Photostructural changes in amorphous semiconductors studied by the modulated photocurrent method

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Abstract.   The optical bias dependence of the modulated photocurrent (OBMPC) is used to study the light-induced changes of sputtered a-As_2Se_3 and glow discharge a-Si:H films. It is found that upon strong light exposure, an increase, by an order of magnitude, in the ratio of the capture coefficient of the majority carriers by the gap states, over the free carrier mobility, c/μ, is taking place in both materials. This is interpreted as a result of the structural modifications that may take place during the light degradation enhancing the capture coefficient. The effect is completely reversible upon thermal annealing.

1. Introduction

A common property of the amorphous semiconductors is the creation of light induced metastable effects. A detailed review is given in [1]. The authors compare the effects reported in the literature for a-Si:H to those for the amorphous chalcogenides and conclude that two types of reversible light induced effect are observed. One is related to defect formation and is responsible for the metastable effects observed in the photoconductivity, the midgap absorption, the photoluminescence and the electron spin resonance (ESR) in a-Si:H and in the amorphous chalcogenide semiconductors. The other type of effect is related to the light induced structural changes. Recent EXAFS measurements in a-As_2Se_3 [2] showed a metastable increase of the average coordination number of the Se in the photoexcited state, accompanied by an increase in the disorder parameter. When the light is turned off, the initial coordination number is restored, while the disorder remains. The data are confirmed by Raman experiments and are attributed to changes in the interactions between LP electrons, which change the bond angles, leaving the length of the bond unchanged. New (wrong) bonding between similar atoms, (As–As and Se–Se (or S–S)) is formed, while the interlayer distances are affected. In the case of a-Si:H, theoretical calculations [3], as well as experimental data [4], provide evidence that, upon illumination, long range structural changes take place as well. The volume expansion observed upon illumination in a-Si:H [5] and in the amorphous chalcogenides [6] demonstrates the extended nature of the structural changes which occur in both materials. All the above mentioned light induced effects are completely reversible upon thermal annealing.

Despite the extensive studies, the microscopic mechanisms responsible for the degradation of the photoconductivity still remain unclear. A simple functional relation between the defect concentration (as measured by electron spin resonance, ESR or by the constant photocurrent method, CPM) and the photocurrent decay is not found [1]. Indeed, the dc photocurrent, i_{ph},

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depends on the density of the recombination centres, N_R and the ratio of the free carrier mobility, μ, to the capture coefficient of the majority carrier by the defect states, c, i.e. $i_{ph} \propto (\mu/c)(1/N_R)$. Usually, it is assumed that the ratio μ/c remains constant during the illumination so it is expected that the i_{ph} will follow the inverse variation of N_R. However, it is found that the i_{ph} is not related by a simple formula to the concentration of the dangling bonds (as measured by ESR) or the concentration of the deep defects (as measured by the CPM or photothermal deflection spectroscopy, PDS).

In the present work, we will show that, in both a-As$_2$Se$_3$ and a-Si:H, the ratio μ/c strongly decreases under light exposure and that it is the main reason for the degradation of the i_{ph}. In the following we will argue that the decrease of the μ/c is mainly due to the increase of c which is caused by the light induced increase of the structural disorder in those materials.

In the last few years, we have used the optical bias dependence of the modulated photocurrent (OBMPC) technique [7] in order to study the light induced phenomena in a-As$_2$Se$_3$ [8] and in a-Si:H [9]. Computer simulations of the phase shift spectra of a-Si:H [9] were made in order to determine the location of the quasi-Fermi level of the trapped majority carriers, E_t, in the frequency domain. These simulations showed also that reasonable fittings could be obtained *only* when the capture coefficient of the gap states was increased by an order of magnitude upon illumination.

In the present work the defect density is determined by the CPM, while the position of the E_t level is determined directly from the out of phase modulated photocurrent spectra without using any computer simulation. During the light soaking (LS) and the subsequent thermal annealing cycle of a-Si:H and a-As$_2$Se$_3$, a reversible shift of the E_t in the frequency domain is observed. This shift will be attributed to reversible changes to the ratio of the capture coefficient over the free carrier mobility, c/μ, induced by the structural rearrangements that take place during the illumination.

In the following, the OBMPC method is described in section 2 and the experimental details in section 3. The results are presented and discussed in sections 4 and 5 respectively.

2. The OBMPC method

The method of the modulated photocurrent (MPC) was originally developed by Oheda [10] for calculating the density of states in the amorphous semiconductors. The intensity of the bias light has to be sufficiently weak in order to keep the occupancy of the probed gap states close to that of the dark equilibrium state. According to the multiple trapping model, the observed phase shift between the modulated probing light and the resulting MPC arises from the trapping and the detrapping of the majority carriers into the empty probed gap states that are located at a depth E_ω from the respective conduction path (which is at E_C for a-Si:H and at E_V for a-As$_2$Se$_3$). At this level, which is in thermal equilibrium with the conduction path, the thermal emission rate, $e(E_\omega) = cN \exp(-E_\omega/kT)$, is equal to the modulation angular frequency, $\omega = 2\pi\nu$ and is given by

$$E_\omega = kT \ln(cN/\omega) \tag{1}$$

where c is the capture coefficient of the majority carriers by the gap states and N is the effective density of states at the respective conduction band. This equation also defines the energy scale, which depends logarithmically on c. At a constant temperature, by changing the modulation frequency, the probed energy level E_ω shifts at different trap depths and therefore various gap states can be probed.

According to the Simmons–Taylor statistics [11], the quasi-Fermi level of the trapped majority carriers, E_t, is defined as the energy where the emission rate of the majority carriers

to the respective band is equal to the capture rate of electrons and holes and is given by:

$$e(E_t) = nc_n + pc_p = \omega_t. \tag{2}$$

As the predominant contribution to the dark and the photoconductivity is from holes in a-As$_2$Se$_3$ and from electrons in a-Si:H, a good approximation of equation (2) is

$$\omega_t = pc_p \text{ or } \omega_t = nc_n \tag{3}$$

for a-As$_2$Se$_3$ or a-Si:H respectively. The density of the free majority carriers can be calculated from the dc photoconductivity, σ_{ph}; then equations (3) become

$$(c/\mu) = \nu_t(2\pi e)/\sigma_{ph} \tag{4}$$

where c stands for c_p or c_n and e is the electronic charge. By using equation (4) the ratio of c/μ can be calculated if the modulation frequency ν_t is determined experimentally. Indeed, for a given temperature and a given bias light intensity, the frequency ν_t, which corresponds to the E_t level in the frequency domain, can be determined experimentally. The quasi-Fermi level of the trapped majority carriers, E_t, separates the empty states, which act as trap-and-release centres, from the recombination centres. The transition from the trapping–detrapping to the recombination regime is shown in the MPC spectra by the levelling up of the amplitude of the MPC, i_{ac} and the decrease towards zero of the phase shift, Φ. The above transition takes place in a wide region of frequencies and usually computer simulations are needed for the accurate determination of the position of ν_t [9, 12]. In contrast, the spectrum of the amplitude of the out of phase MPC, i_{out}, exhibits a well defined maximum at the transition region. As we are interested in detecting only relative changes in the E_t level in the frequency domain during the LS, we propose to use the frequency ν_p for recording the relative changes of the ν_t.

The goal of this paper is to investigate any variations of c/μ that may be induced during the LS in a-As$_2$Se$_3$ and a-Si:H. For this purpose, the MPC spectra were recorded in various exposed states under two different bias light conditions. The bias light intensity was properly adjusted in order to produce the following.

  (i) *Constant dc photocurrent (i_{ph} = constant).* Due to the degradation of the dc photocurrent upon illumination, the intensity of the bias light had to increase in order to keep the dc photocurrent constant. As the i_{ph} was kept constant, the quasi-Fermi levels of the free and the trapped carriers were expected to remain fixed in the energy gap. Any observed shift of the ν_t, that determines the location of the E_t level in the frequency domain, may then be attributed to light induced changes in the ratio of the c/μ (see equation (4)).

 (ii) *Constant dc generation rate (G_{dc} = constant).* Due to the decay of the dc photocurrent upon light soaking, the quasi-Fermi level was expected to shift towards deeper energy levels in the energy domain or the ν_t towards lower frequencies in the frequency domain. Any other changes observed would be attributed to changes in the ratio of the c/μ (see equation (4)).

3. Experimental details

The a-As$_2$Se$_3$ films were 0.85 μm thick and were deposited by rf sputtering on water-cooled substrates. The base vacuum was 3×10^{-7} Torr, while during the deposition the Ar pressure was 5×10^{-3} Torr. The a-Si:H films were 1 μm thick and deposited at the Energy Conversion Devices (ECD), USA, by the capacitively coupled rf glow discharge technique at substrate temperature 250 °C using pure disilane at a pressure of 200 mTorr.

The composition of the a-As$_2$Se$_3$ films was found to be very close to stoichiometry and the oxygen contamination of a-As$_2$Se$_3$ and a-Si:H was less than the detection limit (0.2 at.%) of the WDS (wavelength dispersive method) which was used for the composition analysis.
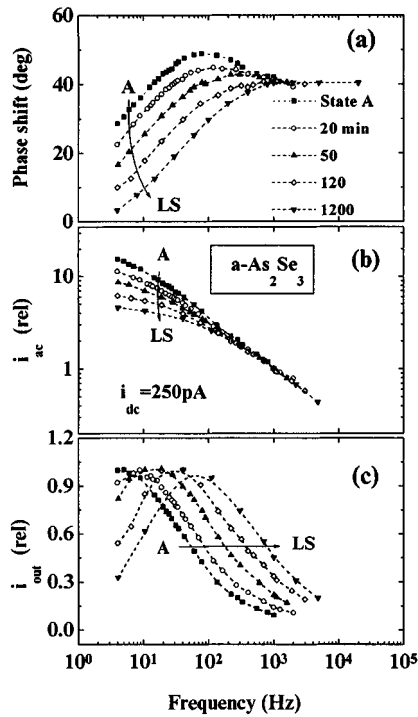
Figure 1. The MPC spectra for a-As$_2$Se$_3$ in the annealed (state A) and for various light exposed states recorded with constant photocurrent and E_{fp} at 0.65 eV. In (a) is shown the phase shift, Φ, in (b) the amplitude of the MPC, i_{ac}, normalized to the value corresponding to $\nu = 1$ kHz, and in (c) the amplitude of the out of phase MPC, i_{out}, normalized to the maximum value.
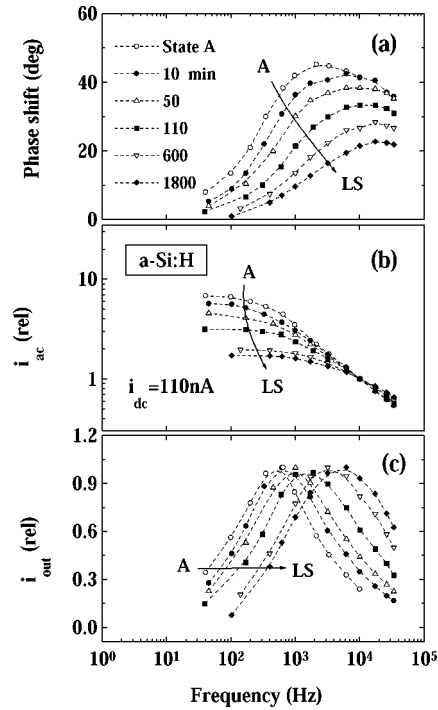
Figure 2. The MPC spectra for a-Si:H in the annealed (state A) and for various light exposed states recorded with constant photocurrent and E_{fn} at 0.55 eV. In (a) is shown the phase shift, Φ, in (b) the amplitude of the MPC, i_{ac}, normalized to the value corresponding to $\nu = 1$ kHz, and in (c) the amplitude of the out of phase MPC, i_{out}, normalized to the maximum value.

The as deposited a-As$_2$Se$_3$ and a-Si:H films were annealed at 150 °C for 4 hours and at 185 °C for 3 hours respectively, which is conventionally denoted as state A. 50 mW cm^{-2} of white filtered light from a halogen lamp and 10 mW cm^{-2} of red light from three light emitting diodes (LEDs), at 660 nm, were used for the LS of a-As$_2$Se$_3$ and a-Si:H respectively. The LS and the OBMPC measurements were made under vacuum and at room temperature. At certain time intervals the light exposure was interrupted and the samples were measured. When the LS cycle was completed, the samples underwent a step-like thermal annealing cycle at successively higher temperatures until the state A was reached.

Band gap light of 660 nm, provided by two different LEDs, was used for the probe and the bias illumination during the OBMPC measurements. The amplitude of the probe illumination was sinusoidally modulated and care was taken so that the maximum intensity was 25% or less than that of the bias light and that the intensity of the bias light did not produce measurable metastable effects. The maximum light intensity of the bias light was 10^{13} cm^{-2} s^{-1} in state A and 10^{14} cm^{-2} s^{-1} in the light exposed states. The dc photocurrent was measured with a bias light that produces a generation rate $G_{dc} = 10^{17}$ cm^{-3} s^{-1}.

Evaporated coplanar gold and aluminium electrodes with a 0.15 mm separation were grown on the a-As$_2$Se$_3$ and a-Si:H films respectively for the dark and the photocurrent measurements. To ensure that no space charge regions are formed near the contacts and
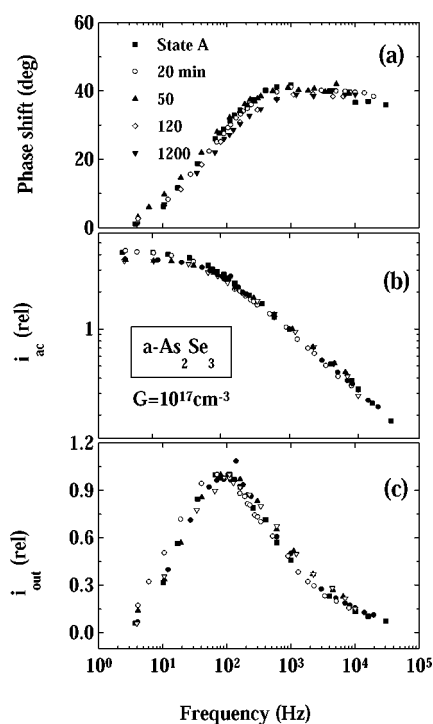
Figure 3. The MPC spectra for a-As$_2$Se$_3$ in the annealed (A) and for various light exposed states recorded under a constant generation rate. In (a) is shown the phase shift, Φ, in (b) the amplitude of the MPC, i_{ac}, normalized to the value corresponding to $\nu = 1$ kHz and in (c) the amplitude of the out of phase MPC, i_{out}, normalized to the maximum value.

that only the bulk properties of the films are probed by the OBMPC measurements, we made sure that the following requirements were fulfilled: (i) a linear variation of the dark current, the dc photocurrent, i_{ph}, and the amplitude of the MPC, i_{ac}, with the applied voltage up to 100 V, (ii) the i_{ac} to have a linear dependence on the intensity of the probe light and also to be equal to zero when the applied voltage was zero and (iii) the phase shift, Φ, to be independent of the applied voltage.

The density of the deep defects was determined by the CPM. The subgap absorption $\alpha_{1.2}$ and $\alpha_{1.3}$, which corresponds to 1.2 eV and to 1.3 eV photon energies, was used to record the changes induced by the LS in the deep defect density of the a-As$_2$Se$_3$ and of the a-Si:H respectively. The optical absorption for $\alpha \geqslant 10^4$ cm^{-1} was determined by optical transmission measurements.

4. Results

4.1. Light soaking (LS) cycle

4.1.1. Constant dc photocurrent, i_{ph} = const.   During the LS experiments, the fully annealed samples (state A) were exposed to a strong light, as described in the previous section. At certain time intervals the illumination was interrupted and the MPC spectra were recorded by using a bias light which produces a constant value of the dc photocurrent, i_{ph}, and locates the E_{fp} at 0.65 eV in a-As$_2$Se$_3$ and the E_{fn} at 0.55 eV in a-Si:H. The spectra for a-As$_2$Se$_3$ and a-Si:H

are plotted in figures 1 and 2 respectively. The spectra of the phase shift, Φ, are shown in (a), the amplitude of the MPC, i_{ac}, in (b) and the amplitude of the out of phase MPC, i_{out}, in (c).

The important feature of figures 1 and 2 is that, upon illumination, the frequency where the Φ starts to decrease towards zero, the i_{ac} to level up, with decreasing frequency, and the maximum of the i_{out}, v_p, are shifted towards higher frequencies. This implies that the E_t level shifts in the frequency domain even though the experimental conditions used were such as to keep its position fixed in the energy gap. The observed shift of the v_p, towards higher frequencies upon LS, corresponds to an increase of the ratio of c/μ (equation (4)). The evolution of c/μ with the illumination time is plotted in figures 4(a) and 5(a) (solid symbols) for a-As$_2$Se$_3$ and a-Si:H respectively and will be discussed in the followings.

4.1.2. Constant dc generation rate, $G_{dc} = $ const. After subsequent light soaking steps, the spectra of a-As$_2$Se$_3$ were recorded with a bias light which produced a constant generation rate ($G_{dc} = 10^{17}$ cm^{-3}). The data are plotted in figure 3; the phase shift, Φ, spectra are shown in (a), the amplitude of the MPC, i_{ac}, in (b) and the amplitude of the out of phase MPC, i_{out}, in (c). Spectra recorded with lower generation rates ($<10^{15}$ and 10^{16} cm^{-3}) show also similar behaviour. It is interesting to notice that all the spectra are practically identical, which means that no shift of the E_t level is observed in the frequency domain even though a degradation of the dc photocurrent occurs. According to equation (4) this could be observed only if the ratio c/μ increases. The evolution with the illumination time of the calculated ratio of c/μ is shown in figure 4(a) with the open symbols.
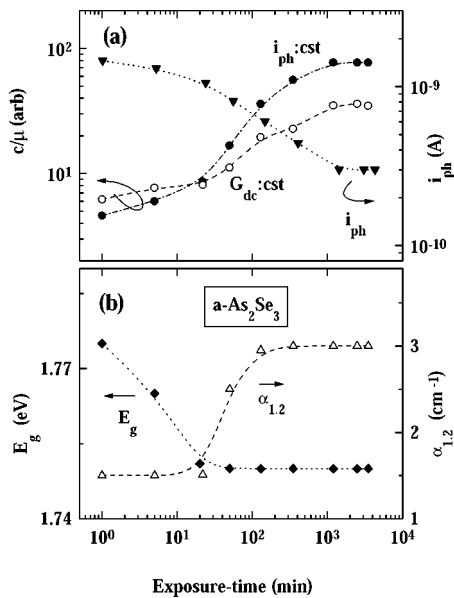

Figure 4. The summarized data of the LS in a-As$_2$Se$_3$. In (a), left axis is shown the ratio of c/μ calculated from the data of $i_{out}(v)$ recorded under a constant photocurrent (solid symbols) or under a constant generation rate (open symbols); on (a), right axis the decay of the dc photocurrent. In (b), left axis are plotted the variation of the optical gap; and on (b), right axis the change of the subgap absorption $\alpha_{1.2}$ as a function of the light exposure time.
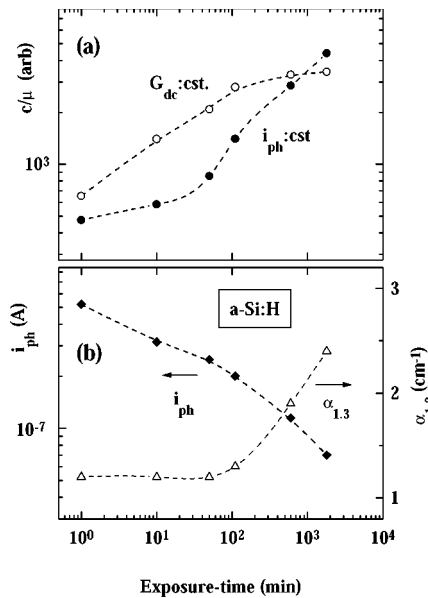
Figure 5. The summarized data of the LS in a-Si:H. In (a) is shown the ratio of c/μ calculated from the data of $i_{out}(v)$ recorded under a constant photocurrent (solid symbols) or under a constant generation rate (open symbols). In (b), left axis is plotted the decay of the dc photocurrent and on (b), right axis the change of the subgap absorption $\alpha_{1.3}$ as a function of the light exposure time.

In a-Si:H, the respective MPC spectra recorded with a constant generation rate show a minor shift of the E_t level upon illumination. From these spectra, which are not shown here, the ratio c/μ was calculated using equation (4) and is plotted in figure 5(a) with the open symbols.

For comparison in figures 4 and 5 are also shown the decay of the dc photocurrent during the light degradation and the changes of the defect density at 1.2 eV and 1.3 eV for a-As$_2$Se$_3$ and a-Si:H respectively. The variation of the optical gap (defined as the photon energy at $\alpha = 10^3$ cm^{-1}) upon LS of a-As$_2$Se$_3$, known as the photodarkening effect, is also included in figure 4(b).

We can see in figures 4 and 5 that the increase of c/μ is roughly inversely proportional to the decay of the dc photocurrent (as expected from equation (4)), while the increase of the defect density is not related to the decay of the i_{ph}. During the first 20 min of the illumination in a-As$_2$Se$_3$ (figure 4) or during the first 100 min in a-Si:H (figure 5), where the defect density is still unaffected, a weak variation of the i_{ph} and of the c/μ are observed in both materials. In a-As$_2$Se$_3$ the defect density saturates after 200 min of illumination even though the dc photocurrent still continues to decay and the c/μ to increase. Both quantities saturate after 1000 min of illumination. In a-Si:H no saturation is observed during the 2000 min of illumination of this experiment. In both materials, at the moment where the defect density starts to increase, an enhancement of the rate of change of the c/μ and of the i_{ph} is observed. We may also notice that the photodarkening effect in a-As$_2$Se$_3$ is completed in the first 20 min of illumination. At this time interval no changes in the defect density are observed.

4.2. Thermal annealing cycle

All the changes of the photoelectric properties, observed during the LS cycle and plotted in figures 4 and 5 are completely reversible upon thermal annealing. The recovery of the optoelectric properties of a-As$_2$Se$_3$ and a-Si:H upon subsequent thermal annealing is shown in figures 6(a) and 6(b) respectively. In both materials, the c/μ and the i_{ph} start to recover just above room temperature, while the defect density starts to recover only above 120 °C. The optical gap in a-As$_2$Se$_3$ (not shown in figure 6) remains constant up to 120 °C and recovers completely around 150 °C, where the state A is reached.

5. Discussion

The important experimental result of this work is that upon LS the ratio c/μ increases by approximately one order of magnitude in both materials (see figures 4 and 5), while the defect density (as measured by the CPM) increases only by a factor of two. It is also found that the evolution of the ratio c/μ and of the i_{ph}, with the light exposure time, are very similar. A higher rate of change occurs when the defect density increases. At the beginning of the illumination, even though the defect density remains constant, both the c/μ and the i_{ph} varied by a factor of two. In the case of a-As$_2$Se$_3$, in the first 20 min of the illumination, the structural changes responsible for the photodarkening effect take place. We may notice that $\alpha_{1.3}$ (figure 5) shows similar time evolution and annealing behaviour to the photoinduced spin density of a-Si:H [13]. Recent polarized electroabsorption and macroscopic stress measurements [14] showed that the structural disorder increases after illumination in a-Si:H and that the creation of the dangling bonds begins only when the large-scale network structural disorder saturates. The changes observed in the i_{ph} and the c/μ in the first minutes of the illumination may be related to the structural disorder that is induced before the increase of the defect states.

The thermal annealing data of figure 6 show that, for both materials, the i_{ph} and the ratio c/μ start to recover at much lower temperatures ($\sim$50 °C) than the defect density ($\sim$120 °C) or the photodarkening effect ($\sim$150 °C, not shown in figure 6). This also indicates that the energy barriers that have to be overcome in each case are different. The c/μ and the i_{ph} start to anneal from $\sim$50 °C and continuously recover up to 150 °C for a-As$_2$Se$_3$ or up to 175 °C for a-Si:H, indicating that the changes in the c/μ and of the i_{ph} are related to structural modifications which require a wide spread of energy barriers. In contrast, the annealing of the defects, in both materials, starts at $\sim$120 °C, indicating that the related structural rearrangements require higher energy barriers.
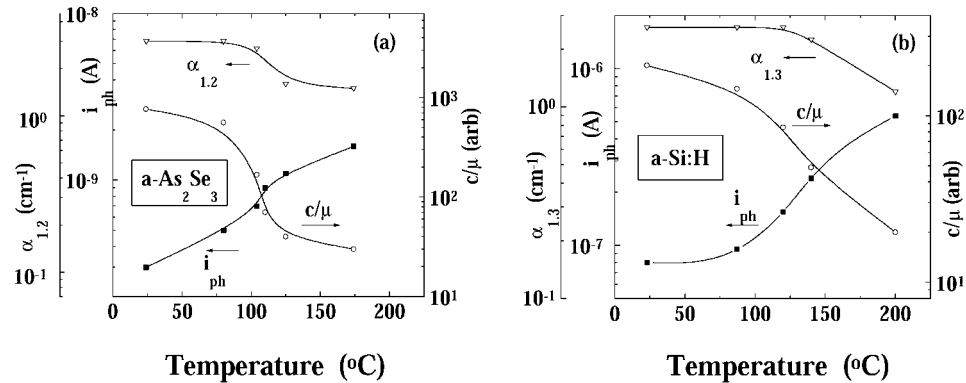

Figure 6. The summarized data of the thermal annealing experiments in a-As$_2$Se$_3$ are shown in (a) and in a-Si:H in (b). The recovery of the dc photocurrent, and the recovery of the subgap absorption $\alpha_{1.2}$ or $\alpha_{1.3}$, as a function of the annealing time, are plotted on the left axis. The ratio of c/μ, calculated from the data of $i_{out}(\nu)$, recorded under a constant photocurrent, are plotted on the right axis.

In the following we will argue that the observed increase, by an order of magnitude, in the c/μ by LS could be mainly attributed to an increase in the capture coefficient of the gap states, c, as the (average) free carrier mobility, μ, in general is considered as a constant. The free carrier or the microscopic mobility, μ, is referred to the transport of the free carriers in the band and is not affected by the presence of defects. As known, TOF experiments, in a-Si:H, show that even if the mobility–lifetime product degrades by an order of magnitude during illumination, the electron mobility remains almost unaffected [1]. Although, some researchers argued that the LS induces long-range potential fluctuations that may affect the free carrier mobility, μ, only recent data, obtained by using the photomixing technique [15], show a decrease of the mobility by a factor of 2. It is argued that this technique measures the mobility close to the 'mobility edge' and then is capable of detecting long-range potential fluctuations.

In contrast, time of flight (TOF) measurements show that upon light soaking, in the As chalcogenides [16] and specifically a-As$_2$Se$_3$ [17], the transit time of the carriers increases by an order of magnitude and is accompanied by the photodarkening effect. This increase of the transit time has been attributed [17] to the changes that occur in the broad range of the localized-state distribution and most likely are caused by the photostructural changes. The temperature dependence of the dark conductivity of a-As$_2$Se$_3$, in contrast with a-Si:H, shows no detectable changes either in the magnitude or in the slope between the annealed and the light soaked state ([17] and our data, not shown here). This indicates that the position of the dark Fermi level and the microscopic mobility are not affected by the illumination. Based on the above discussion, we could argue that mainly the changes of the ratio c/μ observed during

the LS can be attributed to changes that occur in the capture coefficient, possibly related to the accumulation of strain in the lattice during the illumination and/or to structural changes.

In the literature, a reversible increase, by an order of magnitude, in the electron-capture cross section at localized states in P-doped a-Si:H [18], upon light soaking, has been reported. The effect is restored after annealing to 200 °C. In the case of a lightly doped sample (0.01% P), the observed increase of the electron-capture cross section was not accompanied by a reversible photoinduced creation of gap states.

As the capture coefficient of the gap states depends strongly on the charged state of the defect, it could be argued that the observed increase of c by an order of magnitude occurs because charged metastable defects are created upon LS. If a second species of defect were created, below the transport path in a-Si:H, a second peak would appear in the i_{out} spectra, as is deduced by computer simulations. As we can see in figure 2, only a single maximum is present in the out of phase MPC spectra, i_{out}, and no indication of a bump was detected either at state A or at any stage during the LS, suggesting that only a single type of defect is probed. We could then suggest that the observed increase of the capture coefficient in both materials is a result of the accumulation of strain and/or of structural rearrangements, that take place during the LS. It seems that the deformed sites of the lattice or the newly formed bonds may facilitate the capture of the carriers.

In the following, we will propose some possible mechanisms, which may be responsible for the increase of c upon LS. For a-Si:H we have already proposed [9] that the observed increase of the capture probability upon LS could be attributed to an increased concentration of the strained bonds formed around a defect, which may act as a cluster of states, enhancing the effective capture radius of the defect. The carrier first is captured by one of the strained bonds, which are preferentially formed in the vicinity of a defect and subsequently by the defect [19]. Based on recent experimental data which showed that the structural disorder increases in a-Si:H after illumination [14] and on Raman and EXAFS measurements in a-As$_2$Se$_3$ [2] which showed that the photoinduced disorder in the material remains even when the light is turned off, we could argue, that the observed increase of the structural disorder could lead to an enhancement of the capture coefficient.

Finally, we would like to refer to a possible mechanism for the formation of defects. It is believed [20] that under illumination the energy released during the recombination of an electron–hole pair near a strained bond could lead to bond breaking and to lattice rearrangement. From figures 3 and 4 it is evident that, in both materials, during the first minutes of the LS there is no creation of defects whereas c/μ increases and i_{ph} decreases. This is also in agreement with ESR data in a-Si:H [1, 13]. The absence of a bond breaking process at the first stages of illumination can be taken as an indication that highly strained bonds are not present in the annealed state. Bond breaking sets in only after 20 min or 100 min of LS in a-As$_2$Se$_3$ or a-Si:H respectively, indicating that at first strained bonds or structural disorder is accumulated in the lattice, enhancing the c/μ, and later some highly strained bonds break creating defects which enhance the subgap absorption. This observation supports the idea that the accumulation of strains in the lattice is probably the precursor of the defect creation process in both materials.

6. Conclusions

In both materials it was found that upon illumination:

(i) The ratio of c/μ increases approximately by a factor of 10 (see figures 3 and 4), within 1000 min and 2000 min of illumination in a-As$_2$Se$_3$ and a-Si:H respectively.

(ii) The decay of the dc photocurrent is better related to the increase of the ratio of c/μ than

to the increase of the metastable defect density (see figures (3) and (4)). The increase of the ratio of c/μ upon illumination is attributed to an enhancement of the capture radius as a result of the light induced structural disorder. In both materials, the increase of the c/μ is related to structural modifications, which require a wide spread of energy barriers to occur, while the creation of defects is related to processes that require high energy barriers. Finally, the photodarkening effect of a-As$_2$Se$_3$ is related to processes that need to overcome even higher energy barriers than those required for the creation of defects.

Upon LS, the lattice first becomes more disordered and later the defects are formed. The reverse process takes place during the thermal annealing. First the amorphous matrix becomes more ordered and later the defects are annealed.

Acknowledgments

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