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Citation: Applied Physics Letters **109**, 032105 (2016); doi: 10.1063/1.4959557
View online: http://dx.doi.org/10.1063/1.4959557
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APPLIED PHYSICS LETTERS **109**, 032105 (2016)

Revisiting radiative deep-level transitions in $CuGaSe_2$ by photoluminescence

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(Received 20 April 2016; accepted 12 July 2016; published online 21 July 2016)

Recent defect calculations suggest that the open circuit voltage of $CuGaSe_2$ solar cells can be limited by deep intrinsic electron traps by Ga_{Cu} antisites and their complexes with Cu-vacancies. To gain experimental evidence, two radiative defect transitions at 1.10 eV and 1.24 eV are characterized by steady-state photoluminescence on epitaxial-grown $CuGaSe_2$ thin films. Cu-rich samples are studied, since they show highest crystal quality, exciton luminescence, and no potential fluctuations. Variations of the laser intensity and temperature dependent measurements suggest that emission occurs from two deep donor-like levels into the same shallow acceptor. At 10 K, power-law exponents of 1 (low excitation regime) and 1/2 (high excitation regime) are observed identically for both transitions. The theory and a fitting function for the double power law is derived. It is concluded that the acceptor becomes saturated by excess carriers which changes the exponent of all transitions. Activation energies determined from the temperature quenching depend on the excitation level and show unexpected values of 600 meV and higher. The thermal activation of non-radiative processes can explain the distortion of the ionization energies. Both the deep levels play a major role as radiative and non-radiative recombination centers for electrons and can be detrimental for photovoltaic applications. © *2016 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution (CC BY) license (http://creativecommons.org/licenses/by/4.0/).* [http://dx.doi.org/10.1063/1.4959557]

It is well known that in p-type Cu(In, Ga)Se_2 (CIGS) compounds, increasing Ga-content leads to reduced solar cell efficiencies.[1,2] Highest efficiencies of 22.3%[3] can be achieved with Ga/(Ga + In) ratios close to 0.3,[4] whereas $CuGaSe_2$ record cells deliver about half the value or less.[5] Due to the highest band gap, the In-free ternary compound is suited for the top cell of a stacked solar cell. But with increasing Ga-content in CIGS, the open circuit voltage loss with respect to the band gap is significantly increased.[6] One reason might be deep intrinsic defects, which can act as efficient recombination centers for generated electron-hole pairs.

Recent simulations suggest one or two deep electron traps in Cu(In, Ga)Se_2 compounds. Ga_{Cu} antisites and their complexes Ga_{Cu}-$2V_{Cu}$ are expected to be more than 350 meV below the conduction band minimum in $CuGaSe_2$.[7,8] It is shown that the energetic distance to the conduction band minimum increases with higher Gallium contents. The defect levels shift relatively closer to the mid-gap and thus are more effective for non-radiative recombination.[9] In contrast, Baekert *et al*. predicted a shallow donor level for these antisites.[10] Electrical measurements based on photo-induced current transient spectroscopy support the assumption of deep intrinsic defect levels acting as recombination centers for electrons. In Ref. 11, two activation energies in the range of 100–400 meV and 590–680 meV below the conduction band minimum were found. Photoluminescence (PL) studies on $CuGaSe_2$ found donor-acceptor-pair (DAP) transitions between 1.0 and 1.3 eV.[12,13] But estimations of the defect levels in these studies are based on calculations rather than measuring the activation energy directly. In Ref. 12, the distance of next-nearest donor-acceptor pairs for different lattice positions was used, whereas in Ref. 13, the deep levels were calculated from the energetic position of bound excitons. In this paper, we will present further insights into the possible recombination centers in $CuGaSe_2$ and their influence on the intensity dependence of the PL measurement. We will show that two deep transitions around 1.10 eV and 1.24 eV can be attributed to DAP transitions from different deep donors into one common shallow acceptor. Excitation dependent measurements support this result and will be explained by theory and a model-function. Temperature dependent measurements show activation energies of more than 600 meV. But we will conclude that non-radiative processes hinder a direct measurement of the ionization energies of the deep transitions.

The samples studied were grown by metal organic vapor phase epitaxy (MOVPE) on semi-insulating, undoped GaAs-(100)-wafer. In Refs. 14 and 15, a description of the process is given. The substrate temperature was set to 520 °C, and a growth duration of 5 h resulted in a film thickness of approx. 600 nm. The Cu/Ga ratio was tuned by adjusting the partial pressure of the Ga-source. The partial pressure of the Cu-source and the Se/(Cu + Ga) pressure ratio were kept constant. Cu-rich films were etched for 5 min in 10% KCN to exclude influences of $Cu_{2-x}Se$ secondary phases. XRD-measurements on the resulting samples suggest a high crystal quality with full width at half maximum values of 260 arcsec for the 008-reflection from the K_α-line. All results presented in this paper are based on a Cu-rich sample with a Cu/Ga ratio of 1.2 (measured by EDX before KCN-etching). Photoluminescence measurements were performed using the 514.5 nm line of an Argon-Ion laser with a focused spot

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diameter of about 80 μm (4σ-diameter).The photon flux perpendicular to the sample surface was varied between approx. 10^{15}–10^{21} cm^{-2} s^{-1} using neutral density filters. Light from the sample was collected with off-axis parabolic UV-enhanced aluminum mirrors and focused on the fiber-based input of our 303 mm spectrograph equipped with a Si-CCD and InGaAs-array. Measurements are corrected for all optical elements and detector sensitivities with a calibrated halogen lamp as the reference. Temperature dependent measurements from 10 K up to room temperature were done using a continuous-flow liquid helium cryostat.

In Fig. 1, the PL spectrum at 10 K is shown for the full measured spectral range. The peak at 1.709 eV is attributed to an exciton-transition. No intensity-dependent peak shift can be observed and the intensity increases nearly quadratic with the laser power. By adding the exciton binding energy of 13 meV,[16] the band gap is estimated to be $E_g = 1.722$ eV. The value is slightly lower than expected for a single crystal due to strain from the GaAs-substrate lattice mismatch.[16,17] Thicker samples were grown under the same preparation conditions and a shift of the exciton-peak and the XRD-008-peak towards the single crystal value was observed. But for higher thicknesses, the spectrum was disturbed by interference effects. The shallow donor-acceptor pair transitions DA2 in Fig. 1 is in accordance with the literature using the same label.[14] Several phonon replicas with a measured phonon-energy of around 30 meV are observed. For Cu/Ga-ratios above 1.1–1.2, the DA2 transition is expected to dominate the shallow transitions with an energy position at 1.62 eV. The DA2 peak shifts with increasing laser intensity by 2 meV/dec, as expected for a donor-acceptor-pair transition. Based on this low energy shift, the narrow peak width and the observed exciton transition, it can be concluded that the material is uncompensated with negligible potential fluctuations. One shallow donor at 12 meV and three shallow acceptors at 60 meV, 100 meV, and 150 meV have been shown in the past.[18] We can attribute DA2 to the transition

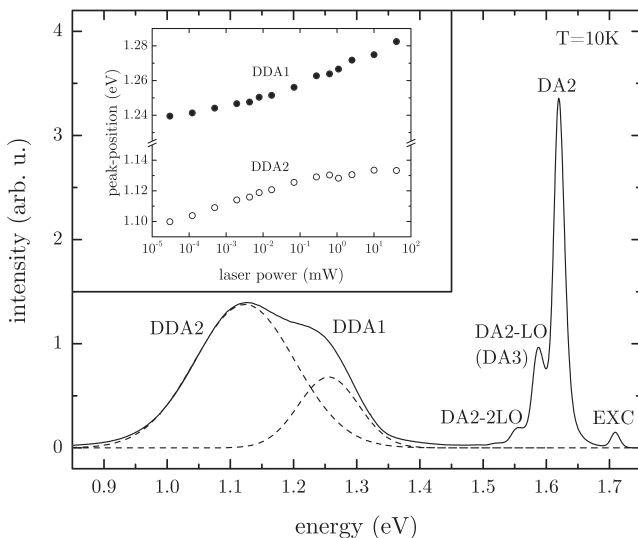

FIG. 1. Spectrum of the Cu-rich $CuGaSe_2$. Transitions marked as EXC (exciton), DA (donor-acceptor), LO (phonon replica), and DDA (deep donor-acceptor). Gaussian fits of the deep bands are represented by dashed lines. The inset shows the intensity dependent peak positions of DDA1 and DDA2.

from the shallow donor to the acceptor at 100 meV.[14] At room temperature, the dominant peak is in agreement with the band to band transition of 1.68 eV, and another peak at 1.75–1.76 eV for the second valence band[17] can be observed in our sample.

Two deep broad transitions near 1.10 eV (DDA2) and 1.24 eV (DDA1) can well be fitted with Gaussian distributions and FWHM-values of 190 meV and 110 meV, respectively. Intensity dependent measurements show a shift to higher energies around 5–7 meV/dec for both peaks (shown in the inset of Fig. 1). From this result, a donor-acceptor pair transition is assumed. The blue-shift can be due to the Coulomb-interaction between the donor and the acceptor[19] or due to potential fluctuations,[14] which were ruled out before. In the former case, high values of the blue-shift compared to DA2 can only be explained with the higher effective mass of holes compared to electrons as discussed in the following.

In Eq. (1), the recombination energy for distant donor-acceptor pairs is given with the band gap E_g, the ionization energy of the donor/acceptor $E_{D/A}$ and the Coulomb-interaction of the pair $E(r)$[19]

$$\begin{aligned} E_{DA} &= E_g - (E_D + E_A) + E(r) \\ E(r) &= \frac{e^2}{4\pi\varepsilon_0\varepsilon_r r} \leq 13.6\frac{m^*_{e/h}}{\varepsilon_r^2}. \end{aligned} \tag{1}$$

The Coulomb-energy $E(r)$ is given with the static dielectric constant ε_r (ε_0 in vacuum), the electron charge e and the pair distance r. An upper limit is set from the binding energy in the hydrogen model with the effective mass for electron and holes $m^*_{e/h} = m_{e/h}/m_0$ and the Rydberg-energy of 13.6 eV.[19] Shallow defects have a delocalized wavefunction in comparison to deep defects. The recombination probability is determined by the overlap of these wavefunctions. Therefore, the Coulomb interaction in DA-transitions is determined by the shallower level.[20] For the transition DA2, the donor-level is the shallower one. Inserting the effective mass for electrons in $CuGaSe_2$ $m^*_e = 0.13$[21] and the dielectric constant of $\varepsilon_r = 11.0$[22] into Eq. (1) results in a Coulomb energy limit of 15 meV. Since the peak-shifts of the deep transitions exceed this value by far (see Fig. 1), it is unlikely that a shallow donor is involved. The effective mass for holes $m^*_h = 0.32$[21] has to be used, if the recombination process is determined by a shallow acceptor. The upper limit for the Coulomb energy increases to 36 meV for single charge levels. For this reason, we conclude that the shallower defect in the deep PL bands DDA1 and DDA2 is an acceptor rather than a donor.

The intensity dependence of DDA1 and DDA2 is discussed in the following. Fig. 2 shows the peak-intensities of both PL-bands for varying laser power density. The linear slope in the log-log plot is described by the power-law exponent k. According to Ref. 23, transitions involving deep levels can yield exponents of $k = 1$ in low excitation and $k = 0.5$ in high excitation, respectively. This behavior can be modelled by the occupation probabilities of the donor with electrons f_D and the acceptor with holes f_A. We will derive a fitting model from the following assumptions:

(1) If re-absorption and stimulated emission can be neglected, the PL-intensity of the donor-acceptor-pair

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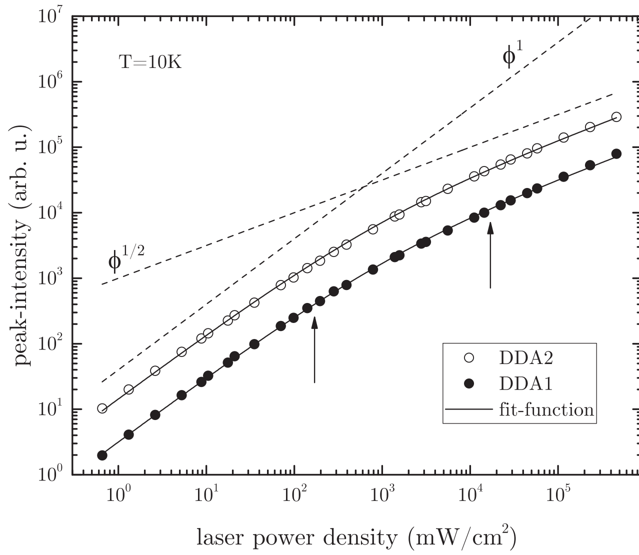

FIG. 2. Intensity dependence of the peak area from the DDA-transitions. The fit according to Eq. (4) is shown with straight lines and its limiting power laws with arbitary amplitude (dashed lines). Arrows indicate the used laser power density of temperature dependent measurements in Fig. 3.

transition is proportional to the occupation probabilities f_D and f_A[24]

$$I_{PL}^{DAP} \propto f_D f_A \propto \left[1+\exp\left(\frac{E_D-F_D}{k_BT}\right)\right]^{-1} \times \left[1+\exp\left(\frac{F_A-E_A}{k_BT}\right)\right]^{-1} \quad (2)$$

with the donor (acceptor) defect level E_D (E_A), the electron (hole) quasi-Fermi level for the donor (acceptor) $F_D(F_A)$ and the thermal energy k_BT.

(2) Thermal activation of free carriers at low temperatures is negligible compared to the photo generated excess carrier concentration.
(3) The Boltzmann-approximation is valid for one of the involved defects. In this model, the acceptor is fully occupied at high excitations and the Boltzmann-approximation is only valid for the donor (the 1 in f_D can be neglected).
(4) The exponential increase of the occupation can be replaced with an unknown power-law

$$\exp\left(\frac{F_D(\phi)-E_D}{k_BT}\right) = c_D\phi^{k_D}$$
$$\exp\left(\frac{E_A-F_A(\phi)}{k_BT}\right) = c_A\phi^{k_A} \quad (3)$$

with the constants $c_{D/A}$, the laser flux density ϕ, and the constant power-law exponents $k_{D/A}$.

Eq. (2) can be rewritten in the following form:

$$I_{PL}^{DAP} \propto c_D\phi^{k_D}\frac{1}{1+c_A^{-1}\phi^{-k_A}} = \frac{c_Dc_A\phi^{k_D+k_A}}{1+c_A\phi^{k_A}}. \quad (4)$$

For low excitations where $c_A\phi^{k_A} \ll 1$, the power-law exponent is determined by the donor and the acceptor occupation. In this case, the Boltzmann approximation can be used for both defects and Eq. (4) converges to $I_{PL}^{DAP} \propto \phi^{k_D+k_A}$. At high excitations ($c_A\phi^{k_A} \gg 1$), the injection of more and more excess holes leads to a saturation of the acceptor level and the intensity dependence follows $I_{PL}^{DAP} \propto \phi^{k_D}$. At this point, the PL signal can only increase due to more excess electrons. The transition point is determined with $\phi_0 = c_A^{-1/k_A}$. In Fig. 2, the intensity dependencies are fitted with Eq. (4) and fixed k-values of 1/2 for the donor- and the acceptor-level. Free parameters for $k_{A/D}$ also result in values close to 1/2 with deviations below 10%. The difference of the transition point ϕ_0 in both curves is small compared to the range of excitation intensity: DDA1: 1190 mW/cm^2, DDA2: 870 mW/cm^2. From this, it is concluded that one common defect gets saturated. Furthermore, the power-law exponent for the shallow DA2 transition is reduced by 1/2 in the same range of excitation. This leads to the conclusion that the acceptor at 100 meV is involved in all of the three observed donor-acceptor-pair transitions. At highest excitation, this acceptor is completely occupied by holes, and the DA intensities can only increase with higher occupations of the donor-like levels with electrons. One shallow donor is involved in DA2, and two deep donors are involved in DDA1/DDA2.

In Fig. 3, the integrated peak intensities of DDA1 and DDA2 are shown in the temperature range of 150 to 300 K. Arrows in Fig. 2 mark the laser power density used for the exemplary low and high excitation case. The activation energies were determined by linear fits of the high-temperature slope in the Arrhenius plot. Appropriate fitting results were also achieved by fitting the full curves with models of two activation energies. But the resulting activation energies are not in accordance with the peak positions according to Eq. (1), even if reasonable values for the Coulomb-interaction are considered. We assume that the temperature dependence has a more complex origin.

The thermal quenching depends on the excitation power for both transitions. With decreasing excitation, the strong intensity drop occurs at lower temperatures and fitted activation energies appear to be higher. A further decrease in the

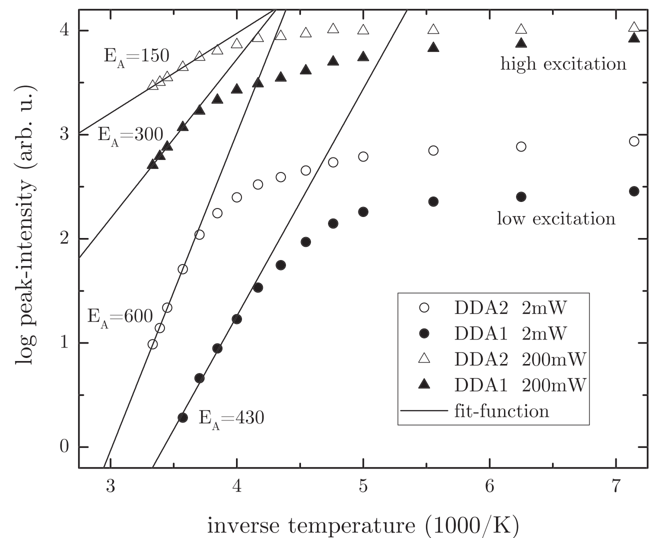

FIG. 3. Logarithm of the peak area from the DDA-transitions versus temperature. Examples with different laser power densities are marked as low and high excitation (arrows in Fig. 2). Linear fits at high temperatures are shown (straight lines) with the activation energies [meV].

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laser power below 2 mW results in activation energies up to 750 meV. These high activation energies are likely an artifact of non-radiative recombination channels, rather than thermal activation to the bands. We assume that these energy values do not reflect the defect levels and we will follow the arguments given in chapter II.B.1 in Ref. 25. The author explains the possible origin of unexpected high activation energies (strong quenching) as well as tunable quenching-points for p-type semiconductors involving deep donors.

When the involved shallow acceptor starts to become thermally activated at a temperature T_0, the density of free holes in the valence band increases rapidly. Electrons which are captured by the deep levels can now easily recombine non-radiatively with the free holes. The occupation of the deep donors with electrons decreases significantly. The thermal activation of the involved shallow acceptor opens the "bottleneck" for the non-radiative recombination channel of the deep donors. In Fig. 3 around 220 K or above the thermal quenching occurs for low excitations. One can assume that in our case, the intensity of the deep donor-acceptor-pair transition is not only reduced by the thermal activation of the shallow acceptor. At the same time, the population of the deep donor level decreases drastically. Since the PL intensity of the pair transition is proportional to both occupations, the simultaneous emptying of both defects results in an intensity drop in a narrow temperature range. Faster emptying of the shallow acceptor results also in faster emptying of the deep donors at the same time. This effect can result in the observed unphysically high activation energies of up to 750 meV and it can explain the tunable quenching-point. Conventional models for the thermal activation cannot be applied.

In conclusion, excitation dependent together with temperature dependent measurements support the assumption that the deep PL transitions DDA1 and DDA2 arise from deep donor levels into the same acceptor around 100 meV above the valence band. From a simple theory of occupation probabilities, a fitting function could be derived to describe the saturation of defects in intensity dependent measurements. From these results, we can conclude that the common acceptor becomes saturated at high excitations, rather than the deep donors. This can also be a possible origin for a limitation of the quasi-Fermi level at room temperature since the deep donors do not become completely filled and act as recombination centers for electrons. It is possible to estimate a lower energy limit for the deep defect levels, if one neglects the Coulomb interaction between the donor and the acceptor in Eq. (1). At lowest laser intensities, the average pair distance is expected to be maximized, resulting in a minimal energy reduction by the Coulomb-term. Inversely, phonon-interactions can shift the observed transition energies to lower values but it will be not considered here. Inserting values of $E_g = 1.72$ eV, $E_A = 0.1$ eV, and $E_{DA} = 1.09$ (1.23) eV in Eq. (1) leads to defect energies of $E_{DD1} < 530$ meV and $E_{DD2} < 390$ meV below the conduction band minimum. Similar values for electron traps are found by Krysztopa *et al.*[11] with $E_1 = 550$–680 meV and $E_4 = 100$–400 meV. Calculated charge transition levels from Huang *et al.*[8] for Ga_{Cu} (620 meV) and Ga_{Cu}-$2V_{Cu}$ (380 meV) below the conduction band minimum can be assumed as possible intrinsic origins for the deep electron traps. Also, the theoretical results from Pohl and Albe[7] suggest that the antisite Ga_{Cu} (420 meV) is the dominant recombination center for electrons in $CuGaSe_2$. Varying preparation conditions with a similar MOVPE-process from Ref. 26 leads to p-type doping-densities in the range of 1×10^{16} to 2×10^{18} which corresponds to dark Fermi levels of 50 to 150 meV above the valence band. Taking this into account, according to Pohl *et al.*, the antisite has low formation enthalpies, even under Cu-rich conditions. Since both theoretical defect studies expect the antisite to form the same or a slightly deeper defect level than the complex with copper, we propose that Ga_{Cu} is involved in DDA2 and Ga_{Cu}-$2V_{Cu}$ in DDA1.

This work was funded by the Fonds National de la Recherche Luxembourg (FNR) in the framework of the ODD project.

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