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Electroabsorption in chalcogenide glasses and its explanation based on barrier-cluster model

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The article deals with an explanation of electroabsorbtion in chalcogenide glasses based on a barrier-cluster model of glass. The model assumes that the structure of chalcogenide glass consists of closed clusters with a potential barrier existing in between. The barrier essentially influences not only electrical transport in glass, but also optical characteristics of the material. The absorption of light in the region of the optical absorption edge is usually connected with tunneling of carriers through the potential barrier. Due to this, the absorption process is influenced by barriers. According to the barrier-cluster model a strong electric field increases the probability of tunneling process and, thus, the value of the absorption coefficient as well. This is the essence of the explanation of electroabsorption on the base of a barrier-cluster model.

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1. Introduction

Optical phenomena in non-crystalline semiconductors represend a number of complex phenomena [1-9]. Under electroabsorption, influence of absorption of a non-crystalline solid by external electric field is understood [1-4]. As experiments show, the influence of the field is relatively small and is proportional to the square of intensity of the electric field. The change of absorption coefficient $\Delta\alpha$, influenced by the field depends also on energy of absorbed photon.

Kolomiec with co-workers [10] observed electroabsorption in amorphous semiconductor As_2S_3. In their study [11], results of measurements of absorption coefficient as well as changes of the coefficient caused by electric field are presented for semiconductors As_2Se_3, $(As_2Se_3)_{0.95}(As_2Te_3)_{0.05}$, $(As_2Se_3)_{0.91}(As_2Te_3)_{0.09}$. The measurements were carried out in the field of 10^7 Vm^{-1}. Similar results for material $(As_2Se_3)_{0.95}(As_2Te_3)_{0.05}$ are presented in [12]. The work [13] of Robertsa et al studied electroabsorption in selen. The experiments show [14,15], that a relative change of optical absorption by electroabsorption is directly proportional to the quadrat of intensity E of electric field, so that $\Delta\alpha/\alpha \sim E^2$. The work describes the measurement methods of electroabsorption [16,17].

Electroabsorption in chalcogenide glasses at the edge of optical absorption tends to be interpreted as Franz-Keldysh effect [18,19] in conditions of fluctuating electric potential [20-22]. In other cases the electroabsorption is considered to be consequence of degeneration of an electron energy level – for instance of excitons level - in a fluctuating inert electric field [23-25]. According to [26] provided considerable deficit of photon energy, the electroabsorption could be connected with electron transition between localized states. The work explains influence of the electric field on optical absorption [27] by a density change of localized states in gap of a non-crystalline semiconductor under influence of the electric field.

In the work, [28] the optical phenomena in non-crystalline semiconductors was explained based on the barrier model. The barrier model assumes that an amorphous solid consists of microscopic regions separated from each other by potential barriers. In this article the phenomena of the electroabsorption in chalcogenide glasses is explained based on the barrier-cluster model.

2. Barrier-cluster model

The barrier-cluster model assumes that an amorphous solid consists of microscopic regions-clusters, separated from each other by potential barriers. The barrier-cluster model [28,29] is an improved barrier model. The ideas of barrier model were published in author's previous works [28, 30-45]. The essential improvement consists in assigning particular physical content to microregions in glasses. The barrier-cluster model identifies the microregions with closed clusters [46-50]. The barrier–cluster model offers a new aspect of density of the states in the gap of the chalcogenide glasses together with a new aspect of electrical and optical phenomena including the electroabsorption.

The barrier–cluster model allows explanation not only of a number of important optical and electrical features of chalcogenide glasses, but also the results of X-ray structure measurements and ESR experiments [46-54]. This concept gives a new look at the density of states within

the forbidden band and at the exponential tails of the optical absorption.

In Fig. 1, the electron spectrum of an amorphous semiconductor, based on the barrier-cluster model, is shown. The potential barriers are depicted inside the conduction and valence bands, separating individual localized energy states at the edge of the band. The electron levels between barriers - due to small dimensions of the microscopic regions (clusters) - exhibit a distinct discrete character. At the lower margin of the conduction band a sub-band with low average mobility carriers (μ_1) is created. The transport in the region of energy levels, lower than the tops of barriers, is connected with a tunneling mechanism. In the papers [28, 30-45] the various electrical and optical properties of the amorphous semiconductors on the base of the barrier model are explained.

3. Absorption of light

The potential barriers are important not only from the point of view of electrical transporting properties of a semiconductor, but from the point of view of optical phenomena as well. The barriers create conditions for a strong electron-phonon interaction. We assume that an electron in an optical transition accepts not only the energy of a photon but also the phonon energy [28, 29]. Thus, the whole energy accepted is $W_e = hf + W_{phon}$ where W_{phon} is the energy acquired from a phonon "field". The quantity hf is positively determined by the wavelength of absorbed monochromatic radiation, while W_{phon} has a statistical character. On the base of this idea the origin of the exponential tails of the optical absorption is in [28,29] explained.

3.1 Optical absorption - low temperature range

At low temperatures (below T_o), absorption of light in a non-crystalline semiconductor can essentially take place only in such a way as it is depicted in a simplified form in Fig. 1. It is a low-temperature mechanism of absorption. Only photons with sufficient energy, exceeding $2W$, can be absorbed by material. The "skewed" optical transition of electron (as depicted in Fig. 1) can be virtually divided into two parts [28,29]: The first part is a vertical transition onto an energy level inside its own localized region (without tunneling); the following second part represents a horizontal tunneling transition onto a real level in an adjacent localized region. Thus, absorption of photon in a low-temperature mechanism is connected with tunneling of electron through a potential barrier. Let us remark that at lower temperatures (below T_o), absorption of light in the vicinity of optical absorption edge could principally run without tunneling process, i. e. within a single localized region. However, probability of such transitions is small due to a distinctly discrete character of the lowest levels as well as due to a small number of such levels in a single microregion. Therefore, absorption connected with tunneling to adjacent regions is more probable. In the case of parabolic potential barrier (Fig. 2), the dependence of potential energy $W(x)$ of electron on its position can be denoted as $W(x) = -ax^2 + W_o$, where the constant W_o means the height of the barrier from the bottom of the conduction band and the quantity a determines "narrowness" of the barrier. For the probability of the tunneling it can be written [28]

$$p(\varepsilon) \approx \exp\{-A\,\Delta W\} \tag{1}$$

$$\text{where} \quad A = \left\{-\frac{\pi}{\hbar}\sqrt{\frac{2m}{a}}\right\}, \quad \Delta W = W_0 - \varepsilon \tag{2}$$

It may seem at the first sight that phonons play no role in the low-temperature absorption.

However, it is not the case. The problem is connected with a clearly discrete character of energy levels in the region between barriers (below barriers peaks). A jump of an electron onto a real level of a neighboring localized region needs the phonons, needs usually small "tuning", i. e. a small correction of the photon energy by a certain value δW. Coefficient of optical absorption α, under the low-temperature mechanism described above, is - as can be supposed - proportional to the product of two probabilities; p_1 being the probability of tunneling through a potential barrier onto a corresponding energy level ε and

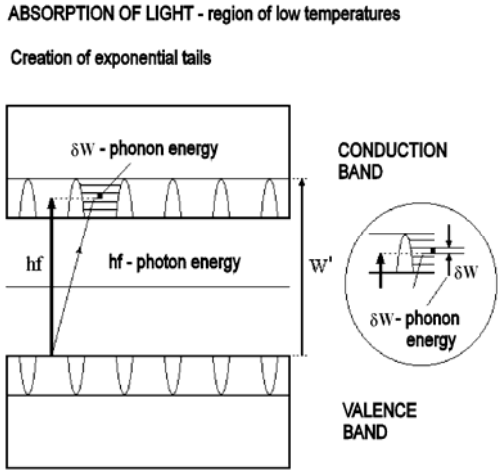

Fig. 1.

$p_2 \approx \exp\left[-\frac{\delta W}{2kT}\right]$ - probability of occurrence of a phonon with the small correction energy δW. For probability p_1, we can write according to (1) $p_1 \approx \exp[-A\Delta W]$. Thus, it can be written for the coefficient α

$$\alpha \approx p_1 p_2 \approx \exp[-A\Delta W] \exp\left[-\frac{\delta W}{2kT}\right].$$

Since for the energy difference ΔW between the level corresponding to the peak of barrier and the energy level on which the tunneling takes place, it holds $\Delta W = 2W + W_o - hf$ we arrive at the expression $\ln \alpha = Ahf - B(T) - C$ which is a mathematical expression for the exponential tail of optical absorption in a logarithmic scale. The term $B(T) = \delta W/2kT$, which depends on temperature will cause parallel shift of the straight lines towards lower absorption with decreasing temperature. Just this effect is typical for the low-temperature absorption in non-crystalline semiconductors. The change of behavior of exponential tails around T_o is caused by a change of mode (mechanism) of optical absorption with decreasing temperature. The increase of slope of tails stops when the slope reaches a certain value corresponding to the transition to a low-temperature absorption mode. Naturally, the transition is continuous.

4. Electroabsorption

4.1 Explanation of electroabsorption based on barrier-cluster model of an amorphous solid

According to what was said before, absorption of light in the region of exponential tails at temperatures below T_o is connected with tunneling of electron through a potential barrier. At that, the absorption coefficient is proportional to the probability of tunneling. As we will show, strong electric field influences the tunneling probability of an electron through a barrier and thus, the value of absorption coefficient as well. Just this is the essence of explanation of absorption on the base of a barrier model.

Tunneling through a barrier in electric field

During tunneling of an electron through a barrier without a presence of external electric field (see Fig. 2), the energy of electron does not change. In external electric field, the electron energy will continuously increase or decrease depending on whether the tunneling process runs in the direction of acting electric force or in the opposite direction. This is true at least in the one-dimensional (1D) case, which is considered now.

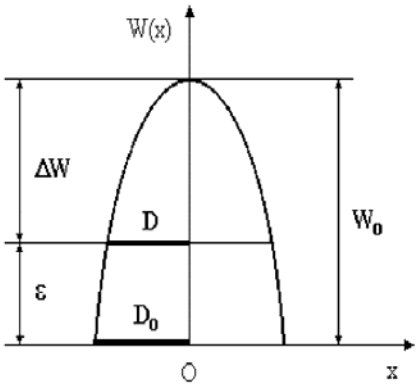

Fig. 2.

If tunneling takes place in homogenous electric field with an intensity E, the electron energy ε will be a function of position [28]. In the case depicted in Fig. 3, where the origin of the x-axis is placed in the middle of the barrier in question, it can be written

$$\varepsilon(x) = \varepsilon_o + eED \pm eEx \tag{3}$$

where $2D$ is the barrier width on the energy level ε_o and ε_o represents initial energy of the tunneling particle. The positive upper sign before the last term applies when the particle energy increases in the field; the negative sign applies on decreasing the particle energy. (See Fig. 3). For the probability of tunneling we obtain

$$p(\varepsilon) \approx \exp \left\{ -\frac{2}{h} \int_{x_1}^{x_2} \left\langle 2m\left[-ax^2 + W_0 - \varepsilon_0 - eED - eEx \right]^{1/2} dx \right\rangle \right\}$$

The lower limit of integration, x_1, is obtained from the condition $W(x_1) = \varepsilon_o$, the upper limit, x_2, from $W(x_2) = \varepsilon_o + eED + eEx_2$, where $W(x) = -ax^2 + W_o$. Thus

$$x_1 = \sqrt{\frac{W_0 - \varepsilon_0}{a}} \quad , \tag{4},$$

$$x_2 = \frac{eE \pm \sqrt{(eE)^2 + 4a(W_0 - \varepsilon_0 - eED)}}{2a} \tag{5}$$

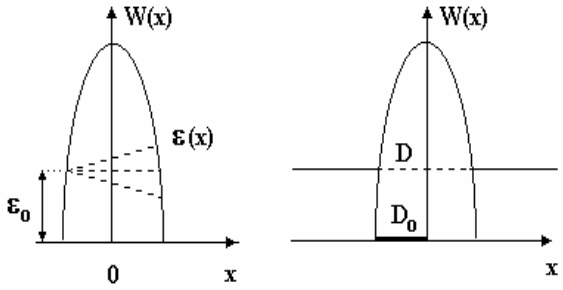

Fig. 3.

After performing integration in the respective limits, we obtain

$$p(\pm E) \approx \exp \left[-A \left(W_0 - \varepsilon_0 \mp eED + \frac{(eE)^2}{8a} \right) \right]$$

where A is given by the formula (2). Supposing that the last term appearing in parentheses in the exponent is negligible as compared to the previous one, the relation can be re-written as

$$p(\pm E) \approx \exp \left[-A(W_0 - \varepsilon_0 \mp eED) \right] \tag{6}$$

Such a transition is conditioned by relation

$$\frac{(eE)^2}{8a} << eED \tag{7}$$

from which, after expressing the parameter a we obtain $eED << 8\,(W_o - \varepsilon_o)$. Its physical sense is clear at the first sight. In the region, not too close to the top, this condition is usually fulfilled satisfactorily. If we put $E = 0$ in the relation (6), we obtain probability

$$p \approx \exp\left[-A\left(W_0 - \varepsilon_0\right)\right] \tag{8}$$

Therefore, the relation (6) can also be expressed as

$$p(\pm E) \approx p \exp\left[\mp\, A\, eED\right] \tag{9}$$

From this, it is clear that the probability of tunneling in electric field increases in one direction and decreases in another direction as compared with the original value for $E = 0$.

Derivation of relations for electroabsorption: According to the barrier model of an amorphous solid, the absorption coefficient in the temperature range below T_o is directly proportional to the probability of tunneling related to the respective energy level. However, two different probability values should be considered during absorption in electric field; the probability of tunneling p(+E) in the direction of acting electric force and the probability p(-E) in the opposite direction. The probability p(+E) is greater whereas the probability p(-E) is smaller than the probability p of tunneling without field. However, these two changes caused by field do not compensate each other. As overall probability P(E) of tunneling an electron in electric field with the intensity E, the sum p(+E) + p(-E) = p(E) will be considered for the one-dimensional (1D) case, where the probabilities p (+E) and p (-E) are given by (9). This is quite similar in absence of electric field, naturally with $E = 0$. The overall probability of tunneling in absence of field, P (0), is thus P (0) = p (0) + p (0) = 2p (0) = 2p whereby for p, the relation (8) holds true. Let us further suppose that a relative change of absorption under influence of the field is directly proportional to the relative change of the overall probability

$$\frac{\Delta\alpha}{\alpha} = \frac{\Delta P}{P} = \frac{p\left(+E\right) + p\left(-E\right) - 2p}{2p}$$

From (4) and (5), we obtain

$$\frac{\Delta\alpha}{\alpha} = \frac{1}{2}\left[\exp\left(AeED\right) + \exp\left(-AeED\right) - 2\right]$$

Supposing that $AeED << 1$ (this condition is usually fulfilled as we can demonstrate in numerical examination), the exponential functions appearing on the right-hand side of this relation can be expanded into the Taylor series with the consideration limited to the first three terms of this expansion. In this way, we obtain

$$\frac{\Delta\alpha}{\alpha} = \frac{1}{2}\left(AeED\right)^2$$

If for $D = x_1$ we substitute the relation (4), we obtain

$$\frac{\Delta\alpha}{\alpha} = \frac{1}{2}\left(AeE\right)^2 \frac{W_0 - \varepsilon_0}{a} \tag{10}$$

from which it can be seen that the relative change (increase) of the absorption coefficient is directly proportional to square of intensity of electric field. This is in agreement with the observation. The relation (10) enables us also to explain the dependence of relative absorption $\Delta\alpha/\alpha$ on the energy hf. However, although the photon energy does not appear explicitly in the indicated formula, the quantity ε_o depends on it. On optical absorption of photons with energy hf, the electrons excite onto the levels inside the conduction band, the highest of which rests in the height of ε_o above the bottom of the conduction band. At that, the relation

$$\varepsilon_o = \mathrm{h}f - 2W$$

holds true, where $2W$ means the actual width of the forbidden band (for the low-temperature region). Substituting this value into (10), we obtain

$$\frac{\Delta\alpha}{\alpha} = -\frac{\left(AeE\right)^2}{2a} hf + \frac{\left(AeE\right)^2\left(W_o + 2W\right)}{2a} \tag{11}$$

This relation is valid for the one-dimensional model of sample. The three-dimensionality of a real situation leads to the conclusion that $E^2/3$ shall be used in (11), instead of E^2. So we find

$$\frac{\Delta\alpha}{\alpha} = -\frac{\left(AeE\right)^2}{6a} hf + \frac{\left(AeE\right)^2\left(W_o + 2W\right)}{6a} \tag{12}$$

and thus, a dependence of the type

$$\frac{\Delta\alpha}{\alpha} = -C_1 hf + C_2 \tag{13}$$

At that, the positive parameters C_1 and C_2 do not depend on energy of the absorbed photon. Thus, the relative change of absorption decreases in a linear manner with the photon energy. It is a consequence of the negative sign before the positive value of C_1. This conclusion is in agreement with the experimental data [15-18].

Confrontation with the experiment: Let us first determine theoretical values of the constants C_1 and C_2, which appear in (13) for the amorphous material As_2Se_3 [1]. For calculation, we use data, which come out from the absorption measurements. The results obtained in this way will then be compared with the values derived from the electroabsorption measurements. From the absorption measurements [28], the value A = 20 eV^{-1} and a = 0,106 Jm^{-2} results for the given material. Based on theoretical relations

$$C_1 = \frac{\left(AeE\right)^2}{6a}, C_2 = \frac{\left(AeE\right)^2\left(W_o + 2W\right)}{6a}$$

which follows from (12,13), for the field intensity $E = 10^7$ Vm^{-1} we obtain $C_1 = 10.06.10^{-3}$ eV^{-1} and $C_2 = 18.1\times10^{-3}$. In theoretical calculation of C_2 we have put $2W + W_o = 1.8$ eV, which is the width of forbidden band of the semiconductor considered.

By numerical analysis of the results of electroabsorption on the semiconductor in question, the values $C_1 = 7.65\times10^{-3}$ eV^{-1}, $C_2 = 14.7\times10^{-3}$ are obtained [1]. As can be seen, the results obtained by both methods are comparable relatively well. At that - and it should be stressed - two different parameters were determined at the same time. It is also worth mentioning that for calculation of C_1 and C_2 in first case two parameters only, *a* and *A*, were used from the absorption measurement. And moreover, there was neither fitting nor any selection of some auxiliary parameters. The agreement of theory with experiment, as far as the dependence of relative absorption on field intensity, *E* and on photon energy is concerned, was already mentioned previously.

5. Conclusion

The article analyzed and explained the electroabsorption in chalcogenide glasses based on the barrier-cluster model. The model assumes that the glass consists of closed clusters with a potential barrier existing in between. The barrier-cluster model enables explanation of the results of the röntgen structural measurements and at the same time the results of the ESR experiments on the chalcogenide glasses. The barrier-cluster model is in a fact an improved barrier model published in author's previous works. The improvement consists in assigning particular physical content to microregions in glasses. The barrier-cluster model identifies the microregions with closed clusters. The barrier–cluster model offers a new aspect of density of the states in the gap of the chalcogenide glasses together with a new aspect of electrical and optical phenomena including the electroabsorption. According to the barrier-cluster model strong electric field increases the probability of tunneling process and, thus, the value of the absorption coefficient as well. The barrier-cluster model might represent a suitable base to explain some further phenomena in non-crystalline semiconductors.

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