Letter

Electrothermal Feedback and Absorption-Induced Open-Circuit-Voltage Turnover in Solar Cells

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Solar panels easily heat up upon intense solar radiation due to excess energy dissipation of the absorbed photons or by nonradiative recombination of charge carriers. Still, photoinduced self-heating is often ignored when characterizing lab-sized samples. For light-intensity-dependent measurements of the open-circuit voltage (Suns-V_{OC}), allowing us to characterize the recombination mechanism, sample heating is often not considered, although almost 100% of the absorbed energy is converted into heat. Here, we show that the frequently observed stagnation or even decrease in V_{OC} at increasingly high light intensities can be explained by considering an effective electrothermal feedback between the recombination current and the open-circuit voltage. Our analytical model fully explains the experimental data for various solar-cell technologies, comprising conventional inorganic semiconductors as well as organic and perovskite materials. Furthermore, the model can be exploited to determine the ideality factor, the effective gap, and the temperature rise from a single Suns-V_{OC} measurement at ambient conditions.

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Photovoltaic devices convert absorbed photons from the Sun into electrical power. During this process, unavoidable thermalization processes lead to self-heating. For silicon solar cells, the relaxation of charge carriers created by photons with energies larger than the optical gap accounts for about 30% of the absorbed energy [1]. Furthermore, the radiative fraction is well below 1% of the total recombination current for most technologies [2]. This leaves nonradiative losses as the dominant recombination mechanism, leading to a temperature increase of photovoltaic devices under illumination. The influence of temperature on solar-cell parameters, for instance on the open-circuit voltage, is well known [3]. For most material systems, higher temperatures are unfavorable for the power-conversion efficiency due to the decrease in V_{OC}, making active cooling of the devices an option [4,5]. In a few cases, some technologies, such as organic solar cells, can benefit from higher temperatures, at which thermally activated hopping transport becomes more efficient, resulting in increased fill factors, which can counteract the drop in V_{OC} in limited temperature ranges [6,7].

At open circuit, almost 100% of the energy is converted into heat since an external current flow is suppressed. As a consequence, internal or external series resistances do not alter the open-circuit voltage (V_{OC}), a fact exploited in Suns-V_{OC} measurements [8,9]. With this technique, the V_{OC} is recorded for various intensities and photocurrents, yielding a pseudo-current-voltage curve that follows the Shockley equation for open-circuit conditions, i.e., $J(V_{\mathrm{OC}}) = 0$:

$$J_{\mathrm{ph}} = J_{00} \exp\left(\frac{qV_{\mathrm{OC}} - E_a}{n_{\mathrm{id}} kT}\right). \tag{1}$$

Hereby is J_{ph} the photogenerated current at a specific incident illumination intensity (I_{Sun}) and E_a describes an activation energy corresponding to the effective band gap at which recombination takes place, while J_{00} is the maximum theoretical recombination current. Suns-V_{OC} or the more elaborate J_{sc}-V_{OC} measurement are widely used among many material systems and solar-cell communities. These techniques have been introduced to study recombination processes, e.g., by determining the ideality factor n_{id}, or to obtain intrinsic charge-carrier densities of organic solar cells [10,11]. Temperature-dependent Suns-V_{OC} measurements

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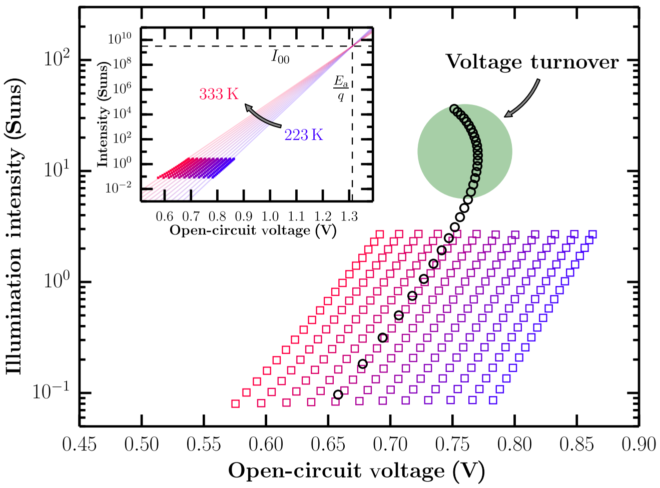

FIG. 1. Suns-V_{OC} measurement of an organic solar cell based on a F4-ZnPc:C_{60} blend and an area of 6.44 mm^2 for different ambient temperatures (colored squares) from 223 to 333 K in steps of 10 K. Using a global fitting routine according to Eq. (1), it is possible to determine the activation energy E_a. Black circles present a Suns-V_{OC} measurement taken at ambient temperatures using a brighter illumination source. At high light intensities, saturation of V_{OC} and a turnover is observed.

(see colored squares in Fig. 1) additionally allow us to determine J_{00} and the effective gap E_a of a solar cell [12].

Although Suns-V_{OC} measurements are valuable, the effect of sample heating has not been discussed thoroughly, despite using illumination intensities of up to 1000 Sun equivalents [13,14]. At high applied intensities, an absorption-induced self-heating of the device becomes likely, ultimately leading to a decreased V_{OC}. Indeed, a saturation of V_{OC} has frequently been observed at strong irradiation, but has been typically attributed to surface recombination rather than heating [15–17]. Moreover, voltage turnovers at increasing light intensities (see black circles in Fig. 1) have been observed for both inorganic [13,14,18] and organic solar cells [12]. Again, this effect is assigned to the contacts and can be modeled as a Schottky diode and parallel shunt resistor equivalent circuit, reducing the voltage at high intensities [13], or it can be an intrinsic feature of silicon heterojunction solar cells [19].

In this work, we provide an alternative simple analytical model describing these voltage turnover effects in Suns-V_{OC} measurements (black circles in Fig. 1) and show that such a behavior can often be solely explained by an electrothermal feedback effect. We validate this model for various solar-cell technologies including small-molecule organic solar cells, perovskite solar cells, and silicon solar cells. Furthermore, understanding the absorption-induced self-heating has also practical significance. The distinct shape of the curve allows us to determine the ideality factor, the relevant energy gap, and the temperature rise from a single measurement at ambient conditions. Thus, the presented adapted Suns-V_{OC} technique allows us to quickly compare different materials and structures regarding these basic parameters without requiring an external temperature control.

In order to demonstrate this method, we first perform a set of Suns-V_{OC} measurements where we continuously illuminate the device with a fixed intensity for a hold time of 30 s and subsequently record V_{OC}. The light intensity is measured with a photodiode and calibrated to Sun equivalents based on mismatch-corrected $J(V)$ characteristics (see Supplemental Material, part III [20]). The red circles in Fig. 2 correspond to these continuous Suns-V_{OC} measurements, which reproduce the typical voltage saturation and turnover for various solar-cell technologies [more details and $J(V)$ characteristics can be found in the Supplemental Material, part I [20]].

In order to explain this observation, we invoke self-heating effects and consider the following: A temperature change ΔT caused by absorption-induced self-heating is given by the product of the dissipating power and a thermal

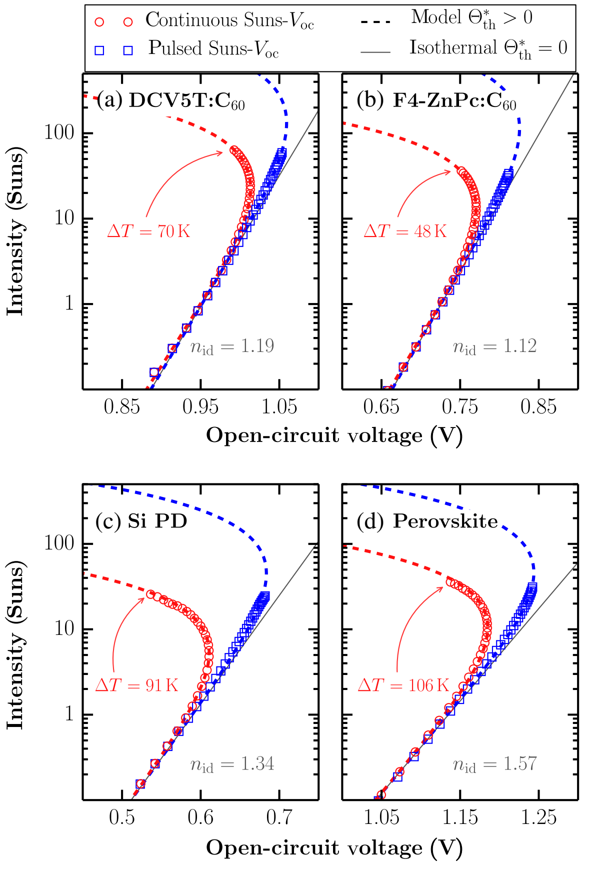

FIG. 2. Suns-V_{OC} measurements of (a) organic solar cell comprising a blend of DCV5T as donor and C_{60} as acceptor material; (b) organic solar cell with F4-ZnPc as donor and C_{60} as acceptor; (c) silicon photo diode; (d) perovskite solar cell. Red circles show a voltage turnover which can be explained by absorption-induced self-heating. For comparison, blue squares depict a measurement with a cooldown time between each data point with reduced electrothermal feedback. The latter lay closer to the isothermal curve.

resistance Θ_{th}. The thermal resistance depends on various parameters, such as the substrate geometry and the heat dissipation to the environment and can be time dependent [21,22]. By definition, there is no current flow at open-circuit conditions, which means that almost all absorbed energy is eventually converted into heat. It, therefore, can be described by the absorption A, the photon flux Φ of the light source, and the energy of the photons E_{ph}:

$$\Delta T = \Theta_{\mathrm{th}} \int A(\lambda)\Phi(\lambda)E_{\mathrm{ph}}(\lambda)d\lambda = \underbrace{\Theta_{\mathrm{th}}\tilde{A}}_{\Theta^*_{\mathrm{th}}} I_{\mathrm{Sun}}. \quad (2)$$

Typically, solar cells are characterized under standard reporting conditions, i.e., the photon flux is well defined by the AM1.5 spectrum [23], while the product of photon flux and energy describes the intensity I_{Sun} in units of Sun equivalents. For simplicity, it is convenient to introduce a reduction factor $\tilde{A}$ accounting for the fraction of incident energy that is absorbed, leading to an effective thermal resistance Θ^*_{th} as fitting parameter. Inserting Eq. (2) into the Shockley equation [Eq. (1)], assuming that the photogenerated current is proportional to the incident illumination intensity, i.e., $J_{\mathrm{ph}} \propto I_{\mathrm{Sun}}$, and solving it for V_{OC} yields

$$V_{\mathrm{OC}}(I_{\mathrm{Sun}}) = (T_{\mathrm{amb}} + I_{\mathrm{Sun}}\Theta^*_{\mathrm{th}})\frac{n_{id}k}{q}\ln\left(\frac{I_{\mathrm{Sun}}}{I_{00}}\right) + \frac{E_a}{q}. \quad (3)$$

Equation (3) takes into account self-heating and below we show that it fully describes the observed saturation and turnover effects. Note that Eq. (3) is valid only at open-circuit conditions and the extracted parameters cannot reproduce the full J-V characteristics with the ideal Shockley equation. However, they can be used in combination with more elaborated models, e.g., for transport-limited solar cells that include charge-carrier mobilities and recombination rates [9] or for drift-diffusion simulations. To fit the measurement data with Eq. (3), it is convenient to reduce the number of fitting parameters by substituting I_{00} in Eq. (3) with Eq. (1), taking a reference point on the experimental curve (V_{ref}, I_{ref}), where self-heating is negligible ($\Delta T < 0.5$ K).

$$V_{\mathrm{OC}}(I_{\mathrm{Sun}}) = (T_{\mathrm{amb}} + I_{\mathrm{Sun}}\Theta^*_{\mathrm{th}})\frac{n_{id}k}{q} \times \left[\ln\left(\frac{I_{\mathrm{Sun}}}{I_{\mathrm{ref}}}\right) + \frac{qV_{\mathrm{ref}} - E_a}{n_{\mathrm{id}}kT_{\mathrm{amb}}}\right] + \frac{E_a}{q}. \quad (4)$$

Using Eq. (4), we can describe the experimental data for all investigated solar-cell technologies. Table I summarizes the values of the fitting parameters E_a, n_{id}, and Θ^*_{th}. Both representative small-molecule organic solar cells, DCV5T:C_{60} [24,25] and F4-ZnPc:C_{60} [26], show ideality factors close to 1, comparable to the ones observed previously at solar intensities for a ZnPc:C_{60} blend [27]. For both organic solar cells, activation energies nicely correspond to values obtained from temperature-dependent Suns-V_{OC} measurements as described by Tvingstedt and Deibel [12] (see Fig. 1 for F4-ZnPc:C_{60}; for DCV5T: C_{60} see Supplemental Material, part III [20]). However, here we use an adapted technique, which requires no external temperature control and solely knowledge of the ambient temperature. Since both small-molecule devices have the same device architecture and substrate, the effective thermal resistances of both samples are indeed very similar, on the order of 1 K/Sun. Even below intensities of 10 Suns we see deviations from the isothermal curves, which already originate from a photoinduced sample heating. For the perovskite solar cell [28], we find an ideality factor of 1.57, a bit lower than reported elsewhere [29]. We determine an activation energy of 1.65 eV, which is close to the expected gap for this material system [30]. The effective thermal resistance is among the highest of the investigated devices, which can be explained by the very strong absorption of the perovskite [see Eq. (2)]. The same applies for the silicon photodiode, with an about threefold effective thermal resistance as compared to the organic representatives. We are able to fit the data with a band gap of 1.21 eV, which is slightly higher than the well-known band gap for crystalline Si of 1.12 eV [31]. However, a similar value of 1.23 eV has been determined from temperature-dependent voltage measurements [32]. For a solution-processed solar cell based on P3HT:PCBM, a material system with comparably low absorption, we barely see a voltage turnover, but a saturation of V_{OC} is present at 30 Suns (see Supplemental Material, part IV [20]). Based on our model, we can estimate temperature changes of up to 100 K for the highest intensities used in this work (at ≈35 Suns for the perovskite sample). The voltage turnover itself typically happens at smaller temperature rises ΔT in the range of 20 to 30 K. Overall, we are able to extract a single global ideality factor for this intensity range, despite changes in the slope of the Suns-V_{OC} measurements.

TABLE I. Fitting parameters of the experimental data shown in Fig. 2 according to Eq. (3). $\Theta^*_{\mathrm{th},c}$ and $\Theta^*_{\mathrm{th},p}$ correspond to the effective thermal resistance of the pulsed and continuous Suns-V_{OC} measurement, respectively.

Material	n_{id}	E_a (eV)	$\Theta^*_{\mathrm{th},c}$ (K/Sun)	$\Theta^*_{\mathrm{th},p}$ (K/Sun)
DCV5T:C_{60}	1.19	1.44	1.10	0.20
F4-ZnPc:C_{60}	1.12	1.31	1.33	0.18
Silicon	1.34	1.21	3.58	0.49
Perovskite	1.57	1.65	2.97	0.80

In order to further verify our model, we perform a second measurement where we minimize self-heating. Instead of keeping the intensity constant at each point for 30 s, we illuminate the devices with a rather short pulse of a few hundred milliseconds, followed by 30 s of darkness. These pulsed Suns-V_{OC} measurements are depicted as blue squares in Fig. 2. When pulsing the incident light, none of the investigated solar cells heats up sufficiently to show a voltage turnover. This indicates that neither the contact of the solar cell nor approaching the built-in voltage can deliver a full explanation of the voltage turnover. Still, our model is able to describe these pulsed Suns-V_{OC} measurements with a lower effective thermal resistance (Table I). It is also evident, that even in the pulsed measurement the data points deviate from the ideal isothermal case (gray solid line). Our automated measurement routine produces light pulses with a length of about 500 ms. In order to prove whether there still is a significant self-heating effect leading to deviations from the isothermal curve, we perform time-resolved measurements of the open-circuit voltage within the 35 s upon illumination of the F4-ZnPc:C_{60} and perovskite solar-cell sample for different fixed intensities (Fig. 3). For F4-ZnPc:C_{60} [Fig. 3(a)] we find voltage drops of a few millivolts already after a few 100 ms, a change sufficiently high to affect the pulsed (blue) measurement in Fig. 2. After 500 ms, the solar cells already lose a few millivolts as compared to the maximum values at high intensities. In contrast to F4-ZnPc:C_{60}, perovskite-based solar cells need much longer to reach an electronic steady state [33]. Any measurement with shortly applied pulses would characterize these cells in an electronic nonequilibrium (see Supplemental Material, part VII [20]), making a comparison between different structures and material compositions impossible. For this reason, the pulsed measurement is done with a 2-s pulse, which is sufficient to reach the electronic steady state. However, it makes the device more vulnerable to self-heating and thus explains the higher effective thermal resistance (Table I). This finding highlights the necessity of our model for material systems where long light pulses are needed to reach the electronic steady state. With our approach, one can directly benefit from a temperature change, thus, self-heating is not to be prevented. Depending on the measurement conditions, such as the light intensity and geometry and heat dissipation of the sample, it can be necessary to use time-resolved measurement techniques to investigate the solar cell close to isothermal conditions.

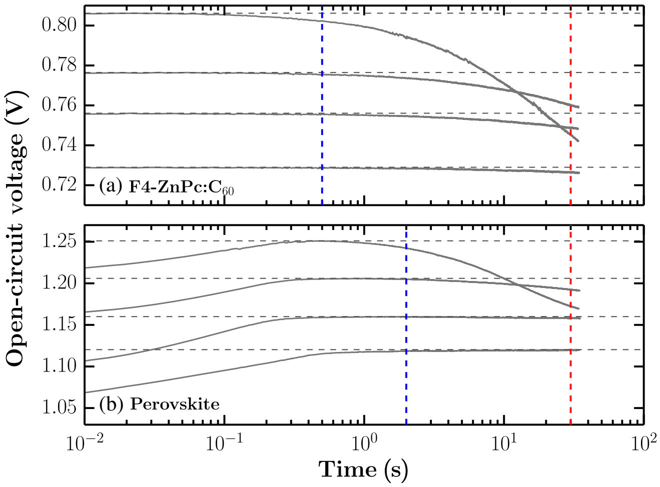

FIG. 3. Dependence of the open-circuit voltage as a function of time for various intensities for (a) the F4-ZnPc:C_{60} sample and (b) for the perovskite sample. Because of self-heating and a consequential increase of the cell temperature, V_{OC} decreases over time. The blue and red dashed lines mark the point in time at which V_{OC} is measured in the pulsed and continuous Suns-V_{OC} measurement, respectively.

In order to understand the physical origin of the voltage turnover at high light intensities in more detail, we show in Fig. 4 a plot of Eq. (3) for different effective thermal resistances. In the absence of absorption-induced self-heating, i.e., $\Theta^*_{\mathrm{th}} = 0$, the open-circuit voltage scales linearly with the logarithm of the illumination intensity as illustrated by dotted black lines in Fig. 4. Upon changing the ambient temperature T_{amb}, the slope will change but all curves will ultimately cross the (E_a, I_{00}). With increasing effective thermal resistance, the solar cell will heat up, reducing its open-circuit voltage, and eventually leading to a voltage turnover. Such an electrothermal feedback mechanism can also be found in thermistor devices as well as in organic light-emitting diodes, where the power dissipation increases faster than the heat can be conducted [34,35].

Taking the derivative of Eq. (3) gives an expression for the intensities of the turnover points TOP_1 and TOP_2:

$$\mathrm{TOP}_{1,2} = I_{00}\left[W_{0,-1}\left(-\frac{eT}{\Theta^*_{\mathrm{th}} I_{00}}\right) - 1\right], \qquad (5)$$

where W_0 and W_{-1} are the upper and lower branch of the Lambert W function [36], respectively, and e is Euler's number. For $W_{0,-1} = -1$, both turnover points coincide,

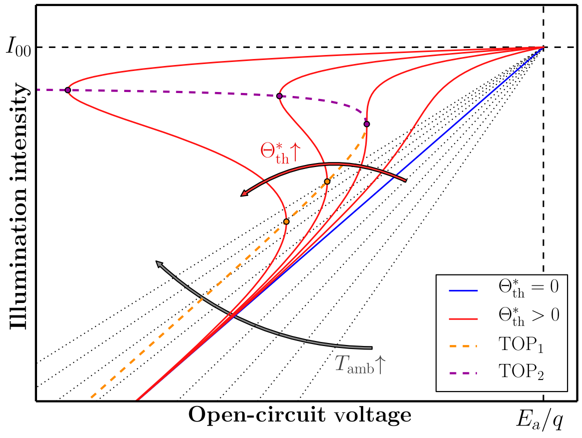

FIG. 4. Schematic illustration of the self-heating model. The black dotted lines show the isothermal behavior for different temperatures. All curves meet in the point $(I_{00}, E_a/q)$. Adding absorption-induced self-heating results in deviations from the isothermal curves (red lines), i.e., the voltage decreases with increasing self-heating coefficient Θ^*_{th}. The turnover points $\mathrm{TOP}_{1,2}$ (orange and purple dashed line) exists for $\Theta^*_{\mathrm{th}} > e^2 T_{\mathrm{amb}}/I_{00}$; their dependence on Θ^*_{th} and T_{amb} are shown in the Supplemental Material, part V [20].

which is the case when $\Theta^*_{\mathrm{th},0} = e^2 T / I_{00}$ (see Supplemental Material [20] for more details), while no turnover points are defined for thermal resistances smaller than $\Theta^*_{\mathrm{th},0}$. The intensity at which the first turnover point (TOP_1) occurs, decreases for increasing effective thermal resistance and lower ambient temperatures (see Supplemental Material [20] for more details). Therefore, the effect is even stronger when conducting Suns-V_{OC} measurements at low ambient temperatures, i.e., the turnover appears at lower light intensities. Indeed, we find data in the literature which support this trend [12–14]. The second turnover point (TOP_2) will always appear very close to I_{00}, thus making it very unlikely to reach this point experimentally (see Supplemental Material [20]).

In conclusion, we present a model that includes absorption-induced self-heating for the analysis of Suns-V_{OC} measurement. It explains the saturation of the open-circuit voltage with increasing light intensities for various photovoltaic technologies. The results reveal that commonly invoked causes such as surface recombination, the density of states, or the back contact are not necessarily needed. While it is common to control the temperature of solar cells to determine the performance according to standard testing conditions, we want to emphasize the importance to also consider self-heating in other measurement techniques. In particular, we show that for the perovskite device, longer illumination durations are required to reach a steady state which inevitably leads to self-heating. Additionally, our adapted Suns-V_{OC} measurements also allow us to determine the effective gap from a single curve at ambient temperature, which is of special interest for alternative technologies such as small-molecule organic, polymer, and perovskite solar cells.

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