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Invited Lecture

Photoconductivity studies of discrete defect levels in amorphous chalcogenides

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The disordered nature of the amorphous semiconductors results in the presence of a significant density of localised electronic states in the energy gap between their valence and conduction bands. In the chalcogenide materials this leads to band edges being widened into tails of localised states with densities well above 10^{16} $\mathrm{cm^{-3}eV^{-1}}$ as far as 0.5 to 0.7 eV into the gap. Discrete defect levels, caused by specific co-ordination defects, often have a lower density and remain, consequently, frequently undetected by optical spectroscopies. Through the use of photoconductivity techniques, where transport and recombination processes follow the initial optical absorption, it becomes possible to locate some of those discrete defect levels.

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1. Introduction

The non-periodic lattice of amorphous semiconductors, with its distribution of bonding angles and bond lengths, generates a distribution of interatomic bonding strengths and leads to a continuous distribution of localised electronic states at the valence and conduction band edges, the so-called band tails. Since the bonding orbitals are more directly affected by the disorder than the anti-bonding orbitals, the valence band tail will be the wider one. In addition to these tail states, the lattice disorder also results in a number of atomic sites with non-standard bonding configurations – for instance 3-fold rather than 2-fold co-ordinated chalcogen atoms in the chalcogenide glasses – that will contribute energetically rather well-defined defect levels to the electronic density of states (DOS). Based on the observation of an exponential Urbach tail in the optical absorption spectrum of amorphous semiconductors, it is widely accepted that the band tails will exhibit an exponentially decreasing density of localized states in the band gap. Given that the absorption reflects a convolution of the valence and conduction bands, it is especially the wider valence band tail that is generally linked to the Urbach slope. In a number of instances the notion of an exponentially varying DOS has been reinforced by the modeling of other experimental observations. Primary examples include the transient photocurrent (TPC) experiments on As_2Se_3 [1] and the interpretation of drift mobility data sets obtained for hydrogenated amorphous silicon (a-Si:H) from time-of-flight (TOF) photoconductivity measurements [2].

Amongst the co-ordination defects, the best-known example is undoubtedly the Si dangling bond, easily detected by means of its electron spin resonance (ESR) signal. For intrinsic a-Si:H, it introduces a localized defect level near the middle of the gap. However, in most of the amorphous chalcogenide semiconductors, no such ESR signals can be observed [3]. Indeed, rather than containing defect sites of the dangling bond type, i.e. with a single unpaired electron, the lattice flexibility inherent in the two-fold co-ordinated chalcogen sites allows the material to lower its internal energy by polaronic deformation around charged defect sites that do not incorporate unpaired electron spins. Still, those 'negative-U' defects are expected to introduce their own set of discrete defect levels in the gap, involving different transition energies for optical and thermal processes due to the polaronic distortion of the site configuration upon changes in the site occupancy [4]. Unfortunately, direct experimental observation of these defect levels against the background of the band tails often proves to be difficult, as evidenced by the near-perfect exponential valence band tail deduced for As_2Se_3 [1] or the fact that the sub-gap optical absorption in As_2S_3 is largely dominated by the intrinsic presence of some 1% of As-As homopolar bonds [5]. Nevertheless, for many of the chalcogenide compounds and through the use of a range of optoelectronic characterisation techniques, it becomes possible to locate the defect levels in the gap.

In the present contribution, the use of photoconductivity techniques to determine the defect levels in the amorphous chalcogenides will be explored. Both steady-state and transient methods can contribute to that goal, with their effectiveness depending on the nature and the position of the defect. As a rule those techniques will be more successful for materials with a high photosensitivity, such as a-Se, than for compounds like the phase change material $GeSb_2Te_5$ that have (due to the high dark conductivity) a low photosensitivity.

2. Direct observation of defect levels

In a number of instances it is not only possible to just observe the presence of discrete gap states in the semiconductor, but also to determine the actual energy positions of those levels directly from the experimental photocurrent data. We call this the direct observation of those defect levels. In different ways, both steady-state and transient measurements can produce such results.

2.1. Steady-state methods

Under proper experimental conditions, the temperature dependence of the steady-state photocurrent (SSPC) in chalcogenide semiconductors, an early example of which is shown in Fig. 1, reveals two thermally activated regions with activation energies that are linked to the location of two discrete recombination centres in the semiconductor band gap [6,7]. The change-over from increasing to decreasing photocurrent occurs where the photogenerated free-carrier density overtakes the thermal equilibrium density. It was subsequently shown [8,9] that these recombination centres, one of which was considered acceptor-like and the other one donor-like, correspond to the thermally accessible levels of the negative-U centres. While such defect levels could not be resolved against the background from either the TPC spectra in a-As_2Se_3 [1] or the optical absorption in a-As_2S_3 [5], they control the behaviour of the steady-state photocurrent since the charged nature of the negative-U defects makes them much more effective recombination centres than the surrounding electrically neutral gap states. Discrete recombination levels could thus also be identified in a-As_2Te_3 [6,10], a-$GeSe_2$ [11] and a-Se [12], be it not as readily in the latter two as in the arsenic compounds. The high-temperature region is in both cases the problematic one: Due to a low photosensitivity, fluctuations of the thermally-activated dark current become more important than the photocurrent at high temperatures for the germanium selenides, while the a-Se combination of high photosensitivity and low glass transition temperature makes it difficult to let the dark current outweigh the photocurrent over a sufficient range of temperatures.

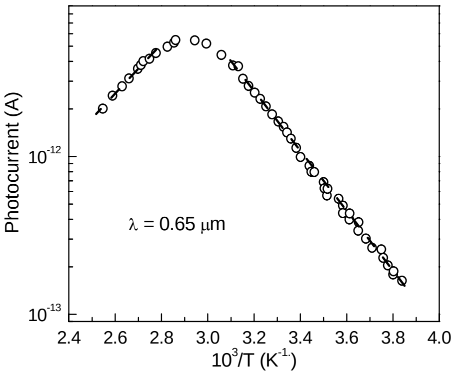

Fig. 1. Dependence on the inverse temperature of the steady-state photocurrent in a-As_2Se_3, showing the two thermally activated regions at high and low temperatures (after [6]).

The SSPC spectral distribution also offers the possibility of detecting the presence of defect levels in the gap. For instance, a predicted enhancement of the number of positively charged negative-U centres by the incorporation of negatively charged ions was confirmed by the appearance of a prominent 1.4 eV shoulder on the a-As_2Se_3 action spectrum upon the introduction of 1% of a-As_2O_3 [13]. Whenever a defect takes the form of a crystalline inclusion, sharp features in the SSPC action spectrum may be anticipated. Fig. 2 shows one such example from an a- $(Ge_2Se_7)_{88}Bi_5Sb_7$ sample, as-prepared and after annealing at 150 °C. The sharp peak in the annealed sample is due to an electronic excitation from a valence band spin-orbit-split $p_{1/2}$ orbital component to the conduction band of Bi_2Se_3 crystallites that were formed during the heat treatment. The build-up of this Bi_2Se_3-related photocurrent is, with time constants of over one hour, orders of magnitude slower than what is observed at surrounding wavelengths. Such slow processes, and the information they contain about the crystalline inclusions with them, would be missed by standard optical spectroscopy methods.

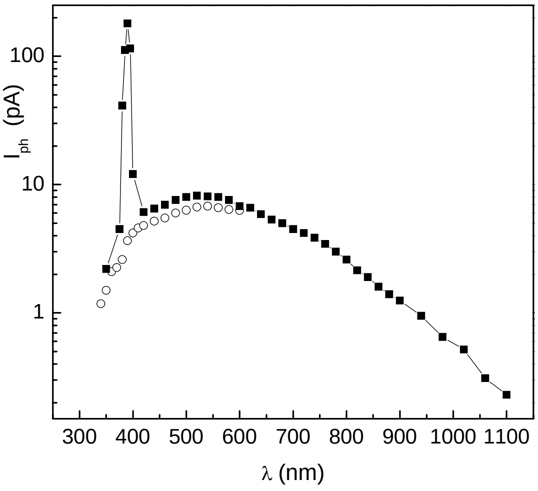

Fig. 2. Spectral dependence of the stationary photocurrent at 23 °C in an a-$(Ge_2Se_7)_{88}Bi_5Sb_7$ bulk sample, as–prepared (*) and after annealing at 150 °C (%).

Photoconductivity has the further advantage over optical absorption spectroscopy that it involves, besides photon absorption, also carrier trapping in the transport process and the eventual carrier recombination, both of which are sensitive to defect characteristics. An example of this advantage for the study of chalcogenide defect centres is shown by the normalised SSPC and thermal deflection spectroscopy (PDS) curves for a-Se in Fig. 3. The PDS data, directly proportional to the optical absorption coefficient, show a monotonic absorption increase above 1.6 eV, while the SSPC results, normalised to a constant number of incident photons, exhibit a clear minimum around 1.95 eV. The latter value corresponds to the optical gap of a-Se at room temperature. Geminate recombination is responsible for that dip as it is indeed more probable to occur for electron-hole pairs that are created with just the gap energy than for electrons and holes that are generated with higher photon energies and that subsequently move apart in thermalising to lower energy states. That geminate recombination becomes less efficient again at below-gap excitation is clearly due to the nature of the negative-U defects whereby polaronic lattice deformation at the occupied defect sites breaks up the geminate pairs. The effect of the applied voltage on the 1.95 eV dip in Fig. 3 supports the above interpretation: Higher fields will more effectively separate the photogenerated electrons and holes and thus hinder geminate recombination.

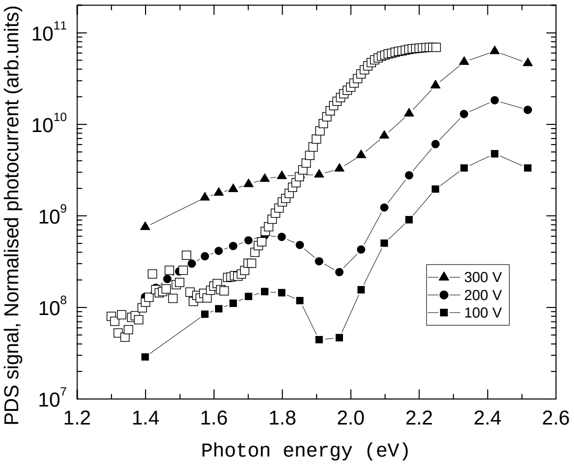

Fig. 3. Normalised photocurrents (full symbols) and PDS signal strength [14] (open symbols), at room temperature, as a function of incident photon energy for an evaporated a-Se layer.

2.2. Transient photocurrents

The analysis of steady-state photocurrent decay transients upon illumination cut-off, as frequently used for defect levels in crystalline semiconductors, is hindered in amorphous semiconductors by the wide and continuous energy range of occupied gap states. Only in the mostly narrow range of parameters that lead to monomolecular recombination is it possible to extract correct DOS information [15]. On the other hand, for transient experiments that use pulsed optical excitation, the multiple-trapping transport model in conjunction with an essentially empty distribution of localised states does allow a good determination of the DOS [16]. This can be achieved through Fourier (or Laplace) transformation of the time-domain measurements, but when sufficiently well-defined defect levels are involved, the $I(t)$ curves themselves reveal the energy position of the defect. Indeed, while for a purely exponential DOS a power-law decay of the transient photocurrent after pulsed excitation is observed, the addition of a discrete protruding feature does produce a downward cusp in this power-law current decay, with the position in time of that cusp, t_c, being linked to the energy distance E_t between the discrete trap and the transport level by $t_c = \nu^{-1}\exp(E_t/kT)$, where ν is the attempt-to-escape frequency for that trap, k the Boltzmann constant and T the temperature [17,18]. Since holes are the more mobile charge carriers in chalcogenides, they will dominate the standard photocurrent and features that are resolved from the current decay will correspond to elements of the DOS in the valence band tail. Such is the case for the TPC traces seen in Fig. 4. They were obtained with a 15 µm thick, thermally evaporated a-Se layer on a set of interdigitating gold electrodes, the whole illuminated by a 440 nm laser pulse of 6 ns duration. The cusps reveal the presence of a shallow defect level just (0.20 ± 0.02) eV above the valence band and with an attempt frequency $\nu \approx 2 \times 10^{10}$ s^{-1} that is characteristic for neutral defect sites.

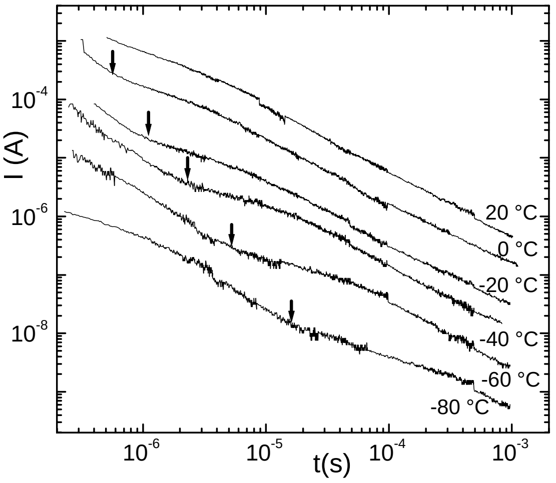

Fig. 4. Transient photocurrents after pulsed excitation of a-Se gap cell for the temperatures shown. Individual curves have been offset for clarity, with the arrows pointing to the values of t_c.

Transient photocurrents of the TOF type, whereby the sample is placed in a sandwich configuration between blocking electrodes and illuminated through a semi-transparent one, can also be used to locate discrete gap states. One advantage of the technique is that, depending on the sign of the voltage applied across the sample, either electrons or holes will drift through and generate negative or positive currents in the external circuit. It has, for instance, thus been possible to deduce the position of a shallow defect level, some 0.3 eV from the band edge, in the a-Se conduction band tail by replicating the above TPC analysis on the pre-transit part of the electron TOF signals [19]. That shallow defect also proved to be electrically neutral.

Mostly though, the TOF technique proves useful in locating deep trapping centres in the chalcogenides by means of an analysis of the photocurrents in the post-transit region, i.e. the time domain following the arrival of the majority of the drifting carriers. Since only primary photocurrents are measured in the TOF experiment, the observed current in that post-transit region reflects the emission of charge that was trapped in the deeper gap states, with a clear maximum in the current-time product $I(t)\times t$ around the time that corresponds to the maximum emission probability from a significant discrete trap. Application of this analysis to the post-transit TOF signals from a-Se sandwich cells confirmed the energy positions of the discrete negative-U levels as obtained from the SSPC results [20].

3. Indirect evidence for defect levels

Photoconductivity results can also be used in less direct ways to reveal the presence of discrete defect levels in the chalcogenide band gap. One indirect way, which allows a good estimate for the energy location of the defect level as well, consists of modelling experimental data sets. Sometimes it is the functional dependence on temperature or light intensity of the SSPC signal that points to the presence of a discrete trap, but without revealing its precise location in the gap.

3.1. Theoretical modelling

It has been generally accepted that – for temperatures above ~150 K – photoconductivity in chalcogenide semiconductors can be characterised by the concept of trap-limited band transport (also known as multiple-trapping transport) whereby photo-excited carriers are repeatedly trapped in some of the localised states in the gap and will only take part again in the conduction process after being thermally emitted to the transport path. In this process, individual charge carriers 'sample' the localised state distribution, and that sampled information is therefore contained in the photocurrent. Extracting that information from the experimental transient photoconductivity data in terms of the broad DOS background has been standard practice for many years [1], but more recently such efforts have been broadened to include the retrieval of information about discrete defect levels. The TOF experiment has been used for that purpose since its distinct pre- and post-transit regions make up a good test for any proposed DOS model.

The multiple-trapping formalism was used as the basis for both Monte-Carlo simulations [21] and analytical calculations [21,22] of TOF transients in a-Se. Simulated and calculated transients were obtained for a model DOS with an exponentially decreasing background and a number of superimposed Gaussian peaks, and compared in an iterative sequence with a set of experimental electron TOF transients. The methods used in [21] relied on a discretised version of the DOS. The analytical method of [22], on the other hand, made use of the detailed formulation of carrier transport in disordered systems for an arbitrary continuous distribution of states as described in an epochal set of papers by Arkhipov and Rudenko [23]. An example of the agreement achieved over a range of temperatures and applied fields between the experimental TOF data and the analytical calculations is shown in Fig. 5.

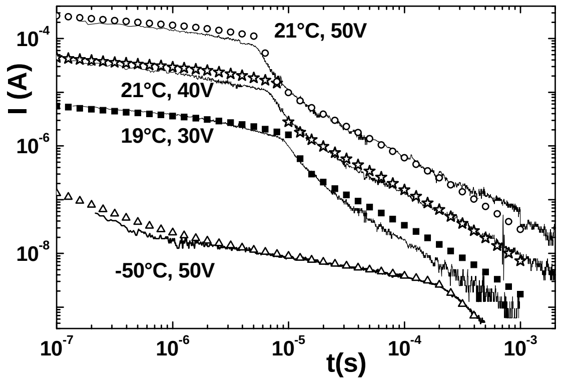

Fig. 5. Experimental electron TOF curves from an 18 μm thick a-Se sample (full lines) together with theoretically calculated photo current traces (symbols) [22]. Individual curves were offset vertically for clarity.

Both the Monte-Carlo and the analytical approaches led to the conclusion that the exponential background had to be supplemented with defect peaks at 0.3 eV and 0.5 eV below the conduction band edge in order to reproduce the TOF signals with sufficient consistency. Differences between the two methods in the relative strength of the two peaks and, especially, the width of the 0.5 eV peak could be ascribed to the fact that the results obtained in [21] were matched to TOF traces over a more restricted time domain than the one shown in Fig. 5, i.e. the one used for the analytical calculations in [22]. The 0.3 eV peak resolved here corresponds to the one mentioned in Section 2.2 above as being resolved in the time domain from the pre-transit electron TOF current, while the 0.5 eV defect matches the upper thermally accessible negative-U level that was identified in a-Se from the SSPC and post-transit TOF results [20,24].

3.2. Experimental indications

The general treatise on *Concepts in Photoconductivity* by Rose [25] discusses several instances where discrete trapping or recombination centres have a marked influence on SSPC current levels. They include phenomena such as negative photocurrents or a super-linear light-intensity dependence of the photocurrent (photo-sensitisation), both of which have been observed in chalcogenide compounds. Such observations, therefore, point to the presence of a discrete trapping level, but not necessarily to its precise energy position in the gap.

Negative photoconductivity has been reported in the past in a-GeSe$_2$ films prepared through a plasma-enhanced chemical vapour deposition process [26], and was also observed after repeated heat-treatment in the a-(Ge$_2$Se$_7$)$_{88}$Bi$_5$Sb$_7$ sample discussed above. The spectral dependence of this negative photocurrent, shown in Fig. 6, is a virtual mirror image of the one seen in Fig. 2 between 400 and 800 nm. In order for this condition to occur it is necessary that the material contain an important majority carrier trap that does not communicate with the transport path. Photo-excitation of carriers then has the effect of introducing extra minority carriers that will recombine with the dark majority carrier population, thus reducing the total current. As mentioned, only the presence and not the position of the trap can in general be inferred from this, but in the specific case of the heat-cycled a-$(Ge_2Se_7)_{88}Bi_5Sb_7$ sample, conduction band states of incipient Bi_2Se_3 grains could have the required characteristics. Potential barriers between the grains and the surrounding amorphous tissue would prevent normal communication with the rest of the conduction band. It implies of course that in a-$(Ge_2Se_7)_{88}Bi_5Sb_7$ electrons are the majority carriers. Repeated heat cycles clearly result in different intermediate states of the material.

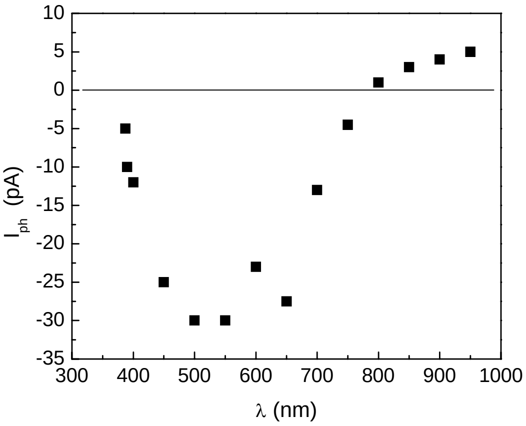

Fig. 6. Spectral dependence of SSPC after repeated heat treatment of the a-$(Ge_2Se_7)_{88}Bi_5Sb_7$ sample used for Fig. 2 [27].

Photosensitisation is to some extent the counterpart of the negative photoconductivity. Here it is required that the changes in the illumination intensity or of the temperature move the quasi-Fermi level for the majority carrier past a specific trapping level that has a large capture probability for the minority carrier, combined with a very small capture probability for the majority carrier. Recombination of the majority carrier then becomes less probable, with an enhanced photocurrent as a consequence. The phenomenon was, for instance, encountered below 10 °C for SSPC in a-Se [19] where – given the low temperature and low light levels involved – it was taken as evidence for the presence of a deep trap in the neighbourhood of the equilibrium Fermi level. The occurrence of photosensitisation (and hence the action of a discrete trapping level with the required characteristics) can be deduced from changes in the characteristic exponent γ of the Lux-Ampère relationship between photocurrent and light intensity, $I_{ph} \propto (Int)^{\gamma}$. In chalcogenides, the values of γ will normally vary from 1.0 in the monomolecular recombination regime at high temperature to 0.5 at lower temperatures where recombination becomes bimolecular. Sensitisation is then signalled by γ values above 1.0, or by rising values of γ with decreasing temperatures in the bimolecular region. Both types of behaviour have been observed in a-Se films [19,28].

Te-rich chalcogenide materials such as the $GeSb_2Te_5$ that is currently being used widely as active phase-change layer in re-writable optical storage discs do have poor photosensitivity due to their small band gap and concomitant high dark conductivity. Consequently, the photoconductivity methods described above proved unable to provide information on possible presence of discrete defect levels in the band gap of a-$GeSb_2Te_5$ samples. However, in structurally less well organised amorphous $GeSb_2Te_4$ layers photosensitisation could be detected, as evidenced by the γ values in excess of 1.0 that are displayed in Fig. 7. In other words, a-$GeSb_2Te_4$ does have at least one discrete defect level in its band gap, be it that its energy position cannot be determined.

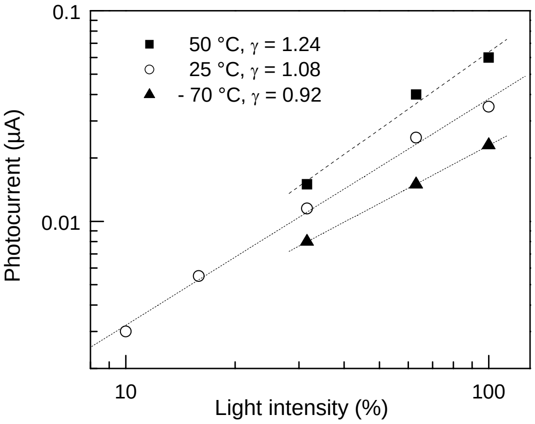

Fig. 7. Lux-Ampère characteristics of $GeSb_2Te_4$ film illuminated with 15 mW He-Ne laser.

4. Discussion

The spectrum of photoconductivity techniques offers a whole range of opportunities to detect and investigate the presence of discrete defect levels in the band gap of the amorphous chalcogenides. They have the advantage over customary optical absorption measurements, whether of the standard transmission/reflection type or by means of photo-thermal deflection spectroscopy, that they do not only reflect the optical absorption event but also additional transport and recombination aspects that may show a specific sensitivity to some of the defect centres in the material. It thus became possible to detect and locate the charged negative-U centres in the chalcogenide semiconductors due to their increased capture cross-section over the surrounding neutral trapping sites, even where they are outnumbered by the neutral tail states that dominate the optical absorption.

The elucidation of the defect level diagram in the a-Se band gap forms a particularly good example of the power of the photoconductivity methods. Whereas even the actual presence of negative-U centres in a-Se was questioned at one time [29], the combined weight of SSPC, TPC and TOF results leaves no doubt about either their occurrence or their charged and polaronic characteristics, and is actually able to reveal their energy position in the a-Se band gap as well. Indeed, similarly to the historic a-As_2Se_3 example shown in Fig. 1, two discrete recombination levels that define mono- and bimolecular recombination regimes could be identified in the a-Se SSPC, and their energy positions could be confirmed through the analysis of post-transit TOF signals [24]. The charged nature of the defects that give rise to those energy levels could be positively identified through the observation of a Poole-Frenkel effect, which predicts a weak dependence of the resolved energy on the applied field for charged centres [20]. The fact that this peak in the post-transit emission of photo-generated carriers is so easily resolved does by itself already suggest that the corresponding defect has an above average capture probability, as is of course the case for charged centres. In other words, photoconductivity data offer convincing evidence for the presence of negative-U defects in a-Se, and for the location in the band gap of their thermally accessible energy level at positions that – while in agreement with the general notions of the negative-U model – had not been resolved before. An overall comparison of the a-Se defect levels resolved from various photoconductivity experiments at the University of Leuven and the ‘standard’ model used in the literature, as quantified by Abkowitz [30], is given in Table 1. The ‘standard’ model does not contain the levels that actually relate to the negative-U centres.

Table 1. Comparison of energy positions of a-Se defect levels as assumed in the literature, and as deduced from recent photoconductivity measurements.

	Abkowitz [30]	University of Leuven
Shallow levels	$E_V + 0.26$ eV $E_C - 0.35$ eV	$E_V + 0.20$ eV $E_C - 0.30$ eV (neutral)
Thermal negative-U levels		$E_V + 0.43$ eV $E_C - 0.50$ eV (charged)
Deep levels	$E_F + 0.07$ eV $E_F - 0.07$ eV (charged)	Close to E_F [31]

It may be pointed out that the neutral shallow levels, as resolved in Leuven from TPC and pre-transit TOF data, agree with a prediction [32] that a structural anomaly whereby the lone-pair orbitals at two neighbouring Se sites would be aligned parallel rather than perpendicular does produce split-off energy levels in the gap at 0.2 to 0.3 eV from the bands. The energies shown for the shallow levels in the Abkowitz model, on the other hand, are based on misinterpretations of the hole and electron drift mobility activation energies determined by the TOF method.

With some of the techniques discussed above, simplifying assumptions that can lead to erroneous results are often made in the analysis. For instance, when using the transient techniques described in Section 2.2, the relationship $t_c = \nu^{-1}\exp(E_t/kT)$ is sometimes used with an arbitrary value of ν for lack of sufficient data at different temperatures to explicitly determine its actual value. Such was the case in [33], where a cusp in the TPC curve from a-Se was translated into a shallow state between 0.25 and 0.3 eV above the valence band by using an attempt frequency of $\nu \approx 10^{13}$ s^{-1}, rather than its (then unknown) real value near 10^{10} s^{-1}

Another widely used assumption that bears pointing out concerns the use of equal capture cross-sections for all traps in the multiple-trapping modelling. This assumption, while certainly not correct in general, would – to first order – mainly reflect on the density of a given defect rather than its energetic position. Expressing results in terms of a density and capture probability product would (formally) remedy that problem, but in practice the consequences of that assumption are considered to be sufficiently understood to allow its continuing general use.

5. Conclusion

The fact that photoconductivity is a complex process involving photon absorption, charge carrier transport and eventual electron-hole recombination, makes it a versatile tool for examining the DOS of amorphous semiconductors. It becomes especially useful in the chalcogenide ones due to the presence of charged defects with different capture cross sections from the other localized gap states.

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