Semiconductor Physics, Quantum Electronics & Optoelectronics, 2007. V. 10, N 2. P. 58-61.

PACS 71.20.Nr; 71.23.Cq; 71.55.Jv; 72.20.Ee; 72.30.+q; 73.20.At; 73.20.Hb

Frequency-dependent dielectric coefficients of $TlInS_2$ amorphous films

S.N. Mustafaeva[1], M.M. Asadov[2], K.Sh. Qahramanov[3]
[1]Institute of Physics, Azerbaijan National Academy of Sciences
[2]Institute of Chemical Problems, Azerbaijan National Academy of Sciences
[3]"Selen" Production Association, Azerbaijan National Academy of Sciences
AZ 1143 Baku, G. Javid Ave., 33
E-mail: solmust@gmail.com

Abstract. The frequency dispersion of the loss tangent (tgδ) and the ac conductivity (σ_{ac}) of amorphous films prepared by evaporation of $TlInS_2$ has been investigated at frequencies $f = 5\cdot10^4\ldots3.5\cdot10^7$ Hz. It is shown that, at $f > 10^6$ Hz, relaxation losses take place. It is established that the hopping conduction near the Fermi level occurs in $TlInS_2$ amorphous films at frequencies up to $3\cdot10^6$ Hz. The density of localized states at the Fermi level, the mean time for phonon-assisted tunneling, and the hopping distance have been evaluated for polymorphic $TlInS_2$ films. For frequencies above 10^7 Hz, $\sigma_{ac}(f) \sim f^2$. Such a behavior is caused by optical transitions in $TlInS_2$ amorphous films.

Keywords: amorphous film, vapour deposition, Fermi level, dielectric properties, electrical conductivity.

Manuscript received 11.04.07; accepted for publication 24.04.07;

1. Introduction

$TlInS_2$ single crystals are typical representatives of layered wide-band semiconductors [1, 2] which are characterized by a low mobility of current carriers. Such materials are very perspective for the fabrication of solid-state electron devices on their base. Layered crystals usually contain structural defects, such as dislocations and vacancies. The presence of these defects results in a high density of localized states near the Fermi level. In [3, 4], it is established by experiments that, in $TlInS_2$ single crystals along the *C*-axis in constant (dc) and alternative (ac) electric fields at $T \le 200$ K and $f = 10^5\ldots10^6$ Hz, the hopping conductivity in localized states near the Fermi level takes place.

Of some interest is the study of the dielectric properties of thin evaporated $TlInS_2$ films in alternate electric fields. The investigation of the electric properties of semiconductor materials in ac-electric fields gives information about the nature of charge transport and localized states in the forbidden gap. Such measurements allow one to determine the permittivity (ε), dissipation factor (tgδ), and optical absorption coefficient. In order to establish the mechanism of charge transport, it is necessary to know the frequency dependence of these parameters. The aim of the given paper is the investigation of the frequency-dependent dielectric parameters of $TlInS_2$ amorphous films and the clarification of the mechanism of charge transport.

2. Experimental techniques

Conditions for $TlInS_2$ thin films to be formed have been studied by the method of electron diffractometry. It has been established that amorphous films of $TlInS_2$ are polymorphous, *i.e.* there appear three different amorphous films with various $S = 4\pi\sin\theta / \lambda$ that are crystallized in tetragonal, monocline, and rhombic syngonies on the condensation surface [5]. Amorphous films $TlInS_2$ – I with $S = 20.32$; 26.06; 38.43 nm^{-1} are crystallized in monocline syngony [6]. Amorphous films $TlInS_2$ – II with $S = 23.61$; 39.25; 62.74 nm^{-1} are crystallized in tetragonal syngony [7]. And amorphous films $TlInS_2$ – III with $S = 15.02$; 24.73; 38.86 nm^{-1} are crystallized in rhombic [7] syngony. $TlInS_2$ – I, $TlInS_2$ – II, $TlInS_2$ – III films were prepared by the vacuum evaporation. Glass plates with conducting SnO_2 layer were used as substrates. The method of "three-temperatures" [9] or the evaporation from different sources was used for preparation of $TlInS_2$ films. At condensation of $TlInS_2$ films, the temperature of the glass substrate was equal to 300 K. This method of evaporation produces films of the stoichiometric composition; which was verified by X-ray spectroscopic analysis. An electron microscopic study demonstrated the amorphous structure of $TlInS_2$ – (I, II, III) films obtained under these conditions. The thickness of the $TlInS_2$ films was of the order of 1 μm. Thin film samples for dielectric measurements were prepared in a sandwich structure (Fig. 1). The contact materials used were silver and SnO_2.

Measurements of the dielectric coefficients of $TlInS_2$ – (I, II, III) films were performed at fixed frequencies in the range $5\cdot10^4…3.5\cdot10^7$ Hz by the resonant method using a TESLA BM 560 Qhmmeter. For electrical measurements, the samples were placed in a specially constructed screened cell. All measurements were performed at T = 298 K. The accuracy in determining the resonance capacitance and the quality factor $Q = 1 / \mathrm{tg}\delta$ of the measuring circuit was limited by errors related to the resolution of the device readings. The accuracy of the capacitor graduation was ±0.1 pF. The reproducibility of the resonance position was ±0.2 pF in capacitance and ±(1.0 – 1.5) scale divisions in quality factor.

3. Experimental results and discussion

Figure 2 shows the experimental frequency dependences of the dissipation factor tgδ for $TlInS_2$ – I (curve 1); $TlInS_2$ – II (curve 2), and $TlInS_2$ – III (curve 3) amorphous films.

As seen from Fig. 2, the tgδ(f) curves have two branches: a monotonically descending one (at $f < f_0$) and a rising one (at $f > f_0$). The hyperbolic decrease of tan δ with increase in the frequency is the evidence of the fact that conductivity loss becomes the main dielectric loss mechanism at $f < f_0$. A significant dispersion in tgδ at $f > f_0$ is observed for $TlInS_2$ – I film (curve 1). The increasing branches of tgδ(f) curves in $TlInS_2$ films allow us to confirm that relaxation losses take place at $f > 10^6$ Hz.

Figure 3 shows the experimentally measured frequency dependence of the ac-conductivity of $TlInS_2$ – (I, II, III) amorphous films at 298 K (curves 1-3). Curve 4 is the frequency-dependent ac conductivity of a $TlInS_2$ single crystal with tetragonal structure (a = 0.80; c = 0.67 nm).

The values of dark resistivity (ρ) of the studied materials at 298 K and the dark dc- and ac-conductivities at $f = 2\cdot10^5$ Hz are listed in Table 1. It is seen from Table 1 that the dark resistivities of $TlInS_2$ – (I, II, III) evaporated amorphous films are much greater than those of a $TlInS_2$ single crystal (by 30…800 times). For all investigated samples, the magnitude of ac-conductivity at $f = 2\cdot10^5$ Hz is much greater than that of the dc hopping conductivity: $\sigma_{ac}/\sigma_{dc} = (3.0…5.4)\ 10^2$.

The ac-conductivity of $TlInS_2$ amorphous films can be expressed by the following equation

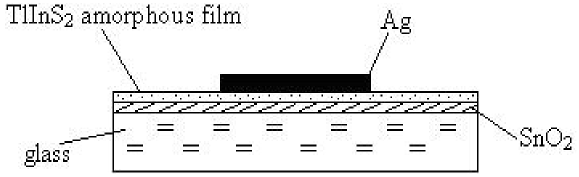

Fig. 1. Configuration of the sample on the base of a $TlInS_2$ amorphous film.

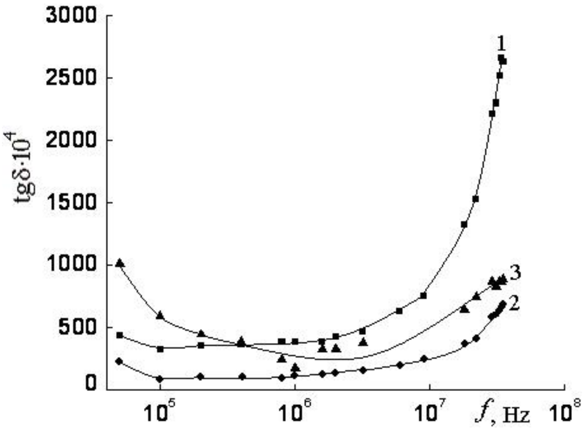

Fig. 2. Dispersion curves of tgδ in amorphous films $TlInS_2$ (I); $TlInS_2$ (II) and $TlInS_2$ (III) at T = 298 K.

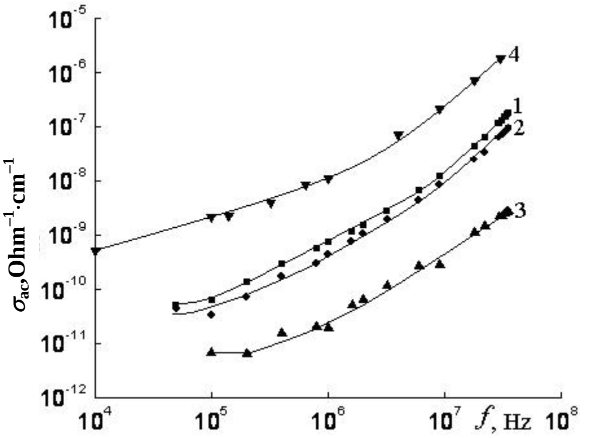

Fig. 3. Frequency-dependent ac-conductivities of $TlInS_2$ – (I, II, III) amorphous films (curves 1-3) and a single crystal (curve 4) at room temperature.

$$\sigma_{ac}(f) = \sigma_0 + \sigma_f, \tag{1}$$

where σ_0 is dc-conductivity, and

$$\sigma_f = \sigma_1 + \sigma_2 + \sigma_3. \tag{2}$$

Table 1. The dc- and ac-conductivities of $TlInS_2$ amorphous films.

Material	ρ_{dc} (Ohm·cm)	σ_{dc} (Ohm^{-1} × × cm^{-1})	σ_{ac} ($Ohm^{-1}cm^{-1}$) at $f = 2\cdot10^5$ Hz	σ_{ac}/σ_{dc}
$TlInS_2$ single crystal	10^{11}	10^{-11}	$3\cdot10^{-9}$	$3\cdot10^2$
$TlInS_2$ – I amorphous film	$3\cdot10^{12}$	$3.3\cdot10^{-13}$	$1.4\cdot10^{-10}$	$4.2\cdot10^2$
$TlInS_2$ – II amorphous film	$7.5\cdot10^{12}$	$1.3\cdot10^{-13}$	$7\cdot10^{-11}$	$5.4\cdot10^2$
$TlInS_2$ – III amorphous film	$8\cdot10^{13}$	$1.4\cdot10^{-14}$	$7\cdot10^{-12}$	$5\cdot10^2$

In (2), $\sigma_1 \sim f^n$ ($n \le 0.5$), $\sigma_2 \sim f$, and $\sigma_3 \sim f^2$. The $\sigma_{ac} \sim f$ dependence indicates that the mechanism of charge transport is the hopping over localized states near the Fermi level [10]. This charge transport mechanism is characterized by the following expression obtained in [11]:

$$\sigma_{ac}(f)=\frac{\pi^3}{96}\,e^2\,kT\,N_F^2\,a^5\,f\left[\ln\left(\frac{\nu_{ph}}{f}\right)\right]^4, \qquad (3)$$

where e is the elementary charge, k is the Boltzmann constant, N_F is the density of localized states near the Fermi level, $a = 1/\alpha$ is the localization length, α is the decay parameter of the wave function of a localized charge carrier, $\Psi \sim e^{-\alpha r}$, and ν_{ph} is the phonon frequency. Using expression (3), we can calculate the density of states at the Fermi level from the measured values of the conductivity $\sigma_{ac}(f)$. Calculated values of N_F for investigated $TlInS_2$ – (I, II, III) amorphous films are given in Table 2. The localization radius is chosen as 0.8 nm for $TlInS_2$ amorphous films (usually, a = 0.8 nm [10] in amorphous materials).

The theory of ac hopping conductivity provides an opportunity to determine the average time τ of charge carrier hopping from one localized state to another using the formula [10]

$$\tau^{-1} = \nu_{ph}\exp(-2R\alpha), \qquad (4)$$

where R is the average hopping distance:

$$R=\frac{1}{2\alpha}\ln\left(\frac{\nu_{ph}}{f}\right). \qquad (5)$$

The calculated values of τ and R for $TlInS_2$ amorphous films are given in Table 2.

Table 2. Parameters of $TlInS_2$ amorphous films obtained from high-frequency dielectric measurements.

Material	N_F, $eV^{-1}\cdot cm^{-3}$	τ, μs	R, nm
$TlInS_2$ – I amorphous film	$2.2\cdot10^{18}$	0.65	5.4
$TlInS_2$ – II amorphous film	$1.7\cdot10^{18}$	0.65	5.4
$TlInS_2$ – III amorphous film	$3.6\cdot10^{17}$	0.63	5.3

As seen from Fig. 3, at $f > 10^7$ Hz, $\sigma_{ac} \sim f^2$ in $TlInS_2$ amorphous films. The conductivity proportional to f^2 is related to optical transitions in semiconductors and is dominant at high frequencies. Such a conduction is characterized by the expression [10]

$$\sigma(f)=\left(\frac{\pi e^2}{\hbar}\right)N_F^2a^5(hf)^2\left[\ln\left(\frac{I_0}{hf}\right)\right]^4, \qquad (6)$$

where I_0 is determined from the equation

$$I = I_0\exp(-R\alpha), \qquad (7)$$

where I is the resonance energy of two localized centers, and distance between these centers is

$$R_f=\left(\frac{1}{\alpha}\right)\ln\left(\frac{2I_0}{hf}\right). \qquad (8)$$

6. Conclusions

Thus, the experimental results of high frequency dielectric measurements on $TlInS_2$ amorphous films allow us to establish the nature of dielectric losses and the mechanisms of charge transport at various frequencies and to evaluate the density of localized states near the Fermi level, average hopping time, and distance.

References

1. T.J. Isaaks, J.D. Feichther, Growth and optical properties of $TlGaSe_2$ and β-$TlInS_2$ // *J. Solid State Chem.* **14** (3), p. 260-263 (1975).
2. S.N. Mustafaeva, M.M. Asadov, V.A. Ramazanzade, Modification of photocurrent spectra of $TlInS_2$ single crystals at intercalation // *Neorgan. Mater.* **31** (3), p. 318-320 (1995) (in Russian).
3. S.N. Mustafaeva, M.M. Asadov, V.A. Ramazanzade, Dielectric properties and ac-conductivity of $TlInS_2$ single crystals // *Fiz. Tverd. Tela* **38** (1), p. 14-18 (1996) (in Russian).
4. S.N. Mustafaeva, V.A. Aliev, M.M. Asadov, DC hopping conduction in $TlGaS_2$ and $TlInS_2$ single crystals // *Fiz. Tverd. Tela* **40** (4), p. 612-615 (1998) (in Russian).
5. D.I. Ismailov, M.F. Alieva, E.Sh. Alekperov, F.I. Aliev, Electron diffractometry research of multistructural amorphous films of polymorphous $TlInS_2$

// *Fizika i Technika Poluprovodnikov* **37** (7), p. 772-775 (2003) (in Russian).

6. T.J. Isaaks, Determination of crystal symmetry of the polymorphs of thallium indium disulfide $TlInS_2$ // *Zeitsch. fur Crystallogr. B* **141** (1), p.104-108 (1971).
7. H. Hahn, B. Weltman, Uber – ternare chalkogenide des thalliums mit gallium und indium // *Naturwissensch. B* **2** (1), p. 42-45 (1967).
8. D. Muller, F. Poltman, H. Hahn, Zur-struktur ternare chalkogenide des thalliums mit aluminium, gallium und indium // *Zeitsch. Fur. Naturforsch. B* **29** (1, 2), p. 117-118 (1974).
9. *Handbook of Thin Film Technology*, Edited by Leon I. Maissel and Reinhard Glang. McGraw-Hill, New York, 1970.
10. N.F. Mott, E.A. Davis, *Electronic Processes in Non-Crystalline Materials*. Clarendon Press, Oxford, 1971.
11. M. Pollak, Frequency dependence of conductivity in amorphous solids // *Phil. Mag.* **23**(3), p. 519-542 (1971).