OPTICAL BIAS EFFECT ON TRANSIENT PHOTOCURRENT IN AMORPHOUS As_2Se_3: Sn FILMS

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Abstract

Optical bias effect on transient photocurrent of As_2Se_3: Sn_x (x=0÷1.0 at.%) amorphous films, excited by a monochromatic light pulse with long-wavelength of 0.63 μm was investigated. The relaxation of the photocurrent has been recorded in the wide time interval (from 0.05 up to 25 s) and was determined by capture on the deep acceptor-like traps. In a non-doped sample of α-As_2Se_3 the relaxation of the photocurrent weakly depends on a level of optical bias. For the samples with tin impurity, even the low levels of optical bias result in sharp reduction of the photocurrent. The experimental results are discussed in frame of the model of recombination controlled by capture on deep states.

1. Introduction

It is known, that in a large time-domain (from 10^{-3} up to 10 s) the photocurrent relaxation in amorphous semiconductors is essentially determined by the capture of non-equilibrium carriers on defect and/or impurity deep states, and then due to recombination [1-4]. Results of other studies, based on measurements of the photocurrent in conditions not far from the equilibrium (time-of-light measurements [5-7], modulated photocurrents [8-10]), also indicate these processes and are interpreted in frame of the multiple-trapping model. Nevertheless, these measurements do not specify the well-determined process of deep capture and thus not assume the structured form of the density-of-states distribution with distinct levels of defects [11].

In this paper the studies of transient photocurrent in As_2Se_3: Sn_x samples: pure (x=0) and with tin impurity (x~1 ат. %), and various levels of optical bias, are presented. These studies are continuation of our previous work in which the detection of persistent photoconductivity in a-As_2Se_3: Sn, samples associated with the deep states was reported, and also have the aim to clear up this phenomenon by experiments with the optical bias. It is supposed, that owing to quasi-Fermi-level movement, optical bias changes population of these centers and can result in suppression of deep capture and/or increase in the recombination rate [2,12]. Based on the photocurrent relaxation curve analysis obtained for various levels of optical bias it was shown, that the origin of the long-term of photocurrent decay in As_2Se_3: Sn samples is associated with the special nature of acceptor-like centers induced by tin. These states are characterized by a small value of hole capture cross-section and their density is sufficiently higher.

2. Samples and experimental method

The optical bias effect was measured in amorphous films a-As_2Se_3: Sn_x (x=0÷1.0 at. %), prepared by thermal flash evaporation in vacuum on the glass substrate heated up to 120 ^{0}C.

The thin film of a thickness about L~2 μm were placed between two sprayed aluminum electrodes, one of which (top) was semi-transparent (the electrode area A~0.56 sm^2).

The transient photocurrent was excited by a monochromatic light pulse from SPM-2 spectrometer (wavelength λ=0.63 μm, intensity F = $(1\div2)\bullet10^{13}$ photon/(cm^2 s), with the spectral resolution $\Delta\lambda$ < 20 nm), and was formed by the electromechanical shutter (time of operation of about 0.02 s). The wavelength of the additional illumination corresponded to the a-As_2Se_3 optical absorption edge provided homogeneous generation of photo-carriers in bulk of the sample. The light intensity was adjusted by the spectrometer slip width, and controlled by the Ge-photo diode.

The applied level of optical bias was created by an usual incandescent lamp illumination passed through the red filter. The level of optical bias was set by a current in a circuit of the lamp and determined by the value of steady-state photocurrent.

The positive polarity of constant voltage bias of about 2 V was applied to the top electrode (anode). Transient photocurrent was amplified by an electrometer and then recorded by the two-coordinate (X-Y) potentiometer; its operation was synchronized with a pulse of exciting light. The relaxation of the photocurrent was measured at room temperature in the time interval 0.05 - 25 s.

3. Experimental results

3.1 Non-doped sample

Fig. 1 illustrates results of measurement of a transient photocurrent in non-doped sample of As_2Se_3 without optical bias (curve 1) and for four consistently increased levels of optical bias (curves 2-5). The appropriate levels of optical bias (in values of steady-state photocurrent I_{OB}) are given in explanations of figure 1. Curve 1 in the figure obtained without optical bias, is similar to those obtained earlier for samples As_2Se_3 [13,14]. It is seen that after the light is switched on the photocurrent sharply increases and then at t~1 s achieves saturation. It is usually supposed that in this region of saturation the current is determined by electron-hole pair generation rate and their recombination.

The relaxation of a photocurrent at the moment when the light is switched off and without optical bias also is characterized by fast decay (curve 1 in Fig. 1). The decay time constant τ~0.05¸ 0.15 is close to the reported values obtained in multi-trapping model for a-As_2Se_3 films [15]. The following curves 2, 3, 4 and 5, situated below, were obtained at the presence of optical bias (I_{OB}~$2\cdot10^{-10}$, $4.3\cdot10^{-10}$, $8\cdot10^{-10}$ and $8\cdot10^{-9}$ A, respectively). They are similar to a curve 1 however the saturating value of photocurrent (near 1 s) is lower. This is in the consent with reduction of the decay of time constant τ at illumination and specifies correlation between the photocurrent decay and recombination [4,16,17].

Let's note related to optical bias effect the weak change of a relaxation rate and the form of decay curves on long time (t >1 s) shown in Fig. 2, when processes of carrier capture and release from deep traps are essential. Curves of photocurrent decay are normalized to it's value at the moment t=0 s, appropriate to termination of the exciting light pulse. For the lowermost curve 5 obtained at the largest intensity of illumination, the relaxation is observed in a time interval limited by noise of measuring installation.

3.2 Doped sample

Results of optical bias effect on transient photocurrent in samples doped with tin impurity (x=1.0 at. %) are presented in Fig. 3 and Fig. 4. At low intensities of additional

illumination and in darkness (curves 1, 2 and 3 in Fig. 3) the impurity influence is opposed to optical bias effect. Really, after the light is switched on the delayed character of transient photocurrent from time and absence of saturation is seen at long times (comparable with an excitation pulse duration t~5 c). Thus, the absolute value of photocurrent is higher than in a non-doped sample.

After light is switched off the decay curves weakly vary in time and, at t>0.05-0.1 s, are characterized by an extended plateau with a very small decay rate (the time constant τ is about 1.0-3.0 c). It is persistent photoconductivity effect, which was revealed and described by us in the previous works [14,18].

Photocurrent on this slow portion of a relaxation appreciably decreases at illumination and, already at a small level of optical bias (I_{OB}~4.5 x 10^{-9} A), the full suppression of effect takes place. Simultaneously to reduction of photocurrent on this portion, there is a reduction of a decay time constant. The fast portion of decay curves (t<0.5 s) as well as in a non-doped sample weakly varies at illumination.

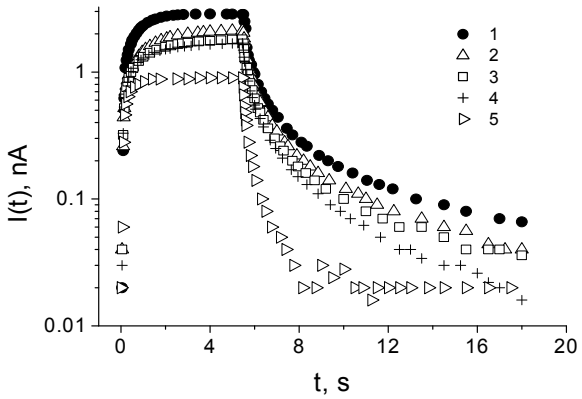

Fig. 1. Photocurrent transient recorded in darkness and for four consistently increased levels of optical bias for a non-doped sample of a-As_2Se_3. A current of constant bias at a voltage of 2 V in A: 1 - 8.0 x 10^{-11} (in darkness), 2 - 2.0 x 10^{-10}, 3 - 4.3 x 10^{-10}, 4 - 8.0 x 10^{-10}, 5 - 8.0 x 10^{-9}.

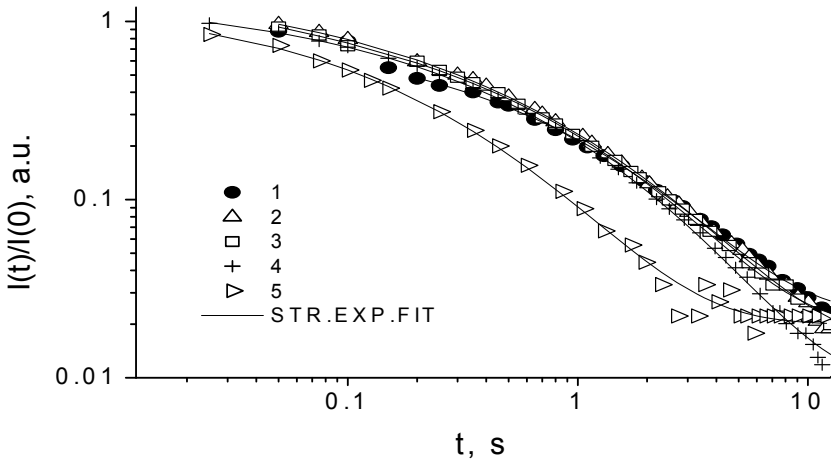

Fig. 2. Normalized photocurrent decay (after light is switched off), deduced from curves in Fig. 1 numbering by the same manner. Solid lines are result of the best adjustment with stretched exponent. Coincidence of curves 1-4 is observed in the overall time interval.

Thus, distinction between curve Fig. 2 and Fig. 4 is shown in the range of large times (0.1-10 s) and is associated to a portion of a slow relaxation in a sample of a-As_2Se_3: Sn. Optical bias dramatically influences the form and decay rate of curves, reduces photocurrent. Only at the initial stage of a relaxation and/or the high optical bias the behavior of transient photocurrent well coincides with the curves in Fig. 2 obtained for a non-doped sample. Limiting value of photocurrent and decay time constant (curve 5) becomes same, as well as in a non-doped sample.

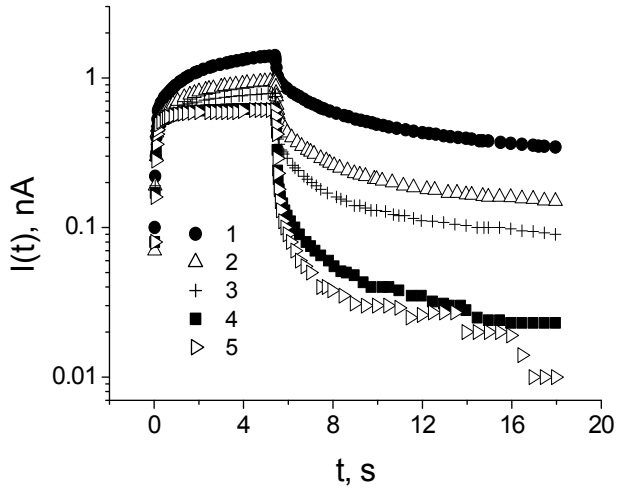

Fig.3. Photocurrent transient recorded in darkness and for four consistently increased levels of optical bias for a doped sample of a-As_2Se_3: Sn. A current of constant bias at a voltage of 2 V in A: 1 - 1.76 x 10^{-9} (in darkness), 2 - 2.5 x 10^{-9}, 3 - 3.0 x 10^{-9}, 4 - 4.0 x 10^{-9}, 5 - 4.5 x 10^{-9}.

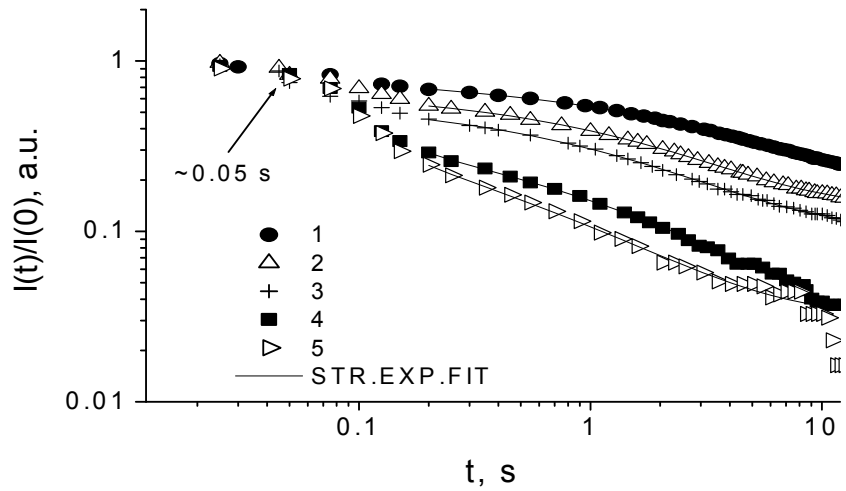

Fig. 4. Normalized photocurrent decay (after light is switched off), deduced from curves in Fig. 3 numbering by the same manner. Solid lines are result of the best adjustment with stretched exponent. Concurrence of curves is observed only at small times t < 0.05 s.

4. Relaxation of photocurrent controlled by deep capture

Systematical studies of the optical bias effect on transient photocurrent in amorphous semiconductors have begun from known paper by R. Pandya and E. A. Schiff [20]; it was shown, that for interpretation of the effect, it is necessary to take into account the final population of traps, which changes under optical bias and influences the issue of relaxation. In

papers [21,22] transient photocurrent was measured in thick (up to 10 μm) As_2Se_3 and As_2Se_3: Sn layers in the time-of-flight mode and at the higher levels of optical bias. It was shown that under these conditions the drift mobility essentially increases but, vice-versa, the recombination time decreases. These studies were limited by small times (from 10^{-5} up to 10^{-1}s) in which only the fundamental band tail has been revealed in experiment.

The results presented in our paper were obtained in the wide time interval (up to 20 s) and mainly related to processes of capture and recombination on deep states of defects. Hence, the model which would examine photocurrent relaxation on large times and which would take into account these states is necessary for the analysis of the obtained results. Recently such model was proposed by V. I. Arkhipov [23]. It was used for analysis of the slow photocurrent decay in amorphous As_2S_3 chalcogenide films doped with ions of praseodymium [23].

It was assumed that due to a high density of gap states in amorphous semiconductors the majority of non-equilibrium charge carriers are localized already at the first time of relaxation. Therefore, decay of photocurrent on large times is determined by the dynamics change of captured hole density within the quasi-continuously distributed localized states. For bimolecular mode of recombination (typical for As_2Se_3 at room temperature [7,24]) the kinetic equation takes the form

$$\frac{dp(t)}{dt} = -Rp_c(t)p(t)\,, \tag{1}$$

where p_c is the free holes concentration, R is the recombination's constant, p is the full hole concentration. Then it was assumed that in the experimental time interval t >1 s the slow relaxation of a photocurrent is completely controlled by carrier release from the localized states above the quasi-Fermi-level and described by expression [25]:

$$p(t) = \int_0^\infty dE \exp\left(-\frac{t}{\tau_e}\right) g(E) \approx \int_{kT\ln(\nu t)}^\infty dE g(E)\,. \tag{2}$$

Here E is the energy of the localized state, $\tau=\nu^{-1}\exp(E/kT)$ is the time constant of thermal release processes, g(E) is the density-of-states distribution (DOS-function), k is Boltzmann's constant, T is the temperature and ν is the typical phonon frequency.

Substitution (2) in (1) results in expression for time dependence of photocurrent decay I(t):

$$I(t) = Ae\left(\frac{\mu_c}{R}\right) F \frac{kT}{t}\left[\int_{kT\ln(\nu t)}^\infty dE g(E)\right]^{-1} g(kT\ln(\nu t))\,, \tag{3}$$

(e is the elementary charge, μ_c is the mobility of charge carriers in the band of conducting states). As well as in the model describing measurements of phase shift [8,9], it is supposed that the emission rate of captured carriers from deep traps exponentially decreases with increase of energy and restricts to rate of the relaxation process. Nevertheless it is seen that as against result, which is turning out at measurement of modulated photocurrents, this dependence is not related directly to DOS-function.

In Fig. 5 the I(t) x t vs. kTln(νt) diagrams for non-doped (a) and doped (b) samples, along with model curves predicted by equation (3) (solid lines), are presented. In calculations the ratio (μ_C / R) was used as adjusting parameter and DOS-function was chosen as quasi-continuously distributed traps of exponential tail and Gauss-form band of deep traps:

$$g(E) = \frac{N_t}{E_0}\exp\left(-\frac{E}{E_0}\right) + \frac{N_d}{\sqrt{2\pi\sigma}}\exp\left[-\frac{(E-E_d)^2}{2\sigma^2}\right]. \qquad (4)$$

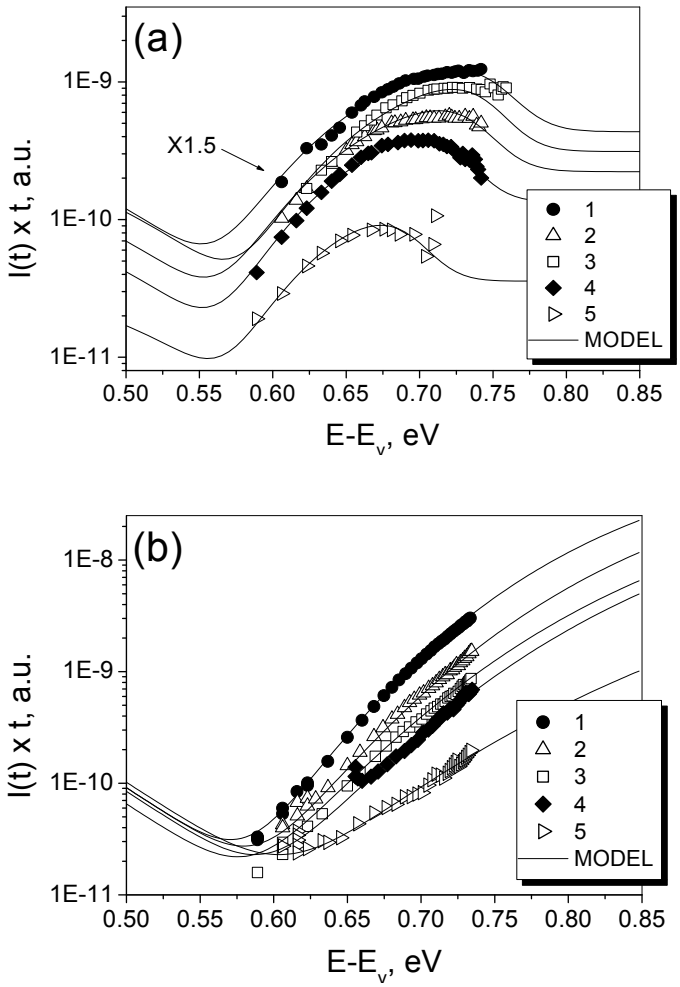

Fig. 5. I (t) x t diagrams obtained for sample As_2Se_3 (a) and As_2Se_3: Sn (b) samples from the curves presented in Fig. 2 and 4, accordingly. Numbering of curves corresponds to Fig. 1 and 3. Solid lines are calculations based on equations (3) and (4) (see text for details).

with the N_t, E_0 parameters of an exponentially band tail and N_d, E_d, σ, are parameters of a band of deep trap distributions listed in Table 1.

Without optical bias and for a non-doped sample product I (t) x t characterizes the effective density-of-states (curve 1) and has a bump features with a maximum at E_d~0.65 eV, which corresponds to characteristic time constant τ~0.1 s obtained for this sample.

Measurements of the modulated currents [26] and transient photoconductivity [3] also result in similar peak near 0.7 eV. It seems obvious that under optical bias the effective concentration of traps is suppressed. It is in accordance with the conclusion based on analysis of a transient photocurrent. Curves for higher levels of optical bias show similar dependence.

Table 1. The parameters of the deep centers obtained by modeling of experimental data I (t) x t in Fig. 5 with the DOS-function described by formula (4) T=289 K, F=10^4 V/cm. The steady-state photocurrent for curves 2-5 characterizes a level of optical bias.

[Sn], at. %	crv. number	I_{dark}, A	I_{OB}, A	$\mu_C / R \bullet 10^8$, $cm^{-1}V^{-1}$	N_t, cm^{-3}	N_d, cm^{-3}	E_d, eV	σ, eV
0.0	1	8e-11		0.0065	5e19	6e15	0.639	0.038
	2		2e-10	0.005	5e19	5e15	0.636	0.036
	3		4.3e-10	0.0068	10e20	1e16	0.648	0.035
	4		8e-10	0.003	1.5e19	1.5e15	0.632	0.034
	5		8e-9	0.0008	2e20	1e16	0.623	0.03
1.0	1	1.76e-9		1.35	4e19	1.2e18	0.875	0.079
	2		2.5e-9	1.35	4e19	1.2e18	0.924	0.09
	3		3e-9	1.1	3.5e19	1.2e18	0.954	0.1
	4		4e-9	1.0	5e19	1.2e18	0.968	0.102
	5		4.5e-9	0.5	1e19	1e17	1.06	0.134

In samples with tin impurity the form of curves specifies a band of traps, their concentration increases towards mid-gap according to the exponential law and also falls with increase of the optical bias level.

6. Discussion

The observed features of a transient photocurrent in As_2Se_3 amorphous films, that are associated with doping, have the general enough character and are not restricted to material of a chalcogenide film and/or impurity type (Sn), chosen in our experiment. For example, in As_2S_3 amorphous films doped with rare earth ions (Pr) [23] the shift of DOS peak to large energies and substantial increase of characteristic decay time constant of the photocurrent relaxation were also revealed. Besides the shift of deep traps distribution into mid-gap was observed in amorphous silicon too [27]. Results obtained in the present work at additional illuminations allow us to look after the occurrence of features in a photocurrent transient and to tie them with a form of DOS-distribution. So the suppression of a maximum on I(t) x t diagrams in a non-doped sample and removal of a plateau accompanying it in curves of Fig. 5a towards the smaller energy (curves 2-5) can be related to increase in the number of recombination centers due to capture of non-equilibrium electrons on gap states [16,17]. The evaluated concentration N_d~10^{16} cm^{-3}, energy position of DOS peak E_d~0,62-0,64 eV and the width of this peak $\sigma \sim 0.034$ eV seem typical for the deep centers in a-As_2Se_3, - the same as also bimolecular character of recombination. It is proved by coincidence of modeling curves with the experimental points.

Situation is different in samples with tin impurity: the trend of curve I (t) x t in Fig. 5b is monotonic and does not reveal a maximum, as in a case shown in Fig. 5a. At the same

concentration of the tail traps $N_t \sim 10^{19}$ cm^{-3} the concentration of deep centers $N_d \sim 10^{18}$ cm^{-3} is essentially larger; significantly deeply and widely a band of the deep states (see Table 1). Along with the enhancement of optical bias level the energy position of the defect band is displaced deep into the gap, which is directly opposite to the considered for As_2Se_3 sample; the value of this shift (~0.13 eV) is also large. Despite significant reduction in recombination constants we think that in this case the mechanism of photocurrent relaxation is controlled by deep capture too as experimental points well coincide with modeling curves. Based on estimations of density-of-states distribution in Fig. 5 and the parameters listed in Table 1 it is possible to assume that in doped samples despite a higher concentration of deep states the suppression of deep capture takes place. Really the effective density of states can be presented as

$$g(E)_{eff} = g(E)[1 - f(E)] = g(E)\left[1 - \frac{\tau_e}{\tau_e + \tau_c}\right] = g(E)\frac{\tau_c}{\tau_e + \tau_c}, \qquad (5)$$

where τ_c is a capture time constant, f(E) characterizes a population of deep trap. It is seen that in a case $\tau_c << \tau_e$ the optical bias can suppress effective density of states. It seems this case is realized in a non-doped sample. On the contrary, for a doped sample it is possible to assume that $\tau_c >> \tau_e$ and effective density-of-states $g(E)_{eff} = g(E)$ doesn't depend at all on a degree of the deep state filling.

The large capture time τ_c can be associated with a special nature of the centers induced by tin impurity, for example, such as pairs (Sn^{2+}-Se_2); these centers can be positively charged to have repulsive Coulomb potential for holes and therefore, - the small value of capture cross-section. This conclusion inferred from X-rays diffraction [28] and the Measbauer's spectra studies [29]. The central result of these studies is that in the samples with tin impurity Se atoms form tetrahedral coordinated structural units such as Sn $(Se_{1/2})_4$ and can promote occurrence of the new acceptor-like deep centers.

Certainly, the mechanism of shift of deep state band under optical bias is not so obvious. We believe that for its explanation the theoretical representations advanced in [30] can be used. According to these representations introduction of compensating impurity should result in formation in the band gap of semiconductor of some additional allowed states caused by increase of amplitude of an impurity potential fluctuations. These states are near the band edges and can result in shift of effective activation energy E_a to mobility edge:

$$E_a \approx E_t - \Delta E \ . \qquad (6)$$

Here $\Delta E \sim Ke^2(NR^3)^{1/2}/(\varepsilon R)$ is the depth of potential well formed by root-mean-square fluctuation of an impurity concentration with characteristic size R (N is the average concentration, K is a constant and ε. is the dielectric permeability of the semiconductor).

It is obvious, that the amplitude of fluctuations and the largest depth of potential wells are limited by size of screening radius: $R \le R_s$. As the amplitude of fluctuations decreases with increase of the concentration of charge carriers, there will be a progressing reduction of effective density-of-states and shift of activation energy deep into the gap, that is seen from Fig. 5b. In strongly compensated semiconductors reduction of radius R_s ($\sim N^{1/3}/p^{2/3}$) occurs by far faster, than at screening by free carriers. Estimations show that at typical values of $N \sim 10^{18}$ cm^{-3} and concentration of the photo-excited carriers p ($=G\tau_R \sim 10^{15}$ cm^{-3}, R_s can be about of 100 nm; appropriate values ΔE make 0.1 eV, that coincides with the found shift of DOS distribution for a doped sample. Besides, modulation of band edges by an impurity potential fluctuations in a doped sample results in a strong reductions of recombination's constant

R_{eff}=Rexp(-ΔE) [31]; the appreciated values of μ_C/R in samples As_2Se_3: $Sn_{1.0}$ more than 100 times exceed these values in As_2Se_3 sample and also decrease at additional illumination (see Table 1). As a result in a doped sample the trend of curve I (t) x t curves is monotonic and does not reveal a maximum as in a case shown in Fig. 5a for a non-doped sample. The calculations reported in [23] show, that a plateau corresponding to hyperbolic law of a photocurrent decay, and seen in this figure, can not be observed at all even if upper limit of a relaxation recording times is indefinitely large.

Thus, the examined model allows us to explain the slow term of a photocurrent decay observed in a doped sample, to tie shift of a maximum of density-of-states distribution at additional illumination by reduction of amplitude of a potential fluctuations. It essentially differs from the model of quasi-Fermi-level movement, which is usually used at interpretation of experiments with optical bias. Estimations show, that the maximal displacement ΔE_{Fp}=kTln (I_{OB}/I_{dark}) at the maximal illumination in our experiment (I_{OB}~10^{-9} A) makes only 0.03 eV whereas for an explanation of the observed DOS shifts the steady-state photocurrent must be larger (~10^{-8} A). The found out the dependences of a photocurrent relaxation rate on intensity of additional illuminations and the energy shift of DOS-distribution in doped samples As_2Se_3: Sn make this model in many respects similar to model of an activation barriers, offered for an explanation of a lot of phenomena as persistent photoconductivity [30,32], relaxation of the energy levels of deep states [33,34] and recently, - the low-frequency 1/f noise in amorphous a-Si: H films [35,36]. .

7. Conclusions

Experimental data obtained in the present work show that the characteristic time constant of carrier capture on deep traps is increased at doping of sample and determines the rate of recombination. Finally it results in occurrence of slow components in photocurrent relaxation related to phenomenon of persistent photoconductivity.

Trend of curves I (t) x t shows that position of a maximum of DOS-distribution changes at doping of samples with tin impurity and is determined by screening processes of potential fluctuations by excess charge carriers appearing at additional illumination. In non-doped sample optical bias only insignificantly shifts these distributions to the band edge.

The analysis of the photocurrent relaxation curve carried out within the model of recombination, which is controlled by deep capture allows us to estimate a number of the important characteristics of a material and to tie them with parameters of DOS distribution.

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