Determination of Charge-Carrier Mobility in Disordered Thin-Film Solar Cells as a Function of Current Density

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Charge-carrier mobility is a fundamental material parameter, which plays an important role in determining solar-cell efficiency. The higher the mobility, the less time a charge carrier will spend in a device and the less likely it is that it will be lost to recombination. Despite the importance of this physical property, it is notoriously difficult to measure accurately in disordered thin-film solar cells under operating conditions. We, therefore, investigate a method previously proposed in the literature for the determination of mobility as a function of current density. The method is based on a simple analytical model that relates the mobility to carrier density and transport resistance. By revising the theoretical background of the method, we clearly demonstrate what type of mobility can be extracted (constant mobility or effective mobility of electrons and holes). We generalize the method to any combination of measurements that is able to determine the mean electron and hole carrier density, and the transport resistance at a given current density. We explore the robustness of the method by simulating typical organic solar-cell structures with a variety of physical properties, including unbalanced mobilities, unbalanced carrier densities, and for high or low carrier trapping rates. The simulations reveal that near $V_{OC}$ and $J_{SC}$, the method fails due to the limitation of determining the transport resistance. However, away from these regions (and, importantly, around the maximum power point), the method can accurately determine charge-carrier mobility. In the presence of strong carrier trapping, the method overestimates the effective mobility due to an underestimation of the carrier density.

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I. INTRODUCTION

Thin-film solar cells based on disordered materials such as organic carbon-based conducting molecules or amorphous inorganic semiconductors have great potential as a low-cost, low-carbon source of electricity. For such devices to be efficient, the photoabsorber must be able to efficiently transport charge to enable low resistive losses and reduced current loss via recombination. The figure of merit for charge transport is charge-carrier mobility. In traditional inorganic semiconductors, this mobility can be as high as $4 \times 10^5 \text{ cm}^2/(\text{Vs})$, while in disordered materials, it is typically in the range of $10^{-5}-10^{-4} \text{ cm}^2/(\text{Vs})$. A low mobility will reduce the fill factor (FF), short-circuit currents, and open-circuit voltage [1,2]. Unfortunately, even for nominally identical disordered materials, there exists a large variation of reported charge-carrier mobility values, with the reported values depending strongly upon the measurement technique employed [3,4]. Part of the reason for this variation is that carrier mobility is a strong function of carrier density in these disordered material systems. This is because at low carrier densities, proportionally more deep trap states will be occupied, and, thus, on average it will be energetically difficult for charge carriers to move. As the charge density is increased due to the Pauli exclusion principle, higher states will be filled where transport is energetically easier [5–11]. However, many measurement techniques inherently neglect this dependence. Furthermore, the measurement conditions (such as applied-bias voltage, light intensity, or device geometry) vary from method to method. This also means that charge-carrier density varies from method to method, making it difficult to compare mobility values between material systems. This creates the need for a reproducible measurement technique, which can consistently measure charge-carrier mobility as a function of charge-carrier density. Furthermore, it should be noted that for mobility to be a useful figure of merit for a light-harvesting device, values should be compared at the maximum power point under steady-state operating conditions; hence, there is a need to be able to reliably evaluate mobility as a function of current. Finally, before going further, it should be noted that measuring mobility in disordered semiconductors is a difficult problem to solve due to the charge-carrier mobility being a function of carrier density and the fact

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that often in disordered materials, any change in device structure (even a change in bottom contact) can change the morphology or electrical properties of the subsequently deposited layers. Thus, it is difficult to use techniques from inorganic semiconductor physics, such as Hall probe measurements, which require special device structures to be fabricated. Consequently, experimentalists have had to come up with ingenious methods to tease mobility out of working devices.

The most common methods to determine mobility are steady-state space-charge-limited current (SCLC) [12], time of flight (TOF) [13], dark-injection transient current (DITC) [14], and charge extraction by linearly increasing voltage (CELIV) [15]. TOF, DITC, and CELIV assume a mobility independent of carrier density and suffer from several drawbacks: For TOF, the dielectric relaxation time must be larger than the transit time. Moreover, the experiment has to be performed at high negative-bias voltage, meaning the background charge density will be lower than is found in an operating device. Finally, photogenerated charges have to be produced close to one electrode. Therefore, the device must be optically thick (around 1 μm), and as material microstructure is often a function of thickness, this makes it difficult to measure functioning solar cells which are typically only 100 nm thin [15]. For the DITC method, pulse parameters such as pulse on and off times have the potential to significantly affect the value of measured mobility due to charge trapping and detrapping effects [16,17]. Furthermore, the models commonly used to interpret the results do not take these trapping effects into account. For CELIV, simulations of organic solar cells have shown that the technique is only able to determine an apparent mobility, which can be up to 2 orders of magnitude different from the actual mobility of the faster carriers [18]. The reason is rooted in the basic assumptions made for the theory behind CELIV. Most important, it requires that the carrier distribution inside the photoactive layer is given by a uniform sheet of charge carriers, which is neatly swept out of the device by the applied voltage. However, due to disorder, most charges in disordered materials reside in trap states. Hence, the extraction of charge carriers is altered by trapping and detrapping of charge carriers. As a result, the shape of the CELIV curve is affected by the density and distribution of these trap states and the average carrier mobility changes during the CELIV measurement.

Unlike the above techniques, with SCLC, the carrier dependence of mobilities can be determined [5,6,8]. However, the great disadvantage of SCLC is that a hole-only or electron-only diode structure is required. Very often, in thin-film devices the electrical properties of the layer will depend on how the material is deposited (or even on which layer it is being deposited), and even nominally identical devices can have differing mobilities. Furthermore, many organic solar cells rely on a bulk heterojunction absorber with a closely intermixed network of donor and acceptor components. In these devices, charge transport is by definition ambipolar [4]. Additionally, the built-in voltage and dielectric constant must be known to apply this technique, the former being especially difficult to measure in disordered devices [19]. Finally, another drawback of SCLC is the difficulty of distinguishing between injection-limited and space-charge-limited currents.

In 2010, Shuttle et al. [7] proposed a method to determine the mobility as a function of carrier density. It is based on the charge-extraction (CE) measurement [20] in which the solar cell is first held at short circuit under illumination. The light source is then switched off and the device simultaneously short circuited. Provided that recombination losses are negligible, the integral of the current leaving the device is a measure of the excess charge in the cell. By varying the bias-light intensity, one can explore a range of carrier densities, and with the aid of an analytical model, carrier-density-dependent mobility can be extracted. Even though the method is attractive because of its simplicity, it suffers from several drawbacks: the mobility is accessible only at short-circuit condition, the analytical model assumes that transport is driven only by one type of carrier, it requires knowledge of the built-in potential, and it does not account for diffusion.

Albrecht et al. [21] extended Shuttle’s method so that any forward-bias condition could be used. Errors due to recombination between the steady-state illumination and the transient charge extraction were minimized by rapidly applying a high reverse bias after switching off the light. The method is based on the following relationship among mobility, carrier density, and transport resistance:

$$\mu_{\text{eff}} = \frac{d}{2q\bar{n}R_{\text{tr}}}.$$  

Here, $\mu_{\text{eff}}$ is an effective mobility of electrons and holes, $J$ the current density, $d$ the device thickness, $q$ the elemental charge, and $\bar{n}$ the average carrier density extracted from the measurement. $R_{\text{tr}}$ is the resistance caused by the transport of electron and holes through the device. In the technique by Albrecht et al., charge-extraction measurements are performed at two different measurement conditions. Under the first condition, a forward bias is applied to the device under a given light intensity before performing charge extraction. Under the second, the device is held at open circuit before performing charge extraction. For this condition, the light intensity is varied until the extracted carrier density matches the one of the first condition. From the voltage differences of these two measurements, $R_{\text{tr}}$ is estimated. Together with $\bar{n}$, this allows the determination of the mobility as a function of current density. Albeit, the method requires about twice as many measurement steps as the one by Shuttle et al., but it is not hampered by the assumptions made therein. Hence, neither the built-in voltage has to be known nor has one to assume that transport is dominated by one type of carrier or only by drift.
However, the underlying analytical model proposed by Albrecht is afflicted by several assumptions, which limit the general applicability of the method. First, in Eq. (1) it is assumed that the electron and hole current densities are equal throughout the device. However, this assumption is generally not true. Second, the theoretical \( \tilde{n} \) in their formula is equal to the geometrical mean of the electron and hole densities, while the measurement extracts the arithmetic mean. This leads to an underestimation of the effective mobility. Next, it neglects the voltage drop across the outer circuitry. Finally, Eq. (1) is only valid for the simplification \( \tilde{n}_e(h) = n_e(h) = \text{const} \). This assumes that the carrier density across the device is uniform, when in fact, in many cases it exponentially decays away from the contacts.

Equation (1) was also independently established by Schiefer et al. using another approach [22]. They assumed a constant and equal mobility for electrons and holes, which they determined via CELIV. \( R_c \) was extracted from a combination of a light \( J-V \) curve and Suns-V\(_{OC} \) curve. With the help of Eq. (1), they were then able to determine the average carrier density. The approach by Schiefer et al. extracts the transport resistance correctly, but it assumes that the average electron and hole current densities are equal and contribute equally to the total current. Furthermore, their approach is limited to the case \( \tilde{\mu}_p = \tilde{\mu}_e = \text{const} \), \( \tilde{n}_h = \tilde{n}_e = \tilde{n} \); however, mobility is known to be a strong function of carrier density, and as we state above, the carrier density decreases exponentially as the distance from the contact is increased.

In this work, we present a simulation study that critically investigates the work of Albrecht et al. and Schiefer et al. Revising the derivation of Eq. (1), we demonstrate that the effective mobility is, in general, a function of the spatially dependent carrier densities. The method is generalized to any combination of measurements that is able to determine the arithmetic mean of the average carrier density of electron and holes, and the transport resistance at a given current density. Drift-diffusion simulations of typical organic solar-cell structures are used to critically assess the advantages and limitations of the method.

This paper is organized as follows: First, we investigate reliability of measuring charge density using the transient photovoltage (TPV) and transient photocurrent (TPC). Then, for a combination of \( J-V \) and transient measurements, we revisit and generalize the theory behind Eq. (1) for the extraction of the mobility. Thereafter, we examine the conditions under which Eq. (1) is valid and where it is not.

II. MEASURING CHARGE-CARRIER DENSITY

For the measurement of the charge-carrier density, two widely employed techniques are equally appropriate: charge extraction as presented by Albrecht et al. or a combination of TPV and TPC measurements [23]. Charge extraction has the advantage that only one measurement has to be performed at each current density. However, a general drawback of charge extraction is the practical aspect of turning off a 1-sun light source and shorting a steady-state solar cell quickly enough as to not influence the measured current transient. Furthermore, the more quickly the cell and light source are switched, the more pronounced inductive ringing and rf effects become in the charge-extraction transients. By using a method called differential charging [23], where TPV and TPC are used together to determine charge density, one needs two measurements at each current density. However, the method is a small-signal method, so one avoids the parasitic effects associated with large-signal measurements. Furthermore, TPV will also provide carrier-lifetime information at the same time, which can be used to calculate diffusion lengths, another important figure of merit. For the above reasons, for this work we choose the combination of TPV and TPC.

The use of transient voltage and current techniques to measure the carrier lifetime of minority charge carriers in \( p-n \) junctions has a long history [24]. They are generally called open-circuit voltage decay (OCVD) and short-circuit current decay (SCCD). When applied to dye-sensitized solar cells (DSSCs), the techniques were more recently dubbed transient photovoltage and transient photocurrent, respectively. These terms now prevail throughout the DSSC, organic photovoltaic, and other novel thin-film solar-cell literature. TPV is mainly used to determine the carrier lifetime [25–27], while TPC is chiefly applied to study trapping and detrapping effects [28–30].

In this work, we make use of both the transient voltage and current techniques, where a steady-state bias light is used in conjunction with a small perturbation light source [23,30]. The steady-state bias light is a white-light source, while the small excitation light source may be a laser or a single-wavelength LED. Its wavelength range should be selected in a way so that the generation profile is as uniform as possible in the device. For this investigation, we opt for a laser of wavelength 370 nm with a short pulse width of 5 ns. In the following, we denote quantities related to the small perturbation pulse, for example, a small change in carrier density, with the term “excess” or “\( \Delta \)” This is to distinguish these quantities from their steady-state counterparts.

Figure 1 illustrates a typical simulation of TPV and TPC experiments and the information extracted from them. The upper left panel depicts a TPV experiment, while the lower left panel depicts a TPC experiment. The illumination induces the generation of charge carriers in the device, which is plotted as a red line in Figs. 1(a) and 1(b). At the start of the experiment, the device is illuminated with a steady-state bias light until an equilibrium between generation, recombination, and in the case of TPC, extraction is reached. Thereafter, an additional small light pulse is applied to the device, which is discernible by the vertical red line in Figs. 1(a) and 1(b). When the device is held under open circuit, the bias light produces a bias voltage...
of the small excitation carriers [23,31]. The data are usually
its decay time can be interpreted as the carrier lifetime
voltage transient takes the form of a single exponential
bias-light intensity. For TPV, this is to ensure that the
attenuated so that the perturbation is small compared to the
majority of the excess charge carriers are extracted out of
trapping. In the case of TPC, the current decays since the
small light source, the voltage and current density decay
Δ
charge carriers, which is manifested by an excess voltage
established. The small perturbation light generates excess
signal excitation has been switched off. (b) TPC simulation: The red line shows the same generation rate as in (a). The green line exhibits the
TPC response, which is also made up of the steady-state bias-current density
δ
bias; under short-circuit conditions, a bias current
V
bias or excess current density
J
bias can be plotted against the
bias-intensity curve in the dark. It is also known as Suns-
V
OC curve, which is needed for the determination of the mobility. The
J
bias-intensity curve, in turn, reveals a linear dependence between light intensity and bias-current density.

By varying the intensity of the steady-state light source
I
light, the bias-light-dependent parameters can be determined. When
I
light is independently measured by a calibrated photodetector, V
bias or
J
bias can be plotted against the intensity. This is illustrated in Fig. 1(c) for a simulation of bias-light-dependent TPV and TPC measurements. The
V
bias-intensity curve shows the typical shape of a diode’s
J-V curve in the dark. It is also known as Suns-V
OC curve
[32], and we later use it to construct a
J
SC-V
OC curve, which is needed for the determination of the mobility. The
J
bias-intensity curve, in turn, reveals a linear dependence between light intensity and bias-current density.

By a combination of TPV and TPC measurements, the excess carrier density can be deduced employing a method known as differential charging [23]. During a current transient of a TPC measurement, an excess charge
Δ
Q or average excess carrier density
Δ
n
TPC is extracted from

FIG. 1. Illustration of typical TPV and TPC simulations and the parameters extracted from them. (a) TPV simulation: The red line depicts the generation rate consisting of a steady-state generation and a small-signal pulse. The blue line is the photovoltage response, which is composed of the steady-state bias voltage
V
bias, a small excitation voltage
Δ
V, and the monoeXponential decay after the small-signal excitation has been switched off. (b) TPC simulation: The red line shows the same generation rate as in (a). The green line exhibits the TPC response, which is also made up of the steady-state bias-current density
J
bias, a small-signal current density
Δ
J, and an exponential decay after switching off the small-signal excitation. The light blue area under the current transient is the excess charge
Δ
Q extracted during the small-signal perturbation. (c) 
V
bias and
J
bias as a function of bias-light intensity
I
light. They are extracted from bias-light-dependent TPV and TPC simulations, respectively. (d) Excess charge
Δ
Q and differential capacitance
C =
Δ
Q/
Δ
V as a function of bias voltage. 
Δ
Q is determined from a bias-light-dependent TPC simulation; see (b). 
Δ
V is computed from an adjoined TPV simulation; see (a). The light orange area under the capacitance-voltage curve is proportional to the total photoinduced carrier density at the upper limit of the integration
V.
the device. In a first approximation, it can be obtained by integrating the area under the short-circuit current transient [shown by the light blue area in Fig. 1(b)],

\[
\Delta Q = AdqA\hat{n}_{TPC} \approx A \int_{t_0}^{t_f} J d t, \tag{2}
\]

where \(A\) is the device area, \(d\) the device thickness, \(q\) the elemental charge, and \(t_0\) and \(t_f\) are the start and finish of the current transient, respectively [Fig. 1(b)]. In a TPV measurement performed under the same illumination conditions, the small-signal perturbation induces a photovoltage peak \(\Delta V\) [Fig. 1(a)]. We can, therefore, define a capacitance for the device at any given measurement condition as

\[
C = \frac{\Delta Q}{\Delta V}; \quad \Delta Q \text{ and } C \text{ are depicted in Fig. 1(d) as a function of bias voltage. It can be seen that both } \Delta Q \text{ and } C \text{ exhibit a strong dependence upon } V_{bias}. \text{ Now, differential charging assumes that, starting from dark conditions, the average excess carrier density can be calculated by summing } CdV \text{ while increasing the light bias in small steps. } dV \text{ signifies here the increase of } V_{bias} \text{ between successive measurement steps. Integrating } C \text{ with respect to the voltage gives the expression for the average carrier density } \bar{n}_{TPV,TPC} \text{ as a function of voltage:}
\]

\[
\bar{n}_{TPV,TPC}(V) = \frac{1}{Aqd} \int_{0}^{V} C(V') dV'. \tag{3}
\]

This integration is depicted in Fig. 1(d) by the light orange area. It is clear from this derivation that \(\bar{n}_{TPV,TPC}\) is only composed of photoinduced charge carriers and does not include the dark carrier density. When the bias-light intensity is varied, the dependence of the carrier lifetime on excess carrier density can be determined [23]. From this dependence, other recombination parameters such as the recombination rate, coefficient, or order can be revealed. Here, we use bias-light-dependent TPV and TPC measurements as a means of determining the carrier density \(\bar{n}_{TPV,TPC}\), and from there, the mobility, as we explain in the next section.

The determination of \(\Delta \bar{n}_{TPC}\) is only valid under certain conditions. A theoretical derivation reveals the following relationship between the integrated area under the current transient and the charge-carrier dynamics [33]:

\[
\frac{1}{Aqd} \int_{t_0}^{t_f} \Delta J_{SC} d t = \frac{1}{2} \left( \Delta \bar{n}_{e,\text{init}} + \Delta \bar{n}_{h,\text{init}} \right) - \frac{\Delta \sigma_{\text{EL}}}{2qd} - \int_{t_0}^{t_f} \left( \frac{\Delta J_{\text{surf}}}{qd} + \Delta R \right) d t. \tag{4}
\]

Here, \(\Delta \bar{n}_{e(h),\text{init}} = \Delta \bar{n}_{e(h)}|_{t_0} - \Delta \bar{n}_{e(h)}|_{t_f}\) denotes the average excess carrier density, which is initially generated due to the small-signal pulse. \(\Delta \sigma_{\text{EL}}\) is defined as the change in the charge-extraction method.

\[
\Delta \sigma_{\text{EL}} = (\sigma_d - \sigma_0)|_{t_0} - (\sigma_d - \sigma_0)|_{t_f}, \tag{5}
\]

where \(\sigma_d\) and \(\sigma_0\) are the charge densities (in cm\(^2\)) on the left and right metal electrodes, respectively. \(\Delta J_{\text{surf}} = \Delta J_e(0) + \Delta J_h(d)\) is the total excess surface recombination current density due to net electron extraction at the hole-selective contact \(\Delta J_h(0)\) and net hole extraction at the electron-selective contact \(\Delta J_e(d)\). \(\Delta R\) is the total excess recombination rate due to trapping and Shockley-Read-Hall recombination [33]. With device simulations, we analyze the contribution of each component of the right-hand side of Eq. (4) to the integral under the current transient. For the later discussion, we, therefore, express Eq. (4) in terms of excess carrier densities:

\[
\Delta n_{TPC} = \Delta n_{\text{init}} - \Delta n_{\text{EL}} - \Delta n_{\text{surf}} - \Delta n_{R}. \tag{6}
\]

where we combine the initially generated excess carrier density of electrons and holes in the term \(\Delta n_{\text{init}}\). \(\Delta n_{\text{EL}}\) corresponds to the change in the carrier densities on the contacts. \(\Delta n_{\text{surf}}\) and \(\Delta n_{R}\) stand for the excess carrier densities lost due to surface and bulk recombination, respectively. Thus, \(\Delta n_{TPC}\) is equal to \(\Delta n_{\text{init}}\) if surface and bulk recombination, and the change in charge on the contacts can be ignored. We like to stress the fact that \(\Delta n_{TPC}\) is then equal to half the sum of the initial electron and hole carrier density. This result is derived for any kind of current transient [33] and is, thus, also applicable to the charge-extraction method.

**III. DETERMINATION OF THE MOBILITY**

The concept of mobility in disordered materials is less straightforward than in crystalline semiconductors. Crystalline semiconductors possess a highly ordered regular arrangement of atoms, and Bloch’s theory tells us that this will produce well-defined transport bands with a forbidden region separating them (the band gap). Disordered materials, on the other hand, have no long-range order, which results in a quasicontinuous distribution of transport states with no well-defined bands. In these materials, one can distinguish only between extended and localized states. The dense bands of delocalized states are located above and below the band gap. An exponential or Gaussian tail of localized trap states extends from the delocalized bands into the band gap. The energy that separates the extended states from the localized ones is called the mobility edge. The charge-carrier mobility in the extended states above the mobility edge is much higher than that in the localized states below the mobility edge. At low carrier densities or low temperatures, all carriers reside in deep traps, meaning that the mobility will be very low. However, when the temperature is high or there are many charge carriers, the higher energetic states will be filled, and carriers will be able to move. A common approach is to
express mobility, which is dependent upon the filling of trap states in terms of the ratio of the carrier density located in extended states versus the total carrier density [18]:

$$\bar{\mu}_c = \frac{1}{d} \int_0^d \mu_c(x) \frac{n_{e(h)}^\text{free}(x)}{n_{e(h)}^\text{free}(x) + n_{e(h)}^\text{trapped}(x)} \, dx,$$

(7)

where \(\mu^0_c\) is the mobility of free electrons or holes traveling via extended states beyond the mobility edge, \(n_{e(h)}^\text{free}(x)\) is the density of free carriers, \(n_{e(h)}^\text{trapped}(x)\) is the density of trapped carriers in localized states within the exponential band tail, and \(d\) is the device thickness. Hence, if no carriers are trapped within the band gap, the mobility is constant and equal to the average mobility of carriers moving through the extended states. Otherwise, the presence of the term \(n_{e(h)}^\text{trapped}(x)\) will impinge a carrier dependence on the mobility \(\mu_c\), as trapping and detrapping are strongly affected by a change of operation conditions. We intend to determine exactly this carrier-dependent mobility. It is important to note that in measurements, only the spatially averaged mobility is accessible.

For this purpose, we revisit and generalize the methods of Schiefer et al. [22] and Albrecht et al. [21]. The core of their methods is Eq. (1), which has been independently established by the two research groups using different approaches. Both have in common that constant and equal gradients of the quasi-Fermi-levels of electrons and holes were assumed. We follow mainly the approach of Schiefer et al. [22], while the one by Albrecht et al. [21] is discussed in the Supplemental Material [34].

We assume a solar cell with active layer thickness \(d\) and selective Ohmic contacts. Figure 2 depicts a schematic view of the energy diagram of a solar cell under illumination and an applied forward voltage \(V\). The majority hole contact is located at \(x = 0\), and the majority electron contact at \(x = d\). The transport of charges through the photoactive layer brings about a voltage drop. At a location \(x\) in the device, it is composed of the voltage drop \(\Delta V_{tr}\) due to hole transport from \(x\) to the hole contact and \(\Delta V_{te}\) due to electron transport from \(x\) to the electron contact. These voltage drops can be described in terms of the gradient of the quasi-Fermi-levels of electrons and holes \(\nabla F_{e(h)}\), the driving force for their transport:

$$\Delta V_{tr}(x) = \int_0^x \nabla F_{tr}(x') \, dx' + \int_x^d \nabla F_{tr}(x') \, dx'. \quad (8)$$

The assumption of constant and equal gradients of the quasi-Fermi-levels \(\nabla F_{e(h)} = \nabla F_{tr}\) leads to

$$\nabla F_{tr}(x) \int_0^x \, dx' + \nabla F_{tr}(x) \int_x^d \, dx' = \nabla F \, d = \text{const} = V.$$ \quad (9)

As can be deduced from Fig. 2, the constant gradient of the Fermi level times the thickness \(d\) equals the total voltage drop \(V\). \(\nabla F_{e(h)}\) can be expressed in terms of the current, the mobility, and carrier density as

$$\nabla F_{e(h)} = \frac{J_{e(h)}}{\sigma_{e(h)}}. \quad (10)$$

For any \(x\) inside the photoactive layer, the total current \(J\) is the sum of the electron and hole current densities:

$$J = J_e(x) + J_h(x) = q(\mu_e(x)n_e(x) + \mu_h(x)n_h(x))\nabla F.$$ \quad (11)

Combining Eqs. (9) and (11), we obtain

$$\mu_e(x)n_e(x) + \mu_h(x)n_h(x) = \frac{Jd}{qV}.$$ \quad (12)

The transport voltage \(V\) can be associated to a transport resistance via \(V = JR\). This leads us to the following general relationship:

$$\mu_e(x)n_e(x) + \mu_h(x)n_h(x) = \frac{d}{qR}.$$ \quad (13)

To get from here to Eq. (1), Schiefer et al. and Albrecht et al. made further simplifications already mentioned in the Introduction. Schiefer et al. [22] made two more assumptions: (1) the spatially averaged electron and hole current densities are equal and half the total current density \(J_e = J_h = J/2\), and (2) the electron and hole mobilities are equal and constant \(\mu_h = \mu_e = \text{const}\). Both assumptions
lead to the following simplification for the spatially averaged carrier density: \( n_{\bar{h}} = n_{\bar{e}} = \bar{n} \). In Fig. S2 in the Supplemental Material [34], we demonstrate that all three currents in assumption (1) agree only at \( V_{OC} \) if both the carrier densities and mobilities are fairly balanced. Assumption (2) is a strong limitation for the method and, in general, rarely the case in disordered solar cells [35].

Albrecht et al. [21], in turn, made three further assumptions: (1) the electron and hole current densities are equal everywhere in the photoactive layer: \( J_{e}(x) = J_{h}(x) \) so that \( \mu_{e}(x) n_{e}(x) = \mu_{h}(x) n_{h}(x) \), (2) the extracted charge from the integral of the CE transient is half the sum of the electrons and holes, and (3) the electron and hole carrier densities are constant throughout the device so that \( \bar{n}_{e(h)} = n_{e(h)} \). These assumptions result in an effective mobility \( \mu_{eff} = (\mu_{e} + \mu_{h})/(\mu_{e} + \mu_{h}) \). The first assumption is generally not valid: As soon as a current flows in forward direction, the electron current density increases steeply from \( J_{e}(0) \approx 0 \) at the hole contact to \( J_{e}(d) \approx J \) at the electron contact (see Fig. S2 in the Supplemental Material [34]). The opposite is true for the hole current density, which means that assumption (1) is wrong for \( J > 0 \). At \( V_{OC} \), we have by definition \( J = 0 \) so that \( J_{e}(x) = -J_{h}(x) \). As shown in Eq. (4), assumption (2) is valid as long as recombination and the difference between surface charges on the contacts can be neglected. Finally, assumption (3) puts a strong limitation on the method. Würfel et al. [1] established a different relationship using the geometrical mean of the electron and hole carrier density. However, it was also based on the additional assumption that \( J_{e}(x) = J_{h}(x) \) everywhere in the bulk.

If the total carrier density \( \bar{n} \) is measured in experiment, we show in the Supplemental Material [34] that Eq. (13) can be transformed without any further presumptions into

\[
\mu_{eff}(J) = \left( \frac{1}{\mu} + \frac{\Delta \mu}{\bar{n}} \right)^{-1} = \frac{d}{2q\bar{n}(J)R_{U}(J)}.
\]

\( \mu \) is the arithmetic mean of the electron and hole mobilities: \( \mu(x) = (\mu_{e}(x) + \mu_{h}(x))/2 \), and \( \Delta \mu \) is the difference between the mobilities and the arithmetic mean: \( \mu_{e(h)}(x) = \mu(x) \pm \Delta \mu(x) \). The same holds for \( \bar{n} \) and \( \Delta \bar{n} \); the bars denote spatial averages. The plus sign in Eq. (14) corresponds to the case where both the mobility and carrier density of one carrier type are higher than their counterparts [e.g., \( \mu_{e}(x) > \mu_{h}(x) \) and \( n_{e}(x) > n_{h}(x) \)]. The minus sign stands for the case where the mobility and carrier density of the same carrier type show opposite behavior [e.g., \( \mu_{e}(x) > \mu_{h}(x) \) and \( n_{e}(x) < n_{h}(x) \)]. Note that, in general, one cannot assume that because the mobility of one carrier type is higher than the other that its carrier density must be also higher, as this also depends on the density of trap states.

We already indicate above that the TPV and TPC measure the photogenerated carrier density \( \bar{n}_{ph} \) instead of \( \bar{n} \). Instead of expressing carrier densities by their mean and differences, we divide them into an equilibrium carrier density in the dark \( n_{0e(h)} \) and photogenerated carrier density \( n_{ph,e(h)}(x) = n_{0e(h)}(x) + n_{ph,e(h)}(x) \). The relationship between \( \bar{n}_{ph} \) and \( n_{ph,e(h)}(x) \) is given by \( \bar{n}_{ph} = \frac{1}{2}(n_{ph,e} + n_{ph,h}) \). Then, we can find a similar definition of \( \mu_{eff} \) (see the Supplemental Material [34]):

\[
\mu_{eff}(J) = \left( \frac{1}{\mu} + \frac{\Delta \mu(n_{e} - n_{h})}{(n_{ph,e} + n_{ph,h})} \right)^{-1} = \frac{d}{q\bar{n}_{ph}(J)R_{U}(J)}.
\]

We find that in most cases the second term under the overbar in Eq. (15) can be neglected so that \( \mu_{eff}(J) = \left( \frac{1}{\mu} \right)^{-1} \).

IV. DETERMINATION OF THE TRANSPORT RESISTANCE

The key to using Eqs. (14) and (15) is also being able to determine the transport resistance \( R_{tr} \), which is caused by the transport of electrons and holes through the device. For this purpose, any method that is able to measure the series resistance as a function of voltage or current is suitable. This is because the series resistance is composed of \( R_{tr} \) and the circuit resistance \( R_{circ} \). Prominent ways to calculate the series resistance are the comparison of the dark and light \( J-V \) curves [36], the comparison of two \( J-V \) curves at different light intensities [37], or the combination of the \( J-V \) measurement under illumination with a Suns-\( V_{OC} \) or \( J_{SC}-V_{OC} \) curve [38]. The latter two curves are pseudo-\( J-V \) curves, which are equivalent to a standard \( J-V \) curve for a solar cell that has no series resistance. In the approach by Schiefer et al. [22], a \( J_{SC}-V_{OC} \) curve was taken for the simulation, while the Suns-\( V_{OC} \) method was used in the measurement.

Here, we compile a \( J_{SC}-V_{OC} \) curve from the steady-state region of the TPV and TPC measurements at different bias-light intensities \( I_{light} \). \( V_{OC} \) corresponds to the bias voltage \( V_{bias} \) [see Figs. 1(a) and 1(c)]. \( J_{SC} \) is obtained using the superposition of the short-circuit current density at 1 sun, \( J_{SC|1sun} \), and the current-density curve proportional to \( I_{light} \), \( -J_{SC|1sun} I_{light} \) [32,39]:

\[
J_{SC}(V_{OC}) = J_{SC|1sun}(1 - I_{light}(V_{OC})).
\]

\( J_{SC|1sun} \) is determined from the light \( J-V \) curve, while \( I_{light} \) should ideally be measured independently during the TPV and TPC measurements. Alternatively, it may also be computed from the ratio of the bias-current density \( J_{bias} \)
FIG. 3. Simulated J-V and J_{sc-oc} curves obtained from the steady-state part of the TPC and TPV simulations. The dotted lines indicate how the series resistance is computed from the difference of both curves.

and J_{SC|sun}. However, this is only valid if the relationship between J_{bias} and light intensity is linear, as is the case for the simulation shown in Fig. 1(c). Instead, the relationship is generally given by a power law J_{bias} \propto P_{light}^{\alpha}, where the exponent \alpha has been found to vary between 0.7 and 1 depending on the photoactive material [39,40].

Figure 3 depicts the comparison of simulated J-V and J_{SC-oc} curves of a typical organic solar cell. The cell characteristics are described in the Simulation section. The J-V curve is affected by a significant amount of trapping, which manifests itself in a high series resistance (due to the relationship between carrier density and mobility) and, thus, low fill factor. The J_{SC-VOC} curve, on the contrary, is impervious to this effect. As indicated by Fig. 3, the series resistance at current density J is given by the difference between the corresponding voltages of the two curves divided by J:

R_s(J) = \frac{V_{J_{SC-oc}}(J) - V_{J-V}(J)}{J} = R_{tr} + R_{circ}. \quad (17)

In order to determine R_s of a solar cell, the current at the maximum power point is chosen [38]. For our purposes, we extend the method to all current densities except J = 0 and J = J_{SC}. At the latter current, Eq. (17) suggests zero series resistance, which, of course, is not true, as at this operation point the highest current is extracted from the device. R_{circ} can be estimated from R_s at high current in forward direction, where transport resistance is minimal [22].

Even though not stated explicitly, the approach of Albrecht et al. [21] also relies on extracting the series resistance from a combination of the J-V and Suns-V_{OC} curves. The charge extraction under the first measurement condition, where a forward-bias voltage is applied under illumination, corresponds to the J-V measurement. The charge extraction under the second condition, where the device is held at open circuit at a lower light intensity, corresponds to the Suns-V_{OC} measurement in this work. However, instead of subtracting two voltage points at the same current as we do, they compared two voltage points at the same measured carrier density. This is generally not valid and is discussed in the Supplemental Material [34].

V. SIMULATION

To investigate the accuracy of the investigated method, we use an open-source Shockley-Read-Hall-based [41] drift-diffusion-device model, the general-purpose photovoltaic device model (gpvdm) [30,42–45], which is adapted for this work to simulate the steady-state J-V curve, and transient photovoltage and photocurrent. The model can be downloaded from Ref. [45], along with example simulations from this paper. We then use the above method to extract the mobility from the simulated results just as one would do from experimental results. The advantage of this approach is that it enables us to compare the mobility one obtains from our proposed method to the mobility value present within the device model. The device model solves Poisson’s equation between the front and back contact of the device to account for electrostatic effects,

\frac{d}{dx} \epsilon_0 \epsilon_r \frac{d}{dx} \varphi = q(n_f + n_i - p_f - p_i), \quad (18)

where \epsilon_0 is the permittivity of free space, \epsilon_r is the relative permittivity of the photoactive layer, \varphi is the electrostatic potential, and q is the elementary charge on an electron. n_f (n_i) and p_f (p_i) are the densities of free (trapped) electrons and free (trapped) holes respectively. To describe charge transport in the device, the drift-diffusion equations are solved for electrons

J_n = q \mu_n n_f \frac{\partial E_{\text{LUMO}}}{\partial x} + q D_n \frac{\partial n_f}{\partial x}, \quad (19)

and for holes

J_p = q \mu_p p_f \frac{\partial E_{\text{HOMO}}}{\partial x} - q D_p \frac{\partial p_f}{\partial x}. \quad (20)

where J_n and J_p are the current densities of electrons and holes, \mu_n and \mu_p are the electron and hole mobilities, D_n, D_p are diffusion coefficients, and E_{\text{LUMO}} and E_{\text{HOMO}} represent the free-carrier mobility edges. For both electrons and holes, we assume an exponential distribution of trap states of form

\rho(E) = N \exp(E/E_a), \quad (21)

where \rho represents the energetic distribution of trap states, N is the maximum density of trap states, E is
the energy, and $E_a$ is the characteristic tail slope energy representing the degree of energetic disorder in the material system. Under each mesh point in position space, the distribution of trap states in energy space (for both electrons and holes) is divided into ten regions of width $\Delta E$. For each of these regions, the Shockley-Read-Hall capture or escape

$$\frac{dn_i}{dt} = r_{EC} - r_{EE} - r_{HC} + r_{HE} \quad (22)$$

is solved, where $n_i$ is the electron trap density, $r_{EC}$ is the rate of electron escape into a trap, $r_{EE}$ is the rate of electron escape from a trap, $r_{HC}$ is the rate of hole capture into an electron trap, and $r_{HE}$ is the rate of hole escape from an electron trap. Light propagation is modeled using the transfer-matrix method; for computational efficiency, the equations are projected onto a 1D finite-difference grid [46]. Some further details are given in the Supplemental Material [34].

The material parameters are taken from Ref. [30], which are obtained via calibration of the model to multiple data sets including TPV, TPC, charge extraction, and light and dark current-voltage curves (see Table I and Table SI in the Supplemental Material [34]). The parameters represent the standard set of parameters for a P3HT:PCBM organic solar cell.

We model the $J$-$V$ curve at 300 K and 1 sun using the AM1.5g spectrum. For the TPV and TPC simulations, we assume a typical measurement setup with bias light, small perturbation light, and data acquisition via an oscilloscope. The bias light has the same spectrum as above, and its intensity is changed between 0 and 1.26 suns. A cw laser with wavelength 309 nm and a pulse width of 5 ns is used for the small-signal perturbation. The ratio of laser versus bias-light intensity is adjusted in a way so that the ratio of excess photovoltage peak versus bias voltage $\Delta V/V$ is always in the range of 1%-2%. The resulting voltage transients can be almost exactly approximated by a single exponential decay function. In the case of TPV, the voltage is measured across an open circuit, and in the case of TPC, the short-circuit current is measured across a 50-$\Omega$ resistor.

We simulate five different cases of an organic solar cell (see Table I for the main simulation parameters):

1. equal electron and hole carrier densities, equal electron and hole mobilities, and no trapping;
2. equal electron and hole carrier densities, much higher electron than hole mobilities, and no trapping;
3. much higher hole than electron carrier densities, equal electron and hole mobilities, and no trapping;
4. equal electron and hole carrier densities, equal electron and hole mobilities, and strong trapping;
5. a realistic organic solar cell whose measured characteristics have been previously simulated in Ref. [30].

Case 1 is the ideal case and serves to demonstrate the validity of the theory behind the analytical model for extracting charge-carrier mobility. As in this case the mobilities of electrons and holes are equal and no trapping occurs, the spatially averaged mobility is independent of

| Parameter                                    | Unit     | Case 1   | Case 2   | Case 3   | Case 4   | Case 5   |
|----------------------------------------------|----------|----------|----------|----------|----------|----------|
| Electron density on right contact            | $\text{cm}^{-3}$ | $10^{20}$ | $10^{20}$ | $10^{19}$ | $10^{20}$ | $2.65 \times 10^{19}$ |
| Hole density on left contact                 | $\text{cm}^{-3}$ | $10^{20}$ | $10^{20}$ | $10^{20}$ | $10^{20}$ | $5.80 \times 10^{20}$ |
| DOS distribution                             |          |          |          |          |          |          |
| Effective electron trap density              | $\text{cm}^{-3} \cdot \text{eV}^{-1}$ | 0        | 0        | 0        | 10^{20}  | $3.80 \times 10^{20}$ |
| Effective hole trap density                  | $\text{cm}^{-3} \cdot \text{eV}^{-1}$ | 0        | 0        | 0        | 10^{20}  | $1.45 \times 10^{19}$ |
| Characteristic energy for electron tail      | $\text{meV}$ | 60       | 60       | 60       | 60       | 40       |
| Characteristic energy for hole tail          | $\text{meV}$ | 60       | 60       | 60       | 60       | 60       |
| Free-electron mobility                       | $\text{cm}^2/(\text{V s})$ | $2.48 \times 10^{-03}$ | $2.48 \times 10^{-03}$ | $2.48 \times 10^{-03}$ | $2.48 \times 10^{-03}$ | $2.48 \times 10^{-03}$ |
| Free-hole mobility                           | $\text{cm}^2/(\text{V s})$ | $2.48 \times 10^{-03}$ | $2.48 \times 10^{-04}$ | $2.48 \times 10^{-03}$ | $2.48 \times 10^{-03}$ | $2.48 \times 10^{-03}$ |
| Number of traps                              |          | 0        | 0        | 0        | 20       | 20       |
| LUMO electron-capture cross section          | $\text{cm}^{-2}$ | $10^{-29}$ | $10^{-29}$ | $10^{-29}$ | $10^{-26}$ | $2.50 \times 10^{-24}$ |
| LUMO hole-capture cross section              | $\text{cm}^{-2}$ | $10^{-29}$ | $10^{-29}$ | $10^{-29}$ | $10^{-26}$ | $1.32 \times 10^{-26}$ |
| HOMO electron-capture cross section          | $\text{cm}^{-2}$ | $10^{-29}$ | $10^{-29}$ | $10^{-29}$ | $10^{-26}$ | $4.67 \times 10^{-32}$ |
| HOMO hole-capture cross section              | $\text{cm}^{-2}$ | $10^{-29}$ | $10^{-29}$ | $10^{-29}$ | $10^{-26}$ | $4.86 \times 10^{-26}$ |
| Effective density of free-electron states    | $\text{cm}^{-3}$ | $5 \times 10^{20}$ | $5 \times 10^{20}$ | $5 \times 10^{20}$ | $5 \times 10^{20}$ | $1.28 \times 10^{21}$ |
| Effective density of free-hole states        | $\text{cm}^{-3}$ | $5 \times 10^{20}$ | $5 \times 10^{20}$ | $5 \times 10^{20}$ | $5 \times 10^{20}$ | $2.86 \times 10^{19}$ |
current. The same should happen for the one extracted from the method, as long as the condition of constant gradients of quasi-Fermi-levels are met. Cases 2–4 demonstrate step by step the resilience of the method as we move away from the case of a symmetric device with no trapping to an asymmetric device with considerable charge-carrier trapping.

Figure 3 shows the $J$-$V$ and $J_{SC}$-$V_{OC}$ curves of case 5. The remaining curves and the characteristic parameters $V_{OC}$, $J_{SC}$, FF, and solar-cell efficiency are included in the Supplemental Material [34]. Most interesting for our method is the difference between the $J$-$V$ and $J_{SC}$-$V_{OC}$ curves at the same current density, as this is proportional to the transport resistance $R_{tr}$ according to Eq. (17). Cases 1 and 2 lead to a higher FF than for case 5, so that the difference between the $J$-$V$ and $J_{SC}$-$V_{OC}$ curves is smaller than seen in Fig. 3. Case 3 has a lower $V_{OC}$ compared to cases 1 and 2 but a similar FF to cases 1 and 2. Therefore, $R_{tr}$ for those cases is lower, and the mobility is likely to be higher than for case 5, since $\mu$ is inversely proportional to $R_{tr}$. For case 4, the large trapping strongly affects all $J$-$V$ characteristic parameters but above all FF. As a result, it shows a very high transport resistance and, thus, a very low mobility.

VI. RESULTS AND DISCUSSION

At the core of the investigated method is Eq. (15), which states that the effective mobility is inversely proportional to the average carrier density and the transport resistance. Hence, the accuracy of the effective mobility crucially depends on how accurately we can determine these two parameters. The assumptions made in the derivation of Eq. (15) certainly play a role also. We notice that the determination of the carrier density and transport resistance have the highest impact on the shape of the extracted mobility. The spatial variation of the quasi-Fermi-levels, however, plays a minor role, as it is practically constant at $V_{OC}$ and varies only gradually at $J_{SC}$. We choose to outline its contribution in the Supplemental Material (see Sec. III. C in the Supplemental Material [34]).

A. Extraction of the excess carrier density

We first investigate the extraction of the excess carrier density $\Delta n_{TPC}$ from the TPC signal. As we delineate in the theory part, $\Delta n_{TPC}$ can be computed from the area under the current transient. We see that if the difference between carrier densities on the contacts, surface recombination, and trapping are neglected, $\Delta n_{TPC}$ is proportional to this area. In Fig. 4, the excess carrier density calculated from the TPC simulation $\Delta n_{TPC}$ is compared to the components of the right-hand side of Eq. (6) for case 1. Instead of plotting the excess carrier densities against $J_{bias}$, we convert the latter to $J_{SC}$ ($V_{OC}$) using Eq. (16) to better compare it to later results. In Fig. 4, a current density of 0 then corresponds to $V_{OC}$ and the asymptotic limit at high current densities to $J_{SC}$.

For case 1, it can be seen that excess carrier densities are fairly constant throughout the current-density range but drop rapidly towards $J_{SC}$. $\Delta n_{EL}$ is around 2 orders of magnitude lower than $\Delta n_{init}$ for almost the entire current-density range. The number of carriers lost to surface recombination is negligible, since $\Delta n_{surf}$ is more than...
5 orders of magnitude lower than $\Delta n_{\text{init}}$. As trapping is suppressed, bulk recombination does not influence the result. Consequently, we can say that $\Delta n_{\text{TPC}}$ indeed equals the initial excess carrier density at every current density for the case of an ideal solar cell. A similar scenario is found for cases 2 and 3 (see the Supplemental Material [34]).

In Fig. 4(b), the recombination via trap states is added (case 4). The absolute level of $\Delta n_{\text{init}}$ is smaller by about 1 order of magnitude compared to the case without trapping. This is rooted in the fact that the intensity of the small-signal light pulse has to be lowered so that the ratio of the peak excess voltage to the steady-state voltage $\Delta V/V$ is below 2%. As explained earlier, this is to ensure that the excess voltage decays monoexponentially.

The inclusion of trapping leads to a smaller difference between $\Delta n_{\text{init}}$ and $\Delta n_{\text{EL}}$ of about 1 order of magnitude and a much lower $\Delta n_{\text{surf}}$ compared to case 1. However, the bulk recombination via trap levels now plays a major role. At $V_{\text{OC}}$, the bulk recombination is the strongest, where $\Delta n_R$ is only a factor of 2 lower than $\Delta n_{\text{init}}$. For higher current densities, $\Delta n_R$ decreases much more strongly than $\Delta n_{\text{init}}$. According to Eq. (6), $\Delta n_{\text{TPC}}$ is, thus, slightly lower than $\Delta n_{\text{init}}$ because of the impact of $\Delta n_R$. Nevertheless, we can state that $\Delta n_{\text{TPC}}$ is still a good measure of $\Delta n_{\text{init}}$.

Case 5 contains the effect of very unbalanced charge-carrier densities, unbalanced mobilities, and trapping. Despite this, $\Delta n_{\text{TPC}}$ is only slightly lower than $\Delta n_{\text{init}}$, since the former is decreased by recombination and the change in carrier density on the contacts because of trapping and detrapping (see the Supplemental Material [34]). These findings show that the area under the small-signal current transient is indeed a good measure of the initial excess carrier density despite very unbalanced carrier properties or large trapping.

We now compare the average carrier density obtained from the TPV and TPC simulation, $\bar{n}_{\text{TPV,TPC}}$, with the theoretical $\bar{n}$ extracted at each point of the $J-V$ and $J_{\text{SC}}-V_{\text{OC}}$ curves. Figure 5(a) depicts the carrier density as a function of bias-light intensity (upper $x$ axis) and bias voltage $V$ (lower $x$ axis) of case 1. Here, we deal with equal electron and hole carrier densities and equal mobilities. The triangles show the theoretical carrier density $\bar{n}$ extracted from the steady-state part of the TPV signal. This carrier density corresponds to the total carrier density at the bias-light intensity or, alternatively, bias voltage. The diamonds stand for the photogenerated carrier density $\bar{n}_{\text{ph}}$, which is obtained from the former by subtracting the carrier density in the dark. $\bar{n}_{\text{TPV,TPC}}$ is the carrier density deduced from the combination of the TPV and TPC simulations by means of the differential capacitance method.

For very small light intensities, $\bar{n}$ remains nearly constant and equals the carrier density in the dark, since $\bar{n}_{\text{ph}}$ is minuscule in comparison. Only when the light intensity increases considerably, $\bar{n}_{\text{ph}}$ augments and approaches $\bar{n}$. In the description of the differential capacitance method above, we imply that $\bar{n}_{\text{TPV,TPC}}(V)$ includes only the photogenerated carrier density. This is corroborated by the findings of Fig. 5(a), as $\bar{n}_{\text{TPV,TPC}}(V)$ follows the shape of $\bar{n}_{\text{ph}}$. Albeit that, $\bar{n}_{\text{TPV,TPC}}(V)$ overestimates $\bar{n}_{\text{ph}}$. The gap closes only for light intensities above 0.5 suns, or in terms of voltage, close to $V_{\text{OC}} = 0.646 \text{ V}$ (vertical line in Fig. 5(a)).

The representation of Fig. 5(a) is very suitable to focus on the small-intensity range of the TPV measurement due to the exponential relationship between intensity and $V_{\text{OC}}$. Yet, the mobility is defined as a function of current density
in Eq. (15). For this purpose, we convert the bias-light intensity to current densities using the superposition principle as described by Eq. (16). Instead of plotting $\bar{n}$ versus the bias-light intensity or bias voltage as in Fig. 5(a), we now plot it versus the corresponding current density $J_{SC}$. Figure 5(b) depicts the so-deduced $\bar{n}$ as a function of $J_{SC}$ together with $\bar{n}$ computed at each point of the $J_{SC}$-$V_{OC}$ curve. The dashed line represents the photoinduced carrier density $\bar{n}_{ph}$ from the $J$-$V$ curve. The carrier density obtained from TPV and TPC $\bar{n}_{TPV,TPC}$ is plotted with circles.

The carrier densities $\bar{n}$ obtained from the $J$-$V$ and $J_{SC}$-$V_{OC}$ curves stay nearly constant for increasing current density and fall off only slightly when approaching $J_{SC}$. Both curves match perfectly over the entire current range. The carrier densities $\bar{n}_{ph}$ deduced from the $J$-$V$ curve and $J_{SC}$-$V_{OC}$ curves are also in perfect agreement but lie clearly below and exhibit a strong decrease for increasing current densities. As already seen in Fig. 5(a), $\bar{n}_{TPV,TPC}$ follows the same shape as $\bar{n}_{ph}$ but with slightly higher values.

We now shift the focus to Fig. 6, where we add deviations from the ideal case 1 in the form of unequal carrier densities, unbalanced mobilities, or trapping. Figure 6 displays the same meaning for the lines and symbols as in Fig. 5(b). On the left-hand side of Fig. 6, the cases 2 and 4 with equal electron and hole carrier densities are shown. The right-hand side depicts cases 3 and 5 with unbalanced carrier densities. The corresponding figures of the carrier densities as a function of bias voltage are included in the Supplemental Material [34].

For cases 2–5, it can be seen that $\bar{n}$ obtained from the current-voltage curves displays the same behavior as case 1: Both carrier density curves perfectly overlap and are...
much higher than \( n_{ph} \). The perfect agreement between both \( n_{ph} \) curves, on the contrary, is only true for the cases with equal mobilities (cases 3 and 5). For the cases with unbalanced mobilities, the curves coincide only for low current densities. The strongest difference between all cases and the most important one regarding the determination of the mobility is the offset between \( n_{ph} \) from the \( J_{SC}-V_{OC} \) curve and \( n_{TPV,TPC} \).

Now let us turn to each case in detail. For case 2, the hole mobility is lowered by 1 order of magnitude. This has the effect that \( n_{TPV,TPC} \) lies about 20\% below \( n_{ph} \) for the main current-density range. For case 3, the electron carrier density on the left contact is lowered by 1 order of magnitude compared to case 1 [Fig. 6(b)]. In practice, this means that the left contact is changed to a less Ohmic contact. As a result, all carrier densities are lowered by about 30\% over the entire current-density range compared to case 1. Furthermore, \( n_{TPV,TPC} \) overestimates \( n_{ph} \) by about 40\% for the entire current-density range.

Figure 6(c) plots the carrier densities for case 4, where trapping via electron and hole traps is added in comparison to case 1. The trapping does not impact on the total carrier density \( n \) and \( n_{ph} \) determined from the \( J_{SC}-V_{OC} \) curve. However, it causes a much stronger decrease of \( n_{TPV,TPC} \) than for cases 1 and 2. Thus, this decrease is clearly related to trapping, whose impact is expected to be more severe for higher light intensities, which translate to lower current densities via the superposition principle of Eq. (16). The simulation of the excess densities in Fig. 4(b) indeed unearths that \( \Delta n_{TPC} \) is slightly lower than \( \Delta n_{init} \) due to trapping.

Finally, case 5 incorporates all deviations from case 1 into one [Fig. 6(d)]. Regarding \( n_{TPV,TPC} \), two opposing trends come into play: Unbalanced mobilities and, above all, trapping entail that \( n_{TPV,TPC} \) is lowered compared to \( n_{ph} \), while unequal carrier densities at the contacts have the opposite effect. In sum, these affects cancel each other out, so that \( n_{TPV,TPC} \) matches \( n_{ph} \).

**B. Determination of transport resistance**

In Fig. 7, we plot \( R_u \) as a function of current density for all cases. \( R_u \) is obtained from the difference between the \( J-V \) and \( J_{SC}-V_{OC} \) curves [Eq. (17)]. For all cases, \( R_u \) exhibits an increase near \( V_{OC} \) and a sharp peak near \( J_{SC} \). The increase near \( V_{OC} \) stems from the fact that the current density becomes very small. At \( V_{OC} \) itself, \( R_u \) is not defined owing to the definition of Eq. (17). The sharp peak at \( J_{SC} \) is rooted in the fact that for voltages below the maximum power point, the voltage difference between the \( J-V \) and \( J_{SC}-V_{OC} \) curves increases strongly, while the current density remains fairly constant. The curve suggests that \( R_u \) is close to zero at \( J_{SC} \); however, this originates from the particular definition of the series resistance as a function of current density [Eq. (17)]. Converse to the peaked \( R_u \) observed in Fig. 7, Eq. (13) predicts that \( R_u \) should vary only slightly with current density. This is due to the fact that \( n \) is nearly constant except for a slight decrease towards \( J_{SC} \) [Fig. 5(b)]. As a consequence, since \( \mu_{TPV,TPC} \) is inversely proportional to \( R_u \), the features near \( V_{OC} \) and \( J_{SC} \) will be directly imprinted on \( \mu_{TPV,TPC} \) via Eq. (15).

In Sec. IV, we assume that the transport resistance dominates the series resistance. In our simulations, a contact resistance of 2.34 \( \Omega \text{cm}^2 \) is assumed; see the Supplemental Material, Table SI [34]. This contact resistance is taken from the measurement of a typical organic solar cell (case 5) [30]. Only for cases 1 and 3 (no trapping), \( R_u \) as low as the contact resistance for low current densities. Case 5 exhibits \( R_u \) as low as 2.5 \( \Omega \text{cm}^2 \) for very low current densities. For case 4, which includes trapping, the lowest \( R_u \) is about 7 \( \Omega \text{cm}^2 \). For current densities close to \( J_{SC} \), the transport resistance increases strongly and reaches values of 15–25 \( \Omega \text{cm}^2 \) for all cases. Schiefer et al. [22] measured the circuit and transport resistances of an organic solar cell with P3HT:PCBM as active layer using the combination of \( J-V \) and Suns-\( V_{OC} \) measurements. They found a circuit resistance of 2 \( \Omega \text{cm}^2 \), while the transport resistance varied with the current density between 6 and 23 \( \Omega \text{cm}^2 \) from \( V_{OC} \) to \( J_{SC} \). It is clear from these examples that the transport resistance is comparable to the circuit resistance for solar cells for current densities below \( J_{SC} \) and devices without trapping. When trapping occurs or the current densities are close to \( J_{SC} \), the series resistance is dominated by the transport resistance.

**C. Determination of mobility**

In Fig. 8, the mobility is depicted as a function of current density for all cases. For cases with equal electron and hole carrier densities at the contacts, \( \mu \) represents the spatially averaged mobility as defined in Eq. (7). For cases with unbalanced carrier densities, the spatially averaged
The first thing that comes to our attention is that \( \bar{\mu}_\text{TPV} \) slightly overestimates \( \bar{\mu}_\text{TPC} \) at \( J_\text{SC} \) stems from the strong impact of the curvature of the transport resistance. Figure 8(b) depicts case 2, where the hole mobility is lowered by 1 order of magnitude compared to case 1. Here, the theoretical mobilities of electrons and holes also equal the constant, predefined mobilities of the free electron and holes. For the main current-density range, \( \mu_{\text{TPV},\text{TPC}} \) is nearly flat and very close to \( \mu_{\text{eff}} \). It is slightly higher than the arithmetic average of \( 1.4 \times 10^{-4} \text{ cm}^2/(\text{Vs}) \) and clearly higher than the geometric average of \( 7.8 \times 10^{-4} \text{ cm}^2/(\text{Vs}) \).

FIG. 8. Spatially averaged mobilities as a function of current density. (a) Cases 1 and 3, (b) case 2, (c) case 4, and (d) case 5. The left panel shows the cases with equal electron and hole mobilities. Here, the continuous line corresponds to the average mobility \( \bar{\mu} \) as defined in Eq. (7), which is determined at each point of the \( J-V \) curve. The right panel depicts the cases with unbalanced mobilities. Here, the continuous and dashed lines are the average electron and hole mobilities \( \bar{\mu}_e \) and \( \bar{\mu}_h \), respectively. The dash-dotted green line corresponds to the effective mobility \( \mu_{\text{eff}} \) as defined in Eq. (15). In all graphs, the mobility determined from the investigated method \( \mu_{\text{TPV,TPC}} \) is plotted with circles.

The constant, predefined mobility of free carriers shown as a blue line. The theoretical \( \mu_{\text{eff}} \) predicts a slightly higher mobility over almost the entire current range before experiencing a sharp increase at \( J_\text{SC} \). Apart from the regions near \( V_\text{OC} \) and \( J_\text{SC} \), \( \mu_{\text{TPV,TPC}} \) is in good accordance with the theoretical one for both cases. The slightly lower mobilities as \( \mu_{\text{eff}} \) can be explained by the fact that for both cases, \( \bar{n}_{\text{TPV,TPC}} \) slightly overestimates \( \bar{n}_\text{ph} \) [Figs. 5(b) and 6(b)]. The differences of \( \mu_{\text{eff}} \) and \( \mu_{\text{TPV,TPC}} \) at \( J_\text{SC} \) stems from the strong impact of the curvature of the transport resistance.

Figure 8(a) exhibits cases 1 and 3 with equal electron and hole mobilities. The theoretical mobility, therefore, equals the constant, predefined mobility of free carriers shown as a blue line. The theoretical \( \mu_{\text{eff}} \) predicts a slightly higher mobility over almost the entire current range before experiencing a sharp increase at \( J_\text{SC} \). Apart from the regions near \( V_\text{OC} \) and \( J_\text{SC} \), \( \mu_{\text{TPV,TPC}} \) is in good accordance with the theoretical one for both cases. The slightly lower mobilities as \( \mu_{\text{eff}} \) can be explained by the fact that for both cases, \( \bar{n}_{\text{TPV,TPC}} \) slightly overestimates \( \bar{n}_\text{ph} \) [Figs. 5(b) and 6(b)]. The differences of \( \mu_{\text{eff}} \) and \( \mu_{\text{TPV,TPC}} \) at \( J_\text{SC} \) stems from the strong impact of the curvature of the transport resistance.
As $\bar{n}_{TPV,TPC}$ slightly underestimates $\bar{n}_{ph}$ [Fig. 6(a)], $\bar{\mu}_{TPV,TPC}$ is slightly higher than $\mu_{eff}$.

Now we wish to discuss the impact of trapping on the mobility. For balanced carrier densities and mobilities, the effect of trapping can be best appreciated by comparing case 1 and case 4 in the left panel of Fig. 8. There are two changes that affect both the theoretical mobility $\bar{\mu}$ and $\bar{\mu}_{TPV,TPC}$: First, the carriers become less mobile by about 1 order of magnitude. Second, the mobility becomes dependent on current density with a negative slope. The first change is rooted in the fact that the trapped carrier density exceeds the free-carrier density by more than 1 order of magnitude (see Fig. S6(a) in the Supplemental Material [34]). This strongly decreases the ratio of free- to total carrier density and, therefore, $\bar{\mu}$ [Eq. (7)]. The second change stems from the fact that trapping is the strongest at $J_{SC}$ and diminishes with decreasing current density (Fig. S6(b) in the Supplemental Material [34]). Even though $\mu_{eff}$ is very close to $\bar{\mu}$, $\bar{\mu}_{TPV,TPC}$ is higher than $\bar{\mu}$ by about a factor of 2 in the main current-density range. This is a direct cause of the fact that $\bar{n}_{TPV,TPC}$ strongly underestimates $\bar{n}$.

Now coming to the last case, Fig. 8(d) plots the results for a typical organic solar cell that combines all the influences of cases 2–4 into one. On the one hand, $\mu_{eff}$ and $\bar{\mu}_{TPV,TPC}$ exhibit the same current-density dependence as observed in Fig. 8(c). On the other hand, the vertical position of $\bar{\mu}_{TPV,TPC}$ is due to the combined impact of the unbalanced mobilities [Fig. 8(b)] and trapping [Fig. 8(c)]. Hence, owing to the fact that $\bar{n}_{TPV,TPC}$ agrees well with $\bar{n}_{ph}$, $\bar{\mu}_{TPV,TPC}$ is in good accordance with $\mu_{eff}$ in the main current-density range.

In the Supplemental Material [34], we add simulation of further cases. They are variations of cases 2 and 3 for which we increase the unbalance between mobilities and carrier densities. In the variations of case 2 (cases 2b and c), we decrease the hole mobility by 2 and 4 orders of magnitude, respectively, compared to the electron mobility. In the variations of case 3 (cases 3b and c), we reduce the electron charge on the right contact to $10^{18}$ and $10^{10}$ cm$^{-3}$, respectively, so that the ratio of hole to electron charge on the contacts is increased to 2 and 10 orders of magnitudes, respectively. In many thin-film solar cells, highly unbalanced mobilities and charge-carrier densities are a stark reality, for example, caused by a very poor electron-selective contact. Furthermore, many organic materials suffer from a much lower electron than hole mobility [35]. When the hole mobility is decreased by 2 orders of magnitude, $\bar{\mu}_{TPV,TPC}$ is fairly constant and agrees well with $\mu_{eff}$ for the main current-density range (case 2b). Even for a further decrease of hole mobility, a good agreement between $\bar{\mu}_{TPV,TPC}$ and $\mu_{eff}$ is found for lower current densities (case 2c). For larger current densities, $\mu_{eff}$ predicts a much lower mobility as determined by $\bar{\mu}_{TPV,TPC}$. The mobilities are much closer to $\mu_{eff}$ than to $\sqrt{\bar{\mu}_{TPV,TPC}}$, in contrast to what has been postulated in Refs. [1,22]. For the variation of case 3, the following results are obtained: For an electron charge on the right contact to $10^{18}$ cm$^{-3}$, $\bar{\mu}_{TPV,TPC}$ exhibits a very similar agreement as that shown for the two cases in Fig. 8(a). When the electron charge is further decreased to $10^{10}$ cm$^{-3}$, however, $\bar{\mu}_{TPV,TPC}$ becomes strongly dependent on current density and agrees only with the theoretical mobility in a small-current-density range. Nevertheless, $\bar{\mu}_{TPV,TPC}$ determined at mid-current densities remains a good estimate of the theoretical mobility.

VII. Conclusions

A method previously proposed by Albrecht et al. and Schiefer et al. that is capable of determining the charge-carrier mobility in disordered thin-film solar cells is revisited and generalized. Revising the theory unveils that only an effective mobility can be determined, which, in general, is composed of a mix of mobility and carrier density terms. This mix depends on the exact nature of the carrier density measured by the method, that is, the total carrier density or the photoinduced one. Drift-diffusion simulation of organic solar cells shows that in the case of equal mobilities and carrier densities, the extracted mobility is in excellent agreement with the theoretical one. In the case of unequal electron and hole mobilities, the techniques determine a mobility close to $\mu_{eff}$. In the case of unequal carrier densities, $\bar{\mu}_{TPV,TPC}$ is fair close to $\mu_{eff}$ to the ratio of 2 orders of magnitude, the mobility agrees very well with the theoretical one. When trapping is present in the device, the method is able to measure the trapping-induced current-density dependence of the mobility. The accuracy of determining the mobility depends inversely on the one of the experimentally determined average carrier density.

However, the accurate extraction of mobility is restricted to current-density ranges away from $V_{OC}$ and $J_{SC}$. Near $V_{OC}$ and $J_{SC}$, the limitation is rooted in the fact that the transport resistance cannot be correctly determined. It is important to add that this can be circumvented for $V_{OC}$ if the transport resistance is extracted from the $J-V$ and $J_{SC}-V_{OC}$ curves measured at considerably higher intensities than 1 sun. Then, $V_{OC}$ at 1 sun will correspond to a point on the higher-intensity $J-V$ curve, which has the same voltage but a nonzero current. The intensity should be chosen in a manner so that the 1-sun-$V_{OC}$ point moves to the flatter part of the mobility versus the current-density curve.

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