About RC-like contacts in DLTS and Cu(In,Ga)Se$_2$ solar cells

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1

Abstract

The low temperature Deep-Level Transient Spectroscopy (DLTS) signal of two $Cu(In, Ga)Se_2$ samples on glass with different buffer layers is subjected to a thorough study. A similar signal is observed in the DLTS and admittance spectra of many solar cells and is usually labeled as N1. The standard DLTS theory assumes the investigated device to be a Schottky or p-n diode with an ohmic back contact, and relates the spectral components to capture or emission of free carriers by defect levels in the structure. It is well-known, though, that $Cu(In, Ga)Se_2$ thin film solar cells deviate from this ideal structure. However, even for a device like this, where advanced numerical modeling is necessary to describe the equilibrium charge distribution as a function of applied bias, a change in the free carrier concentration at a certain position the device as a result of capture or emission by deep defect levels should satisfy the detailed balance equation. The DLTS experiment performed with conventional and complemental settings for the reverse and pulse bias voltages ($V_r < V_p < 0$ and $V_p < V_r < 0$, respectively) exhibit characteristics that cannot be explained using free carrier transfer between deep levels - in the bulk or at an interface - and the conduction (electrons) or valence (holes) band of a semiconductor as a model. On the other hand, we show that for the solar cells studied here the N1 signals follow the behavior predicted for an non-ohmic RC-like contact, as established in our recent paper [J. Lauwaert et al. Journal of Applied Physics 2011] closely.

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I. INTRODUCTION

Thin film solar cells, and in particular cells with a $Cu(In, Ga)Se_2$ (CIGS) absorber layer, are very relevant as renewable energy source. At the moment cells approaching an efficiency of 20.3% can be made in laboratory environments [1], indicating that this technology may become competitive with Si solar cells in time. Among the major remaining challenges related to this type of solar cells are their electrical characterization and the understanding of their defect structure, as these have not been as thoroughly studied as a silicon solar cell structure. After two decades of capacitance spectroscopy investigation (Deep-Level Transient Spectroscopy (DLTS) and Admittance Spectroscopy (AS)) many observed signals still have not received an unambiguous interpretation. Among these is the so-called N1 signal, very often observed in CIGS cells, quite independent of their origin. Several models have been suggested for its origin [2–7], the controversy persists to date. Part of the difficulties with interpreting the DLTS and AS spectra is related with the complicated structure of thin film solar cell devices, strongly deviating from the ideal Schottky or p-n diode in the text-book explanation of capacitance spectroscopic techniques. Indeed, the N1 signal exhibits characteristics quite atypical for a majority carrier trap in the depletion layer of a diode. Recently, Eisenbarth et al. [2] suggested to assign the N1 signal observed in AS to a non-ideal ohmic, RC-like back contact in the CIGS solar cell structure. This suggestion was based on own experiments and re-interpretation of literature results and the Meyer-Neldel rule[8] was used to classify signals as N1. Their study left the question open whether the N1 signal observed in DLTS could have a similar interpretation. Intrigued by this problem, we investigated the properties of an RC-like back contact in DLTS on model devices where a junction diode is placed in series with a resistor and capacitor in parallel, on the one hand, and a second, reversely polarized diode, on the other. We demonstrated that experiments with conventional and inverted pulses (see section II for the definitions) allow to distinguish RC-like contacts from electrically active bulk defects, as origin of a DLTS signal. In particular four features appeared to be characteristic for RC-like contacts The present paper is a logical continuation of this research. We performed a set of DLTS experiments on two types of CIGS solar cells on glass substrates with different buffer layers, which should allow to identify their origin. In section three a thorough discussion demonstrates how accepting capture and emission by defect states in a bandgap within the solar cell structure leads to

contradiction, for bulk as well as for interface (surface) states. On the other hand, the experiments do follow the predictions from our previous study, characteristic for an non-ohmic contact in the solar cell structure. Hence, the latter emerges as an attractive alternative interpretation and appears to confirm the conclusions of Eisenbarth et al. [2] in AS.

II.   EXPERIMENTAL

DLTS measurements were made using a Phystech Fourier Transform-DLTS setup [9], in combination with a Boonton 72B capacitance bridge, measuring the capacitance at a fixed frequency of 1MHz. CIGS solar cells (Mo / CIGS / Buffer layer / i-ZnO / ZnO:Al / Ni/Al grid) were produced at EMPA on glass substrates where the CIGS absorbers were produced by a three-stage co-evaporation process[10]. InS buffer layer was deposited by ultrasonic spray pyrolisis [11] and CdS buffer was deposited by a chemical bath deposition. The specimens were cooled in a contact gas cryostat (Leybold and Hereaus). Specimens were relaxed in the dark in the cryostat before starting the measurement. Temperature depend measurements were recorded both during cooldown and heatup. In order to remain in this preconditioned state, forward biases have been avoided. Capacitance transients have been observed at quiescent bias V_r after a step from V_p. In conventional DLTS $V_r < V_p \leq 0$ (i.e. $V_r - V_p = \Delta V < 0$) (see figure 1 a) [12]. It should be noted that, in these conditions, for an emission transient the capacitance increases with a certain time constant: $C(t) = C_r - \Delta C \exp\left(-\frac{t}{\tau}\right)$ with $C_r > 0$ and $\Delta C > 0$. In this work we label such a regular, increasing capacitance signal as a positive signal. Besides, DLTS experiments have also been performed using inverted pulses with $\Delta V = V_r - V_p > 0$, as suggested in [13]. This type of measurements will lead to the complementary signal for the altered biases with $V_r - V_p > 0$. Figure 1 shows how voltage evolves over the structure and capacitance evolves in the DLTS experiments both in the situation when the conventional signal is positive (the complementary signal is negative) (b) and when the conventional signal is negative (complementary signal positive) (c). Figure 1 b shows the typical capacitance evolution as a function of time for a deep-level defects in the band gap of a semiconductor for a majority carrier trap with $V_r < 0$, $V_p \leq 0$ and $V_r < V_p$. Figure 1 c shows schematically the transients one observes typically for the N1 signal in DLTS, leading to a negative signal in conventional DLTS experiments.
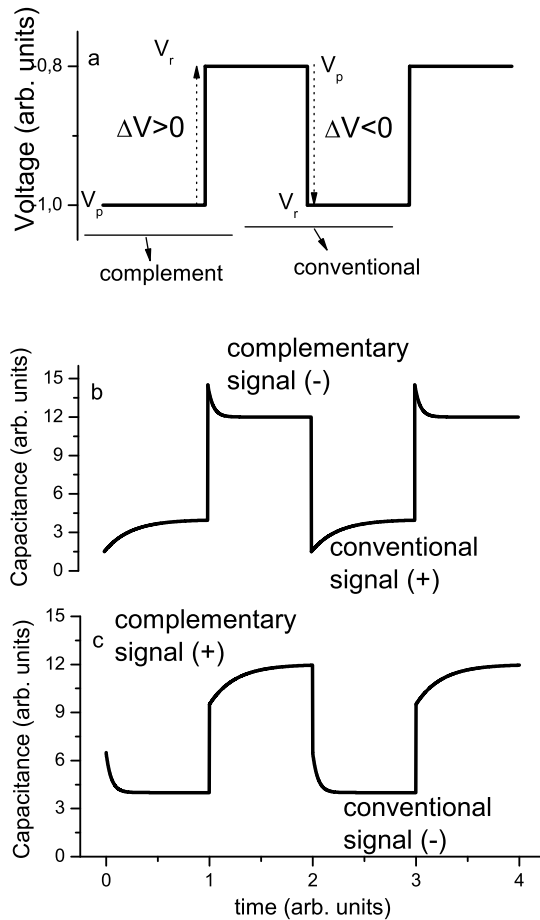
FIG. 1: (a) Time evolution of voltage and (b,c) capacitance during DLTS measurements. (b) Typical cycle for a system where the capacitance transients are due to deep-level defects. (c) Typical cycle observed for N1.

III. DISCUSSION AND RESULTS

DLTS, which in the standard theory uses $V_r - V_p < 0$[12, 14], observes emission of majority carriers from a deep level as a capacitance transient for a quiescent bias V_r after a filling pulse V_p with length t_p (see figure 1, as example). Many textbooks on DLTS use as typical example a uniformly doped semiconducting substrate in a diode structure created using a Schottky barrier. Although such assumptions make the interpretation in terms of emission of carriers from deep levels easier, they are not essential for the observation of a signal. In principle any charge transition in a diode-type device can result in a capacitance transient, which is observable with a DLTS setup if it is sufficiently slow and has a sufficiently high

5

amplitude, i.e. if it is within the detection window time and above the noise level.

To describe a CIGS solar cell structure in this discussion we will start from a more general diode structure. An inhomogeneous depletion layer is assumed that has an n-type and p-type region with a certain charge density $\rho(x)$ depending on the position (x). This charge density could for example also include charges on interface states.

For such a structure the high frequency capacitance can be written as $C = \frac{dQ}{dV}$ [14], with the total charge $Q = \frac{1}{2} \int_{-\infty}^{+\infty} |\rho(x)|dx$ and the potential difference over the structure $V = \frac{1}{\epsilon} \int_{-\infty}^{+\infty} x\rho(x)dx$. Thus one can see that any change in the charge density $\rho(x)$ in principle result in a change in the capacitance. This includes not only the emission or capture of carriers from a deep level, but also the displacements of charges with the Debye time constant $\tau = \frac{\epsilon}{\sigma}$ (σ the conductance and ϵ the dielectric constant). Thus one can see that the equilibrium capacitance at reverse bias V_r, as well as at pulse V_p and during the transients between these, strongly depends on the hole and electron concentration profiles both in the bands and on defect states, including also the interface states between layers. Advanced numerical modelling is necessary [15] to describe even the equilibrium capacitance of a thin film solar cell measured at a certain frequency. Despite all possible complications we may safely assume this junction to have n-type and p-type part or, even more general, to have a positive and negative space charge region. This implies that, even for a charge profile, the high frequency reverse capacitance (C_r) measured at quiescent bias V_r can be written as:

$$C_r = \frac{\epsilon A}{W_n + W_p} \tag{1}$$

Herein is A the area of the junction, ϵ the dielectric constant and W_n and W_p the depletion layer width in the n-type (positively charged) and p-type (negatively charged) region respectively. Even though it may be difficult to point out where the interface between the types of material is situated it might even be dependent on the bias, the structure could be more a MIS-structure it is well-established that the window (ZnO) layer of CIGS solar cells is n-type while the CIGS absorber is p-type. The carrier concentration in the window is expected to be much higher than in the absorber. Below we will also show that our argumentation is not restricted to this case. Charge neutrality in the system will thus require that the depletion width mainly extends in the p-type absorber. Therefore the capacitance for such a structure with $W_n << W_p$ can be approximated by $C_r = \frac{\epsilon A}{W_p}$. Thus one may expect that charge transitions in the heavily doped n-type region (i.e. TCO and buffer layer) will not affect the

6

capacitance strongly. The CIGS absorber is mainly p-type and if one assumes a level within the band gap or states at the interface of this negatively charged region this can result in an observable capacitance transient.

The emission or capture of a carrier by a deep level will change the charge density at a specific position locally. As a consequence the width of the depletion layer has to adapt since it has to fulfill the charge neutrality over the total structure. If one has thus a certain charge transition in this structure due to changes in the minority n or majority p carrier concentration, the time constant of the observable transient (assumed to decay exponentially) can be written as

$$\tau = \frac{1}{c_n n + e_n + c_p p + e_p} \qquad (2)$$

with e_n and e_p the emission rates of electrons and holes respectively, $c_n n$ and $c_p p$ the capture rates for electrons and holes respectively (both are proportional to the carrier concentration). Thus a single level can only result in one time constant dependent of the carrier concentrations n and p and parameters characteristic for the defect level e_n, $c_n = \sigma_n v_{th}^n$, e_p and $c_p = \sigma_p v_{th}^p$. With σ the capture cross section and v_{th} the thermal velocity of the carriers. One can only distinguish four different processes for a level leading to changes in the carrier concentrations: the emission or capture of a hole and emission or capture of an electron. For depleted p-type material having a negative charge density the emission of a hole or capture of an electron will make this region more negatively charged and thus the depletion layer will shrink, resulting in an increasing capacitance at a constant bias after the pulse this will result in a positive signal. Reversely, emission of an electron or capture of a hole results in a decreasing capacitance, thus a negative signal.

We will first examine the consequences of assuming defects in the depletion layer or at an interface as the origin of the transients labelled as N1-signals and demonstrate that in any case this leads to contradictions with experiments.

Figure 2 shows a negative DLTS signal observed for conventional DLTS parameters $\Delta V = -0.2V < 0$ ($V_r = -1V, V_p = -0.8V$) on a CIGS solar cell with a In$_2$S$_3$ buffer [11]. The signature obtained for the observed time constant with the Meyer-Neldel rule discussed by Eisenbarth et al. [2] for the N1 signal in CIGS solar cells. For $\Delta V = -0.2V < 0$ ($V_r = -1V, V_p = -0.8V$), $K_T = 1.3 \times 10^7 s^{-1} K^{-2}$; $E_T = 0.21eV$ (spectrum shown in figure 2) and $K_T = 3.5 \times 10^4 s^{-1} K^{-2}$; $E_T = 0.095eV$ for the cell with CdS buffer could be found. K_T represents the pre-exponential factor (corrected for T^2) and E_T the apparent activation
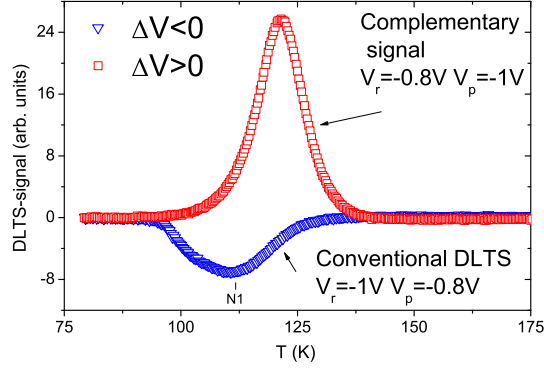
FIG. 2: Conventional temperature DLTS-scan observing both N1 $V_r = -1V$; $V_p = -0.8V$ and its complementary observed for $V_r = -0.8V$ and $V_p = -1V$
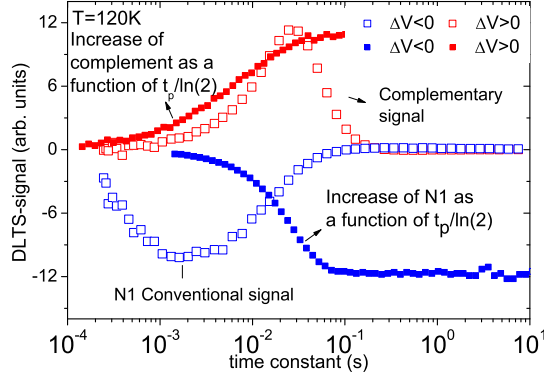

FIG. 3: Isothermal measurements at 120K of N1 ($\Delta V < 0$) for the cell with a In_2S_3 buffer and its complementary observed after $\Delta V > 0$, the increase of these signals as a function of filling pulse duration t_p (corrected for $\ln(2)$) are also included.

energy of the carrier emission rate constant: $e_n = K_T T^2 \exp\left(-\frac{E_T}{k_B T}\right)$. Since the DLTS peaks shift with varying V_r, V_p and t_p other signatures might be observed for different measurement parameters. that these signatures depend on V_p and V_r as will be shown later. It could be noted that the used T^2 correction is typical for emission of carriers and might not be useful to describe the time constant of a barrier. Due to the strongly asymmetric depletion layer, one is tempted to believe that this decreasing capacitance transient can only be the effect of emission of an electron or capture of a hole within a negative space charge region. The most

probable charge transition resulting in the N1-DLTS signal is the emission of an electron as proposed by Igalson et al. [4]. In this situation if one observes the complementary capacitance transient for $V_r = -0.8V$ and $V_p = -1V$ after sufficiently long t_p so as to start from an equilibrium situation, this transient should originate from the capture of electrons by the same levels. Hence the complementary positive signal in figure 2 would represent the capture of electrons while the typical N1 signal would correspond to the emission of electrons. For the same biases it is remarkable that the capture peak appears at higher temperature than the emission peak and that one observes a faster transient for emission than for capture. This is also clearly visible in the isothermal DLTS scan at 120K shown in figure 3, where the N1 signal is faster than the complementary signal. The standard theory does not allow this, since for emission one observes a time constant $\tau = \frac{1}{e_n}$, while the observed capture time constant is $\tau = \frac{1}{e_n+c_n n}$. Thus for a higher electron concentration the capture rate should be larger than the emission rate observed in DLTS. This renders the assignment of the N1 signal to emission of an electron from a defect level very unlikely.

The other possibility resulting in a negative DLTS signal is capture of a hole. This implies that one observes $\tau = \frac{1}{e_p+c_p p}$ for the conventional DLTS signal at $V_r < V_p$, while for the complementary signal $\tau = \frac{1}{e_p}$. Since the N1 signal is observed in reverse V_r and $\Delta V < 0$ it is very unlikely that a higher hole concentration in the negatively charged depletion layer occurs for V_r than for V_p. This also renders the possibility that the N1 signal is the effect of the capture of a hole very unlikely. Let us still assume that $V_r < V_p$ may result in a higher hole concentration. The fact that the conventional-signal has a lower intensity than its complement signal would suggest that a lower number of holes is captured, than is emitted after the complementary pulse. In DLTS it is indeed possible to observe capture from a slow capture region (as discussed in [13, 16, 17]), resulting in a considerably smaller signal. If one would observe only capture by levels from a region with a lower hole concentration, and the complementary signal corresponds to emission, the fast capture component has to be observable in the complementary signal signal (emission) if we monitor its intensity as a function of filling pulse duration for short pulses. In figure 3 the increase of the signals as a function of t_p (corrected for $\ln 2$) is also included. No fast component is observed and the increase of the N1 signal corresponds to the time constant of the complementary signal. The fact that the amplitude of the complementary signal is zero for 0.1ms filling pulse shows that no faster (i.e. normal) capture is present, while the total integrated N1 signal is remarkably
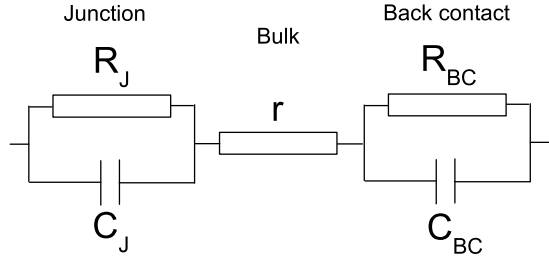
FIG. 4: Equivalent circuit for the solar cell.

smaller than its complement. This is a second, even stronger argument that the characteristics of the observed N1 signal are not compatible with hole capture lying at its origin. Even though one does not expect a wide n-type depleted region, and emission of a hole or capture of an electron in p-type would result in a positive signal, in the further discussion we will show that arguments against defects as origin of the N1 signal are still valid in these cases. Therefore we assume a charge distribution with both a n- and p-type region where W_n is not necessary negligible with respect to W_p (Eq.1). If N1 would be an effect of emission of a carrier within this structure, its complement would be an effect of capture. This leads to a contradiction with observations because it is not possible that the emission rate is higher than the capture rate from the same type of defects. On the other hand if N1 would be due to capture of a carrier somewhere in the structure, its complement would be a result of emission as proposed by Igalson [4]. In this situation it is remarkable that for the same charge transfer ($V_r = -1V$; $V_p = -0.8V$ versus $V_p = -1V$; $V_r = -0.8V$) the capture peak has a much smaller amplitude, suggesting that one is measuring a transient originating in a slow capture regime. However no faster components are observed if one records the increase of the emission peak as a function of filling pulse length.

In summary the properties of the conventional and complementary DLTS signal of N1 cannot be explained as originating directly from the change in occupation of a defect level. Although its appearance may be very well related to defects, it does not in a direct way yield information on the kinetic parameters of carrier trapping defects in the bulk structure or at the interface. Hence another explanation needs to be sought for the appearance of the signal.

Recently we have shown that also a non-ohmic (RC-like) contact in a p-n diode, other than the junction can result in a DLTS signal, with a negative or a positive sign, depending on the device parameters [13]. Figure 4 shows the equivalent circuit the model is based on.

In that paper, both the junction diode (R_J, C_J) and the reverse polarized contact diode (R_{BC}, C_{BC}) were modelled by RC parallel circuits. The model does not require that the additional barrier, although labelled as back-contact, is situated at the back of the device. The measured capacitance C at an angular frequency $\omega = 2\pi\nu$, ν=1MHz for this equivalent circuit can be written as:

$$C(\omega) = \frac{C_{BC}\left(\frac{R_{BC}}{R_{BC}+R_J}\right)^2 + C_J\left(\frac{R_J}{R_{BC}+R_J}\right)^2 + \omega^2\left(\frac{R_{BC}R_J}{R_{BC}+R_J}\right)^2 C_{BC}C_J\left(C_{BC}+C_J\right)}{1 + \omega^2\left(\frac{R_{BC}R_J}{R_{BC}+R_J}\right)^2\left(C_{BC}+C_J\right)^2} \tag{3}$$

For sufficiently high frequencies ν as applied in DLTS measurements one can see that Eq. 3 approaches the total capacitance of C_{BC} and C_J in series:

$$C \approx \frac{C_J C_{BC}}{C_J + C_{BC}} \tag{4}$$

It was verified that this high frequency condition was sufficiently met at our DLTS measurement frequency ν=1MHz. In the condition that $C_{BC} > C_J$, this can be approximated by $C \approx C_J$. Thus we may state that in the measuring conditions, the capacitance is mainly determined by the capacitance of the junction. This is in agreement with the model used in [13] where the bias over the junction $V_J(t)$ is calculated. In this situation the transient $V_J(t)$ results in a measurable capacitance transient over the total solar cell structure. In the limit of small pulse heights the capacitance transients time constant registered can then be assumed to be the same as for the change in potential drop over the junction. In this case also the amplitude can be modelled assuming that the bias dependence of the junction capacitance is that of a junction having a uniform doping profile. Although these assumptions may seem very demanding, we will demonstrate that the N1 DLTS signals observed in this work closely follow the predictions of this back contact model. In [13] we showed that in order to obtain a negative DLTS signal it is necessary that the time constant of the junction impedance is larger than that of the back contact impedance: $R_J C_J > R_{BC} C_{BC}$. For back contact diodes with $R_J C_J > R_{BC} C_{BC}$ four distinctive properties are found [13]: negative signal for $\Delta V < 0$, the time constants converge to the same value for $|\Delta V| \to 0$,
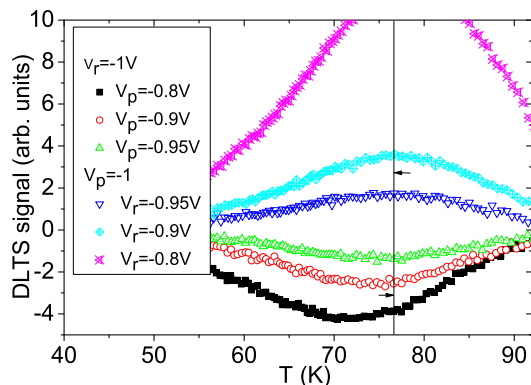
FIG. 5: Conventional DLTS spectrum showing N1 on a CdS buffer solar cell for different pulses. The complementary spectra observed for a pulse amplitude with $\Delta V > 0$ are also shown.

the signal for $\Delta V > 0$ has the largest amplitude, and the amplitude scales with $\Delta V C_r^4$ (for a uniform doping profile and $R_J \gg R_{BC}$). From the explanation above it is clear that in a certain condition a peak at $\tau = R_{BC}C_{BC}$ is expected in an isothermal DLTS experiment. In order for a signal to appear in a conventional temperature-sweep DLTS experiment, this time constant needs to be temperature dependent. As both R_{BC} (the resistance of the additional contact) and C_{BC} (the capacitance of the additional contact) maybe so, it is not unreasonable that τ_{BC} exhibits a temperature dependence. Moreover in Ref. 13 the DLTS signal of a reverse polarized back contact diode OA95 in series with a BA102 junction diode has been directly measured in a conventional DLTS experiment.

Figure 5 shows the DLTS spectrum of the N1 peak and its complement for different pulse amplitudes, including different signs observed on a solar cell with a CdS buffer. For the conventional DLTS experiments one observes a negative DLTS signal labelled as N1, and for $\Delta V > 0$ its positive complement is observed. Figure 6 shows the results of similar experiments performed on a CIGS cell with an In_2S_3 buffer.

Both specimens show a negative N1 conventional signal, with a smaller amplitude than the complement signal.

For a negligible bulk resistance one can write the observed time constant as [13]:

$$\tau = \frac{R_J R_{BC}}{R_{BC} + R_J} \left(C_J + C_{BC} \right) \tag{5}$$

Thus one concludes that for small pulse heights the time constants for the positive and the
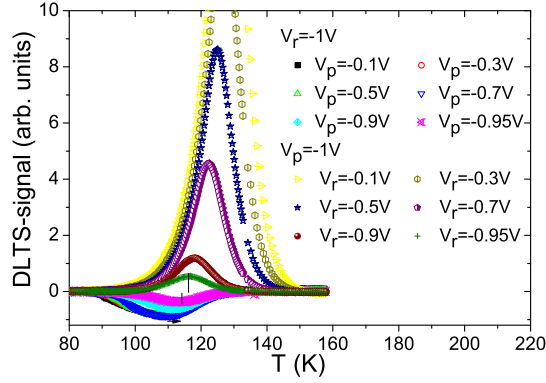
FIG. 6: DLTS spectrum showing N1 on an In_2S_3 buffer CIGSe solar cell for different pulses. Both conventional and complementary spectra are shown.
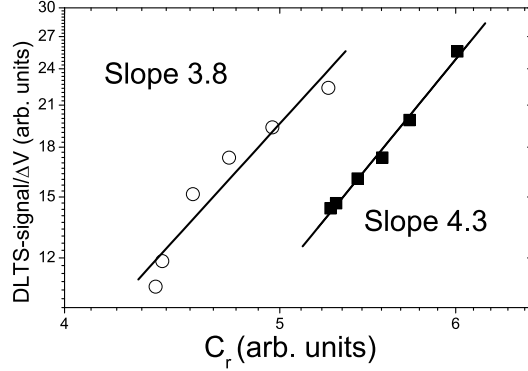

FIG. 7: Pulse corrected amplitude of the signal observed for $\Delta V > 0$ for both the solar cell with the CdS (full symbols) buffer and the In_2S_3 buffer (open symbols) as a function of reverse capacitance.

negative signals should converge to one another. A strong linearization of the equivalent circuit for a junction and an additional barrier, which is reverse polarized, leads to a capacitance amplitude that is proportional to $\Delta V \cdot C_r^4$. Figure 7 shows the pulse corrected DLTS amplitude $\frac{\Delta C}{\Delta V}$ as a function of quiescent reverse capacitance C_r. Both for the In_2S_3 and the CdS one sees that the amplitude follows the predictions by the equivalent circuit model for the RC-like contact. Hence, the N1-signal and its complement features the four typical properties predicted (and experimentally verified as can be seen in figures 4,5 and 6) for an RC-like contact [13], which is the most likely origin of this signal in the samples studied

13

here. In [2] Eisenbarth et al. suggest, based on AS results, to assign all N1-type signals in CIGS solar cell structures to non-ideal ohmic (back) contacts. Although our experiments seem to support this idea, a more systematic study in terms of number of samples should be performed before definitive statements about the nature of N1 can be made.

IV. CONCLUSION

A DLTS study of N1-signals monitored on two solar cells was presented. Based on the electronic properties observed for different signs of pulse heights and isothermal measurements as a function of filling pulse duration, the fact that this signal could directly originate from carrier emission or capture by a defect state within a bandgap in the structure is excluded. Hence, one needs an alternative explanation. In this study the properties of the N1-signal are compared with those expected for an RC-like contact. It is concluded that for the samples studied the N1-signal follows the typical properties derived for an RC-like contact closely, and that such an identification is thus very probable.

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