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Time-resolved photoluminescence measurements for determining voltage-dependent charge-separation efficiencies of subcells in triple-junction solar cells

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Conventional external quantum-efficiency measurement of solar cells provides charge-collection efficiency for approximate short-circuit conditions. Because this differs from actual operating voltages, the optimization of high-quality tandem solar cells is especially complicated. Here, we propose a contactless method, which allows for the determination of the voltage dependence of charge-collection efficiency for each subcell independently. By investigating the power dependence of photoluminescence decays, charge-separation and recombination-loss time constants are obtained. The upper limit of the charge-collection efficiencies at the operating points is then obtained by applying the uniform field model. This technique may complement electrical characterization of the voltage dependence of charge collection, since subcells are directly accessible.

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photo-diode (APD) for detection. Appropriate short and long-pass filters were used to selectively detect the PL of InGaP and GaAs subcells. Second harmonic generation in a beta-barium borate crystal was used to obtain 400 nm. Fast decay components were checked with a tunable laser system (repetition rate 200 kHz) and a streak camera, using filters and a monochromator. The excitation spot sizes, determined by a Si charge-coupled device, were varied between \( \approx 15 \) \( \mu \)m and 0.65 mm in diameter (full width at half maximum, hereafter \( \square \)). The PL detection spot was at the center of the excitation spot, extending about \( \square 13 \) \( \mu \)m. All measurements were performed at room temperature.

Figure 1 shows EQE spectra of the bottom Ge, middle GaAs, and the top InGaP junctions. High EQE values indicate a high-quality device. PL spectra of the subcells are also shown in Fig. 1; the InGaP bandgap energy is 1.8 eV (\( \lambda_{\text{InGaP}} = 690 \) nm), and that of GaAs is 1.42 eV (\( \lambda_{\text{GaAs}} = 875 \) nm). Overlap of EQE spectra is observed for wavelengths with incomplete absorption in a single subcell or photonic coupling between subcells.\(^{18}\) The PL peak positions are consistent with the EQE cutoff wavelengths.

Figure 2(a) shows the time-resolved PL of InGaP for an excitation spot of \( \square 140 \) \( \mu \)m at 400 nm with excitation-power densities varied from 0.11 to 7.6 W/cm\(^2\). No additional PL features have been observed for times later than shown. For all powers, GaAs is only weakly excited via spontaneous emission from InGaP, and no laser leakage occurs. As shown in Fig. 2(a), at 0.11 W/cm\(^2\) a single-exponential fast decay with a time constant of 0.25 ns is observed. At intermediate powers, a second decay component appears, which we denote as slow decay. For elevated powers, the slow decay becomes longer and more pronounced. At 7.6 W/cm\(^2\), two single-exponential decays are observed. The saturated slow decay has a time constant of 7 ns. Other decay features are the initial ultrafast decay (\( t < 0.2 \) ns, due to surface recombination, but not resolved) and the small tail towards the end of the decays (this may be, for example, PL due to a diffusion limited recombination process), but these are not discussed in this letter.

In Fig. 2(b), the PL behavior is analyzed with respect to the power dependence of the slow and fast time constants. The decay was measured at time \( t = 0.9 \) ns, which suppresses the influence of initial-measurement artifacts due to instrument response time. At low excitation-power densities (<1.2 W/cm\(^2\)), a constant fast decay is observed. This constant region defines \( \tau_1 \). Measurements with a streak camera revealed that \( \tau_1 = 0.08 \) ns is faster than the resolution of the APD system (about 0.25 ns). At higher power densities, a slower decay is observed; at 6 W/cm\(^2\), the slow component saturates, defining \( \tau_2 \). Since \( \tau_2 \) was same for repetition rates 8 MHz and 800 kHz, the effect of long-lived carriers is considered to be small.

Figure 2(c) shows that \( \tau_2 \) depends on the excitation spot size. The solid and dotted lines are a guide to the eye (a basic linear function), indicating that \( \tau_2 \) becomes longer for larger spot diameter \( d \), which is attributed to lifetime-enhancing effects such as photon recycling. By decreasing the spot size, the excitation volume decreases drastically, and thus effects from photon recycling are suppressed. For InGaP, we estimate the intrinsic time constant \( \tau_0(d = 0) = \tau_{\text{total}} = 2 \) ns.

FIG. 2. (a) Time-resolved PL for InGaP. Spot size: \( \square 140 \) \( \mu \)m. Excitation: 400 nm, power densities ranged from 0.11 to 7.6 W/cm\(^2\). (b) Power dependence of slow and fast components of InGaP. Spot size: \( \square 140 \) \( \mu \)m. Green triangles: streak-camera measurements. Dotted lines indicate constant values of \( \tau \). (c) Spot-size dependence of saturated time constant \( \tau_2 \) in the InGaP subcell. Black lines are guides-to-the-eye using a linear function. (d) Time-resolved PL for GaAs. Spot size: \( \square 390 \) \( \mu \)m. Excitation: 800 nm, power densities changed from 0.09 to 4.4 W/cm\(^2\). (e) Power dependence for slow and fast components of GaAs. Spot size: \( \square 390 \) \( \mu \)m. (f) Spot-size dependence for the saturated time constant \( \tau_2 \) in the GaAs subcell.
Similar dynamic behaviors are also observed in the GaAs subcell. The time-resolved PL obtained for the excitation of GaAs at 800 nm is shown in Fig. 2(d). Here, the excitation spot size was $\sim 390 \mu m$, while power density was varied between 0.09 and 4.4 W/cm$^2$. A fast decay is observed at low excitation-power densities and a slow decay component appears at high excitation-power densities. The initial PL peak ($t < 2$ ns) found at higher power densities is assigned to PL from the entire junction, before fast carrier re-distribution settled down.

Figure 2(e) shows the power dependence of slow and fast decay-time constants. For power densities up to 0.5 W/cm$^2$, the decay was measured at about 2 ns. Above 0.5 W/cm$^2$, the slow decay was measured at 4.0 ns to suppress influence from the initial PL from the entire junction. The constant value at low power ($< 0.3$ W/cm$^2$) defines $\tau_1 = 0.29$ ns, and the saturated (>3 W/cm$^2$) value of the slow component defines $\tau_2$.

The excitation spot-size dependence for GaAs is shown in Fig. 2(f). Below 30 $\mu m$, the slow time constant abruptly shortens, which is attributed to in-plane bulk diffusion. This region is excluded when determining the built-in voltage. This limit occurs because $\tau_1$ is mainly determined by the built-in electric field, as confirmed by the power dependence in Figs. 2(b) and 2(e). If diffusion of photogenerated carriers from the p and n regions becomes important, which occurs under higher illumination powers, the effective drift velocity will be reduced and a smaller voltage has to be used for normalization, i.e., only when both drift and diffusion are considered, the use of $V_{oc}$ is possible. At $V = V_{oc}$, the last term becomes zero and $\tau_{loss}$ dominates the slow PL decay.

The analytical shape of $V (I_{PL})$ depends on the sample, but may be approximated with that obtained from the Boltzmann relations:

$$I_{PL} \propto \exp \left( \frac{qV(I_{PL})}{k_BT} \right),$$

where $q$ is the elementary charge, $k_B$ is the Boltzmann constant, and $T$ is the sample temperature. The physics embodied by this idealized equation is simple, yet serves as a good test to elucidate our interpretation of $\tau_1$ and $\tau_{loss}$. Several solutions for Eq. (1) are shown in Fig. 3(b). Here, we used parameters near to the experimental values for InGaP ($\tau_1 = 0.08$ ns, $\tau_{loss} = 2$ ns, $V_g = 1.8$ V). For low intensities, a fast decay is observed, while a slow decay becomes dominant for high intensities. The main features of the experimental PL decays can be directly explained by the uniform field model, and thus we believe it is valid.

Here, we used the simplified rate equation (Eq. (1), which neglects diffusion) for a clear understanding of the physical processes determining charge collection. A detailed system of rate equations, accounting for the different regions of the junction, is important for a rigorous test. While $\tau_2$ is an indicator of the losses in the AR, the fast decay observed at high excitation power is strongly influenced by losses in the p and n regions and also diffusion from there. The additional analysis of this decay enables more accurate description of the solar cell under steady-state condition.

Using the uniform field approximation, the charge-collection efficiency of each subcell, which is a function of the subcell operating voltage $V$, is given by

$$\eta_e(V) = \frac{1}{1 + \frac{\tau_1}{\tau_{loss} V_g} \left( 1 - \frac{V}{V_g} \right)},$$

directly derived from Eq. (1). Since we defined $\eta_e$ with $\tau_1$, back-diffusion losses are not included. Consequently, a
well defined upper limit of the charge-collection efficiency is obtained, useful for subcell analysis. As mentioned above, the definition with $\tau_1$ also requires $V$ to be used. The voltage dependence of $\eta(V)$ for both subcells is shown in Fig. 4. The VDCC of GaAs (Fig. 4, red line) has a very high and flat efficiency for low voltages ($V/V_g < 0.8$) and quickly drops to zero for higher voltages. As with the $I$–$V$ curves, the fill factor (FF) of the GaAs VDCC curve is large, indicating high carrier mobility. The VDCC of InGaP (Fig. 4, blue line) shows a reduced overall efficiency and a smoother drop to zero, indicating stronger dependence on nonradiative recombination differences between EQE and $\eta(V)$ are always included in EQE measurements, but their influences are suppressed in optical measurements. Therefore, the differences between EQE and $\eta(V)$ are helpful for the analysis of transport losses, i.e., series resistances.

Additionally, we stress that for achieving maximal efficiencies, the differences due to voltage changes at the operating point can be crucial for current matching. In subcells with low FF, the VDCC efficiency itself changes significantly, even for small voltage changes (for example, the InGaP curve in Fig. 4 has a comparable steep drop near the point of maximum power $V_m$). Thus, the evaluated VDCC curve represents a more accurate description of the device than the constant EQE value. In other words, the accuracy of the VDCC efficiency is derived from voltage-dependent behavior close to that of the real device, rather than from a highly accurate lifetime determination.

By measuring PL decays, the two essential parameters $\tau_1$ and $\tau_{loss}$ are obtained. The mobility-limited charge-collection efficiencies near the actual operating voltages are calculated, and can be used to analyze $I$–$V$ behavior. We found significantly smaller values at the operating point than at SCC, which accounts for additional voltage drops at the operating point, and thus explains losses in subcells that have not been predicted by conventional electrical evaluation.

In summary, we measured PL decay curves as a function of excitation intensity from the top and middle junctions of a triple-junction solar cell designed for use in space. Since the carrier-density dependence of the PL decay corresponds to the voltage dependence of carrier separation and recombination, we evaluated the voltage dependence of the charge-collection efficiency for as-grown subcells, including their interfaces. An experimentally obtained carrier-extraction behavior should enable more accurate device simulations and improvements compared to other methods relying on reference cells or light biasing.

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FIG. 4. VDCC efficiencies for $\eta(V)$ of InGaP ($\tau_1 = 0.08 \text{ ns}$, $\tau_{loss} = 2 \text{ ns}$) and GaAs ($\tau_1 = 0.29 \text{ ns}$, $\tau_{loss} = 12 \text{ ns}$) subcells. The values for the point of maximum power ($V_m \approx 0.6$ and $0.7V_g$ for GaAs and InGaP, respectively) are shown as open black circles.

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