, A. Kumar, Phy. Stat. Sol. (A) **115**, 307 (1989).
- [33] R. S. Kundu, K.L. Bhatia, N. Kishore, P. Singh, Philos. Mag. B **72**, 513 (1995).
- [34] K. K. Srivastava, A. Kumar, O. S. Panwar, L. N. Lakshminarayan, J. Non-Cryst. Solids **33**, 205 (1979).
- [35] A. M. Farid, H. E. Atyia, N. A. Hegab, Vacuum **80**, 284 (2005).
- [36] B. Tareev, "Physics of dielectric Materials" Mir Publishers, Moscow, (1975).
- [37] J. M. Stevels, Handbuch der Physik, Ed. Flugge, Springer, Berlin, 350 (1957).
- [38] N. F. Mott, E. A. Davis, R. A. Street, Phil. Mag. **32**, 961 (1975).
- [39] R. A. Street, N. F. Mott, Phys. Rev. Lett. **35**, 1293 (1975).
- [40] A. E. Streen, H. Eyring, J. Chem. Phys., **5**, 113 (1937).
- [41] M. Pollak, G. E. Pike, Phys. Rev. Lett., **28**, 1494 (1972).
- [42] J. C. Giuntini, J. V. Zanchetta, D. Jullien, R. Eholie P. Houenou, J. Non-Cryst. Solids, **45**, 57 (1981).
- [43] K. L. Ngai, A. K. Jonscher, C. T. White, Nature **277**, 185 (1979).

*Corresponding author: eg_elmetwally@yahoo.com