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Journal Name

ARTICLE

A simple method to experimentally determine the accurate RC-constant in nanosecond timescale transient photocurrent measurements on organic solar cells

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Nanosecond timescale transient photocurrent (ns-TPC) measurements on organic solar cells (OSCs) are commonly used in combination with numerical simulation to study charge transport and recombination phenomenon in these devices. But the ns-TPC measurement itself is influenced by the RC-effects of the test circuit. Thus the RC-constant of the test circuit is needed to mathematically eliminate the RC-effects to reconstruct the accurate TPC signal. Nowadays, an estimated RC-constant is used by researchers to reconstruct the TPC signal. So, a reliable method is needed to experimentally determine the RC-constant accurately to reconstruct the accurate TPC signal. Here, a simple method, by analyzing the transient response of the test circuit after a square voltage pulse excitation, is used to experimentally determine the RC-constant in ns-TPC measurements on typical planar hetero-junction small-molecule organic solar cells and typical bulk hetero-junction polymer solar cells. In the meantime, in order to verify the correctness of the experimentally determined RC-constant, three verification methods, which are valid under specific conditions, are selectively adopted to verify whether the experimentally determined RC-constant is reliable. Finally, all the results given by the verification methods show that this simple method could be used as a reliable method to experimentally determine the correct RC-constant in ns-TPC measurements on OSCs.

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Introduction

Organic solar cells (OSCs) have been attracting more and more attention due to the potential to fabricate light weight, flexible and low-cost devices.^{1,2} Nowadays, the power conversion efficiency of typical thin film OSCs, namely planar hetero-junction small-molecule organic solar cells and bulk hetero-junction polymer solar cells have exceeded 11%.³ In order to further optimize device performance, a thorough understanding of device physics is needed. Nanosecond timescale transient photocurrent (ns-TPC) technique is commonly used in combination with numerical simulation^{4,5} to study charge transport and recombination phenomenon^{6,7} and the energetic distribution of trapping states^{8,9} in OSCs. In addition, this method is used to study the recombination of charge transfer excitons in polymer solar cells.¹⁰ And it is also commonly used in dye-sensitized solar cells (DSSCs).¹¹ Though much progress had been made on this area, ns-TPC measurements on OSCs are suffered from the RC-effects of the test circuit caused by the series connection of load resistor and

device geometrical capacitor.^{5,12,13} Compensation method is thus adopted to reduce the RC-effects of the test circuit. Devices are fabricated with smaller area and connected with smaller load resistor to reduce the RC-constant of the test circuit, thus the impact of the RC-effects would not be that significant.^{6,14,15,16} But the RC-constant still couldn't be reduced to a negligible level, because the usual OSCs are fabricated with large area with large geometrical capacitance. So, mathematical method is proposed to completely eliminate the RC-effects of the test circuit to reconstruct the accurate TPC signal.^{5,12,13} In order to reconstruct the accurate TPC signal, the accurate RC-constant of the test circuit is needed. But an estimated RC-constant of the test circuit is used by many researchers to reconstruct the TPC signal.^{6,10,17} Thus a reliable method is needed to experimentally determine the RC-constant of the test circuit accurately to reconstruct the accurate TPC signal.

Here, a simple method to experimentally determine the accurate RC-constant in ns-TPC measurements on OSCs is proposed. In this method, a waveform generator is used to apply a square voltage pulse to the test circuit, and the RC-constant of the test circuit is determined by simply analyzing the transient response of the test circuit after the square voltage pulse excitation (see Fig. 1(b) and equation (4)). In order to verify the correctness of the experimentally determined RC-constant, three verification methods, which are also experimental but only valid under specific conditions

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are selectively adopted to verify whether the experimentally determined RC-constant is reliable.

Device A, which has the structure of indium tin oxide (ITO)/copper phthalocyanine (CuPc)/fullerene (C₆₀)/tris-8-hydroxy-quinolinato aluminum (Alq₃)/aluminum (Al), is chosen as representative of planar hetero-junction small-molecule organic solar cells to conduct ns-TPC measurements, and the proposed simple method is used to experimentally determine the RC-constant of the test circuit. In the meantime, verification method 1 (see below) and verification method 2 (see below) are here chosen as the suitable verification methods to verify the correctness of the RC-constant got by the simple method. In the end, the proposed simple method gives an RC-constant of 1019.2 ns of the test circuit. The two verification methods give almost identical results, which are 1009.1 ns and 1010 ns, respectively. And the relative error of these three values is only 1%, indicating the correctness of the RC-constant determined by the proposed simple method. Also, device B, which has the structure of ITO/polyethylenimine ethoxylated (PEIE)/ poly(3-hexylthiophene) (P3HT): phenyl C(61)-butyric acid methyl ester (PCBM)/molybdenum oxide (MoO₃)/Al is chosen as representative of typical bulk hetero-junction polymer solar cells to conduct ns-TPC measurements, and the proposed simple method is used to experimentally determine the RC-constant of the test circuit. Because the TPC signal of device B shows a long tail extending to a few microsecond (see Fig. 4(c)), verification methods 1 and 2 are thus not suitable for device B. Thus verification method 3 (see below) is here chosen to verify the correctness of the RC-constant determined by the simple method. In the end, the proposed simple method gives an RC-constant of 204.4 ns of the test circuit and a geometrical capacitance of 1.997 nF of the device B. The TPC signal reconstructed using verification method 3 with the RC-constant of 204.4 ns of the test circuit (see verification method 3 and equation (3)) is almost overlapping with the TPC signal reconstructed using verification method 3 with the geometrical capacitance of 1.997 nF of the device (see verification method 3 and equation (10)), indicating the correctness of the RC-constant determined by the proposed simple method. Thus, all the results above show that the proposed simple method could be used as a reliable method to experimentally determine the correct RC-constant in ns-TPC measurements on OSCs.

Method

A. RC-effects in ns-TPC measurements on OSCs

Fig. 1(a) shows the schematic diagram of the ns-TPC measurement setup. The input impedance of the digital storage oscilloscope (DSO) is switched to 50 Ohms. When the cell is irradiated by a laser pulse, the active layer within the cell will convert the photons to excitons. After the dissociation of the excitons to electrons and holes, the generated charge carriers would drift and diffuse towards their respective electrodes. The moving carriers within the cell will produce the

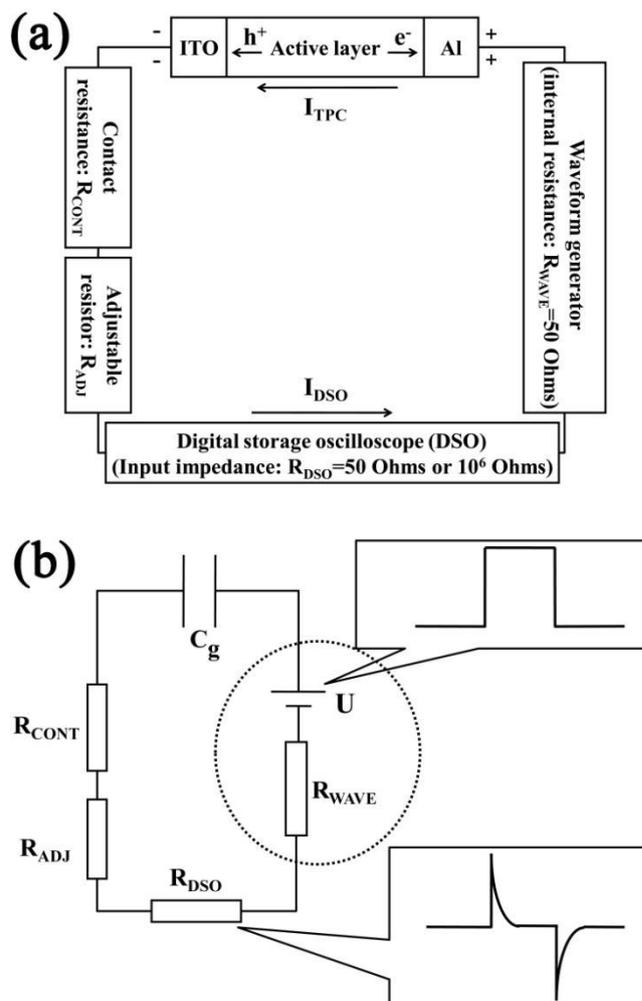


Fig. 1. (a) Schematic diagram of the ns-TPC measurement setup. The total load resistance of the test circuit is $R = R_{\text{CONT}} + R_{\text{ADJ}} + R_{\text{DSO}} + R_{\text{WAVE}}$, where R_{CONT} is the contact resistance, $R_{\text{DSO}} = 50 \text{ Ohms}$ or 10^6 Ohms is the input impedance of the DSO, $R_{\text{WAVE}} = 50 \text{ Ohms}$ is the internal resistance of the waveform generator and R_{ADJ} represents an adjustable resistor that is used to adjust the impedance matching point of the test circuit. (b) Equivalent electric circuit of the ns-TPC measurement setup. Normally the shunt resistance of optimized OSCs is rather large, so the shunt current is negligible. Thus the cell is equivalent to C_g , which is the geometrical capacitance of the cell. The elements within the dotted-line ellipse represent the waveform generator.

TPC signal I_{TPC}

$$I_{\text{TPC}} = \frac{q^+}{L} v^+ + \frac{q^-}{L} v^- = \frac{q^+}{L} [d^+(t)]' + \frac{q^-}{L} [d^-(t)]' = \frac{q}{L} [d^+(t) - d^-(t)]', \quad (1)$$

where $q = q^\pm$ is the quantity of electric charge, v^\pm is the velocity of the carriers, L is the thickness of the cell and $[d^\pm(t)]'$ denotes the rate of change of the carriers' position. At the same time, there is current I_{DSO} flowing through the load resistor of the DSO. The relation between I_{TPC} and I_{DSO} is

$$I_{\text{DSO}} R + \int_0^{I_{\text{DSO}}} dt = \frac{\sigma}{\epsilon} D(t) = \frac{L}{\epsilon \cdot S} \frac{\sigma \cdot S}{L} D(t) = \frac{1}{C_g} \frac{q}{L} D(t) = \frac{1}{C_g} \frac{q}{L} [d^+(t) - d^-(t)], \quad (2)$$

where $R = R_{CONT} + R_{ADJ} + R_{DSO} + R_{WAVE}$ is the total resistance of the test circuit, $I_{DSO}R$ denotes voltage drop across all the load resistors, C_g is the geometrical capacitance of the cell, $\int_0^t I_{DSO} \cdot dt$ denotes the voltage drop across the cell's geometrical capacitor, σ is the electric charge density per unit area, S is the area of the cell, ϵ is the dielectric constant, $D(t)$ is the separation distance of the opposite charges, and $\frac{\sigma}{\epsilon} D(t)$ denotes the electromotive force produced by the separation of electrons and holes. By differentiating equation (2), we get

$$I'_{DSO}RC_g + I_{DSO} = I_{TPC}. \quad (3)$$

So the TPC signal I_{TPC} is influenced by the RC-effects of the test circuit and thus the measured transient signal I_{DSO} using DSO is so not the real TPC signal I_{TPC} . Fortunately if the RC-constant RC_g of the test circuit could be determined, the real TPC signal I_{TPC} could also be worked out.^{5,12,13}

B. A simple method to experimentally determine the RC-constant

A simple method is proposed to experimentally determine the RC-constant of the test circuit. Fig. 1(b) shows the equivalent electric circuit of the ns-TPC measurement setup. It is a typical RC series circuit. As shown in the upper right of Fig. 1(b), if a square voltage pulse is applied to the RC series circuit by the waveform generator U, the DSO will record the transient response of the circuit, as shown in the lower right of Fig. 1(b). This is a typical charging or discharging curve of an RC series circuit. The tail of the charging or discharging curve is a typical single exponential decay curve with constants A and B

$$I = A + B \exp\left(\frac{-t}{RC_g}\right). \quad (4)$$

Thus, by fitting the tail to a single exponential decay function, the RC-constant RC_g of the test circuit could be determined. Furthermore, integral of the charging or discharging curve will produce the quantity of electric charge storing in the geometrical capacitor. Taking the amplitude of the applied voltage pulse into consideration, the geometrical capacitance C_g could also be extracted

$$C_g = \frac{Q}{V}, \quad (5)$$

where V is the amplitude of the applied voltage pulse and Q is the quantity of electric charge storing in the geometrical capacitor. And once the RC-constant RC_g and the geometrical capacitance C_g have been measured, the load resistance R could also be determined.

C. Three verification methods to verify the correctness of the experimentally determined RC-constant

Three verification methods are also used to verify the correctness of the RC-constant measured using the above proposed simple method.

VERIFICATION METHOD 1 - FITTING THE TAIL OF THE I_{DSO} CURVE: As shown in equation (6) and (7), if the TPC signal I_{TPC} decays to zero much faster than the I_{DSO} signal does, the long tail of the I_{DSO} signal is simply the discharging current of the stored electric charge in the geometrical capacitor

$$I'_{DSO}RC_g + I_{DSO} = I_{TPC} = 0, \quad (6)$$

thus

$$I_{DSO} = C + D \exp\left(\frac{-t}{RC_g}\right). \quad (7)$$

By fitting the long tail of the I_{DSO} signal to a single exponential decay function with constants C and D, the RC-constant RC_g of the test circuit could also be determined.

VERIFICATION METHOD 2 - CHARGE CONSERVATION

CONDITION: The integral of the I_{TPC} curve gives the quantity of charge carriers that move within the cell and the integral of the I_{DSO} curve gives the quantity of charge carriers that flow through the load resistor. Normally, for optimized cells, charge recombination is negligible, and so the generated charge carriers by the excitation of a laser pulse will all be collected by the electrodes and finally all flow through the load resistor. Thus the charge conservation condition is

$$\int I_{TPC} \cdot dt = \int I_{DSO} \cdot dt. \quad (8)$$

Thus the I_{TPC} curve that meets the conservation condition is the correct I_{TPC} curve. And the RC-constant RC_g that is used in equation (3) to determine the correct I_{TPC} curve is the correct RC-constant.

For device A, verification method 1 and verification method 2 are adopted as the suitable verification methods to verify the correctness of the experimentally determined RC-constant. For device B, the TPC signal I_{TPC} of device B shows a long tail extending to a few microseconds (see Fig. 4(c)). Verification methods 1 and 2 are thus not suitable for device B to verify the correctness of the experimentally determined RC-constant. This time, verification method 3 is proposed as a suitable verification method to verify the correctness of the experimentally determined RC-constant.

VERIFICATION METHOD 3 - THE TPC METHOD AND THE TPC METHOD SHOULD GIVE THE SAME TPC SIGNAL I_{TPC} :

As shown in Fig. 1(a), if the input impedance of the DSO is switched to 10^6 Ohms, the current I_{DSO} that flows through the load resistor is thus so small that could be ignored. At this condition, the relation between I_{TPC} and I_{DSO} is

$$I_{TPC} = I'_{DSO}RC_g. \quad (9)$$

Since the input impedance of the DSO is much larger than the other load resistance of the test circuit, equation (9) could also be written as

$$I_{TPC} = I'_{DSO} R_{DSO} C_g = C_g \frac{dV_{DSO}}{dt}, \quad (10)$$

where V_{DSO} is the voltage drop across the load resistor of the DSO. This is the TPV method, in which the input impedance of the DSO is switched to 10^6 Ohms and the DSO record the voltage drop across the load resistor of the DSO and once the geometrical capacitance C_g of the cell could be determined, the I_{TPC} signal could be worked out. In addition, equation (3) above describes the TPC method. Thus if the RC-constant RC_g of the test circuit and the geometrical capacitance C_g of the cell could all be measured correctly, the TPC method and the TPV method should finally give almost the same I_{TPC} signal. This is the third verification method to verify the correctness of the experimentally determined RC-constant. Note that verification method 3 is not that easy to use, because it is time consuming to adjust the adjustable resistor to meet the impedance matching condition of the test circuit.

Experimental

Two kinds of devices with an active area of 0.09 cm^2 are fabricated. Device A is a planar hetero-junction small-molecule organic solar cell (ITO/CuPc(20nm)/C₆₀(55nm)/Alq₃(6nm)/Al) and device B is a bulk hetero-junction polymer solar cell (ITO/PEIE/P3HT:PCBM/MoO₃/Al). The device fabrication methods are the same as those used previously.^{18,19} The current-voltage (IV) characterization is performed under the standard 1 sun air mass 1.5 global radiation (AM1.5G) condition, using a solar simulator (Newport 94043A) and a source measure unit (Keithley 2400). And the external quantum efficiency (EQE) is obtained using an EQE characterization system (Newport QEPVSI-B). Then in ns-TPC test, the cells are connected directly in series with an adjustable resistor, a waveform generator and a DSO (Agilent DSO-X3102A) and the input impedance of the DSO is switched to 50 Ohms (the TPC method) or 10^6 Ohms (the TPV method), depending on the specific TPC or TPV method we used (see Fig. 1(a)). Note that the adjustable resistor is used to adjust the impedance matching point of the test circuit, the waveform generator is used to apply the square voltage pulse to the circuit and the amplitude of the square voltage pulse is 20 mV. The exciting source is a diode laser module (Cobolt MLD 0405-06-01-0100-100) that is modulated by a square voltage pulse generated by a waveform generator (Agilent DSO-X3102A). The pulse width and the wavelength of the pulsed laser beams are approximately 5 ns and 405 nm respectively. Also the laser power is carefully tuned to ensure that the maximum change in voltage across a cell caused by the laser illumination is within 35 mV. After laser excitation, the moving carriers within the cell will produce the TPC signal I_{TPC} . At the same time, there is current I_{DSO} flowing through the load resistor of the DSO. Thus the electric charges will accumulate in the geometrical capacitor of the cell. This is equivalent to charging the capacitor by applying voltage to it. Since we have carefully attenuated the laser power and the maximum change in voltage across the cell caused by the laser illumination is

within 35 mV (see Fig. 4(b)), we thus choose to use a waveform generator to apply a square voltage pulse with amplitude of 20 mV to the circuit.

Results and discussion

The IV curves of device A and device B are plotted in Fig. 2(a). According to the curves obtained under illumination, the open circuit voltage (V_{OC}), short circuit current density (J_{SC}), fill factor (FF) and power conversion efficiency (PCE) of devices A and B are 0.475 V, 4.21 mA/cm², 57%, 1.15% and 0.57 V, 10.07 mA/cm², 57%, 3.29%, respectively. Fig. 2(b) shows the EQE spectra of these two devices.

For device A, verification method 1 and verification method 2 are adopted as the suitable verification methods to verify the correctness of the experimentally determined RC-constant. The cell is wired directly in series with an adjustable resistor, a waveform generator and a DSO and the input impedance of the DSO is switched to 50 Ohms, as shown in Fig. 1(a). We first use the waveform generator to apply a square voltage pulse to the test circuit, and the corresponding transient response is recorded by the DSO, as shown in Fig. 3(a) (open circle).

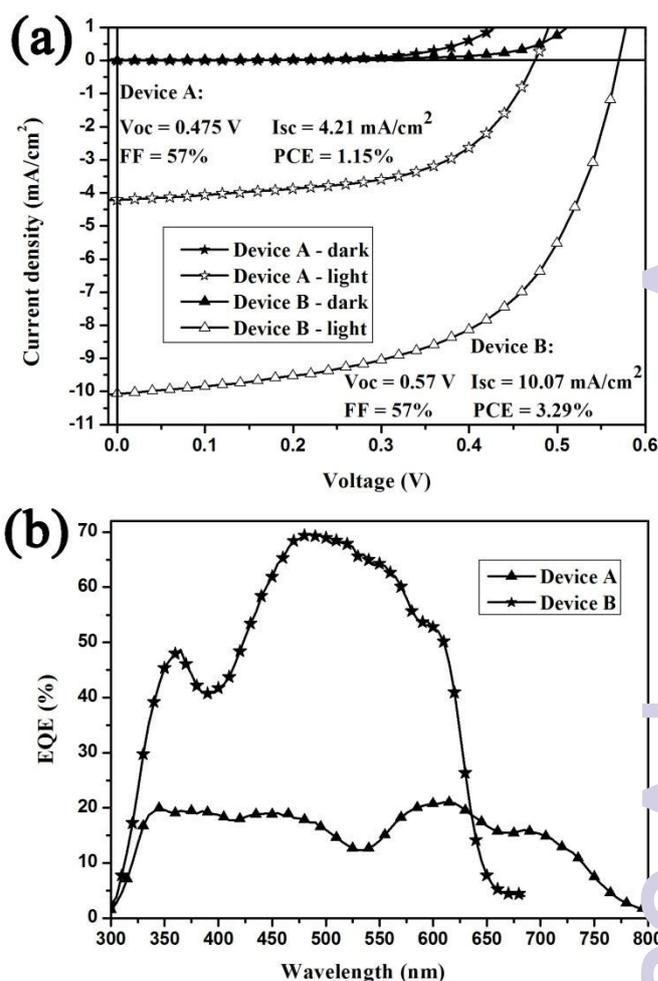


Fig. 2. (a) Light and dark IV curves of devices A and B. (b) EQE spectra of devices A and B.

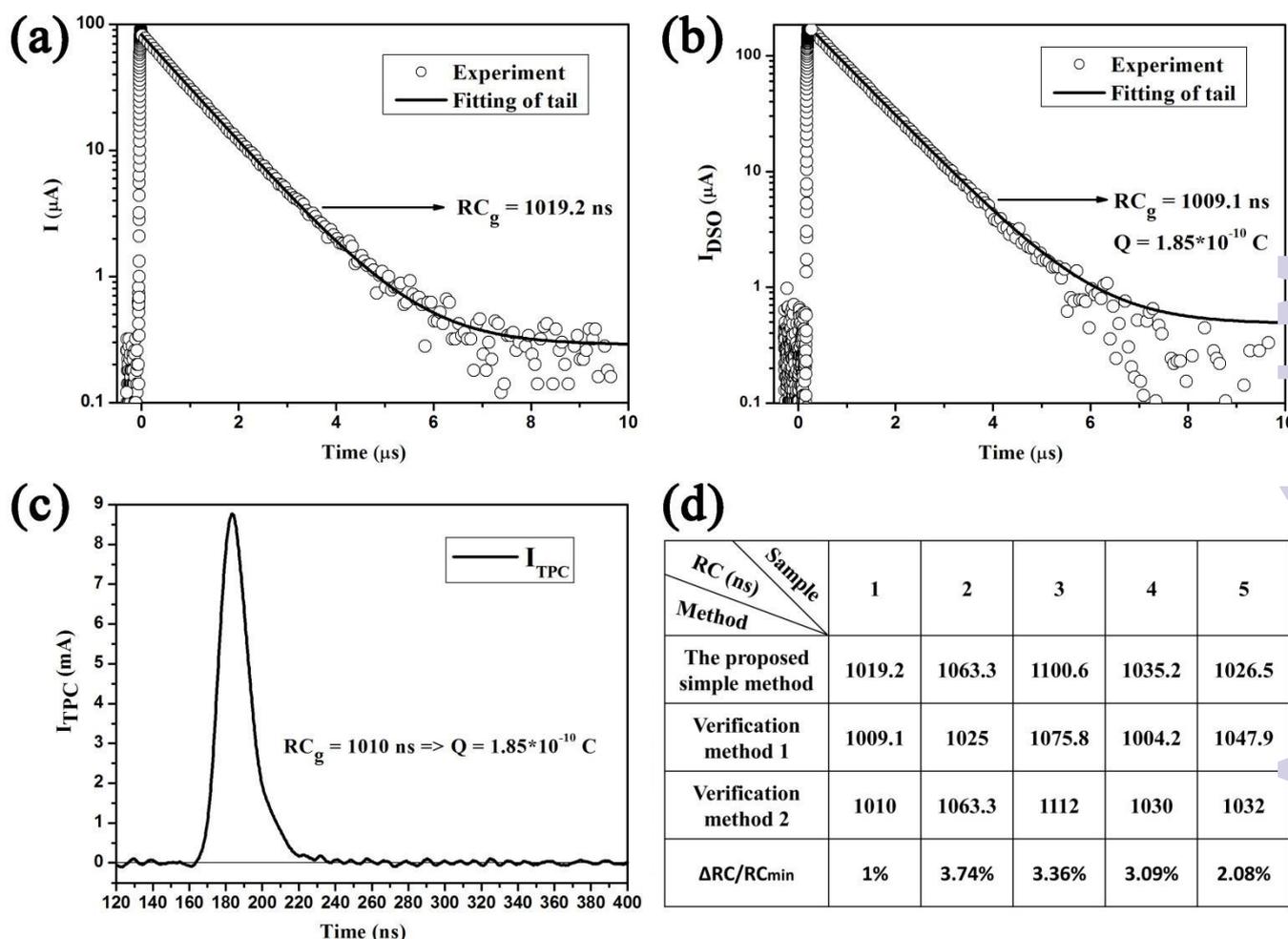


Fig. 3. (a) Transient response of the test circuit after a square voltage pulse excitation by the waveform generator (open circle). The solid line represents the single exponential decay fitting curve of the tail of the charging signal, indicating an RC-constant of 1019.2 ns. Then the waveform generator is set to output a constant 0 V voltage. Following that, a laser pulse is applied to the cell. Fig. 3(b) shows the I_{DSO} signal recorded by the DSO after laser excitation (open circle). Also the solid line in Fig. 3(b) is the single exponential decay fitting curve of the tail of the I_{DSO} signal, indicating an RC-constant of 1009.1 ns. Furthermore, the integral of the I_{DSO} curve gives a total charge of $1.85 \times 10^{-10} \text{ C}$ flowing through the load resistor. At the same time, Fig. 3(c) is the I_{TPC} signal calculated using a guessing RC-constant of 1010 ns. The integral of the I_{TPC} signal also gives a total charge of $1.85 \times 10^{-10} \text{ C}$ moving within the cell. In summary, the proposed simple method gives an RC-constant of 1019.2 ns of the test circuit. The two verification methods give almost identical results, which are 1009.1 ns and 1010 ns, respectively. And the relative error of these three values is only 1%, indicating the correctness of the RC-constant determined by the proposed simple method. Another 4 samples are also used to conduct the same experiment, and all the data is shown in Fig. 3(d). The data show that the proposed simple method could be used as a reliable method to experimentally determine the correct RC-constant of the test circuit. For device B, verification method 3 is adopted as the suitable verification methods to verify the correctness of the experimentally determined RC-constant. In the first place, the

The solid line in Fig. 3(a) is the single exponential decay fitting curve of the tail of the charging signal, indicating an RC-constant of 1019.2 ns. Then the waveform generator is set to output a constant 0 V voltage. Following that, a laser pulse is applied to the cell. Fig. 3(b) shows the I_{DSO} signal recorded by the DSO after laser excitation (open circle). Also the solid line in Fig. 3(b) is the single exponential decay fitting curve of the tail of the I_{DSO} signal, indicating an RC-constant of 1009.1 ns. Furthermore, the integral of the I_{DSO} curve gives a total charge of $1.85 \times 10^{-10} \text{ C}$ flowing through the load resistor. At the same time, Fig. 3(c) is the I_{TPC} signal calculated using a guessing RC-constant of 1010 ns. The integral of the I_{TPC} signal also gives a total charge of $1.85 \times 10^{-10} \text{ C}$ moving within the cell. In summary,

the proposed simple method gives an RC-constant of 1019.2 ns of the test circuit. The two verification methods give almost identical results, which are 1009.1 ns and 1010 ns, respectively. And the relative error of these three values is only 1%, indicating the correctness of the RC-constant determined by the proposed simple method. Another 4 samples are also used to conduct the same experiment, and all the data is shown in Fig. 3(d). The data show that the proposed simple method could be used as a reliable method to experimentally determine the correct RC-constant of the test circuit. For device B, verification method 3 is adopted as the suitable verification methods to verify the correctness of the experimentally determined RC-constant. In the first place, the

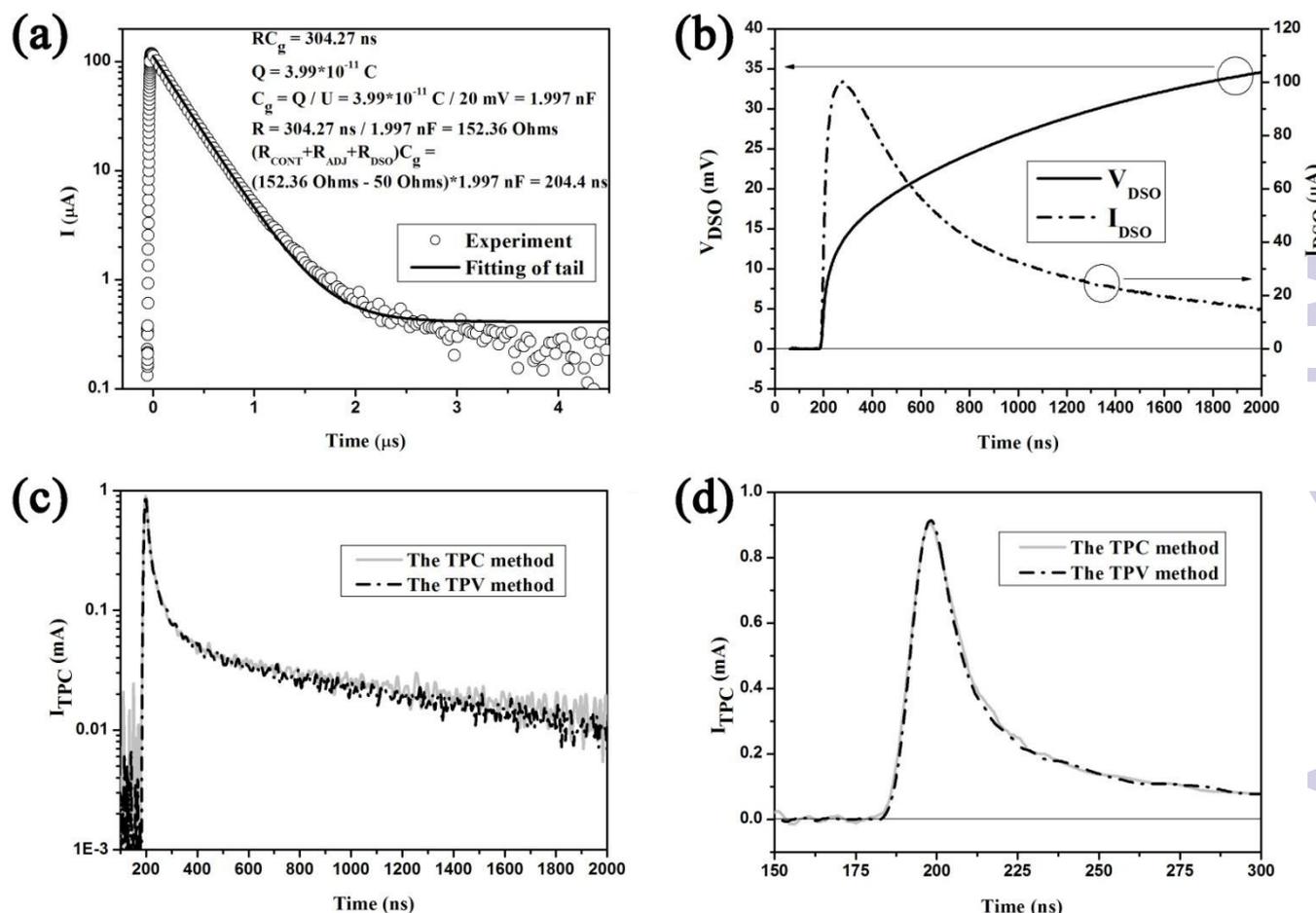


Fig. 4. (a) Transient response of the test circuit after a square voltage pulse excitation by the waveform generator (open circle). The solid line is a single exponential decay fitting curve. (b) I_{DSO} signal (dash dot line) and V_{DSO} signal (solid line) of the cell after the laser pulse excitation. (c) and (d) I_{TPC} signals calculated using the TPC method and the TPV method.

cell is connected in series with an adjustable resistor, a waveform generator and a DSO and the input impedance of the DSO is switched to 50 Ohms, as shown in Fig. 1(a). We use the waveform generator to apply a square voltage pulse to the test circuit, and the corresponding transient response is recorded by the DSO, as shown in Fig. 4(a) (open circle). The line in Fig. 4(a) is the single exponential decay fitting curve of the tail of the charging signal, indicating an RC-constant of 304.27 ns. The integral of the charging curve gives a total charge of 3.99×10^{-11} C storing in the geometrical capacitor. Because the amplitude of the square voltage pulse is 20 mV, the geometrical capacitance C_g is thus calculated to be $(3.99 \times 10^{-11} \text{ C}) / (20 \text{ mV}) = 1.997 \text{ nF}$. So, total load resistance of the test circuit is $R = R_{\text{CONT}} + R_{\text{ADJ}} + R_{\text{DSO}} + R_{\text{WAVE}} = 304.27 \text{ ns} / 1.997 \text{ nF} = 152.36 \text{ Ohms}$. Second, the waveform generator is then

removed from the test circuit and following that, a laser pulse is applied to the cell. The corresponding I_{DSO} signal is recorded by the DSO and shown in Fig. 4(b) (dash dot line). Third, the input impedance of the DSO is switched to 10^6 Ohms and the corresponding V_{DSO} signal is recorded by the DSO after laser excitation, as shown in Fig. 4(b) (solid line). Because the waveform generator has been removed from the test circuit when conducting the ns-TPC measurement, the actual RC-constant is determined to be $(R_{\text{CONT}} + R_{\text{ADJ}} + R_{\text{DSO}}) * C_g = (152.36 \text{ Ohms} - 50 \text{ Ohms}) * 1.997 \text{ nF} = 204.4 \text{ ns}$. Thus the I_{TPC} signals are respectively calculated using the TPC method with an RC-constant of 204.4 ns and using the TPV method with a geometrical capacitance of 1.997 nF, as shown in Fig. 4(c) and (d). The calculated I_{TPC} signals are almost overlapping with each other, indicating the correctness of the values of the

experimentally determined RC-constant of the test circuit and the geometrical capacitance of the cell. Another 2 samples are also used to conduct the same experiment. The calculated I_{TPC} signals are almost the same, too. Thus, the result also shows that the proposed simple method could be used as a reliable method to experimentally determine the correct RC-constant of the test circuit.

Thus, all the results above show that the proposed simple method could be used as a reliable method to experimentally determine the correct RC-constant in ns-TPC measurements on OSCs.

Conclusions

In conclusion, a simple method is used to experimentally determine the accurate RC-constant in ns-TPC measurements on typical planar hetero-junction small-molecule organic solar cells and bulk hetero-junction polymer solar cells. In the meantime, three verification methods, which are also experimental but only valid under specific conditions, are selectively adopted to verify whether the experimentally determined RC-constant is reliable. The results presented in this article show that this simple method could be used as a reliable method to experimentally determine the correct RC-constant in ns-TPC measurements on OSCs. We believe that the proposed simple method could be used in typical ns-TPC measurements on OSCs to experimentally determine the accurate RC-constant and thus to reconstruct the TPC signal more accurately.

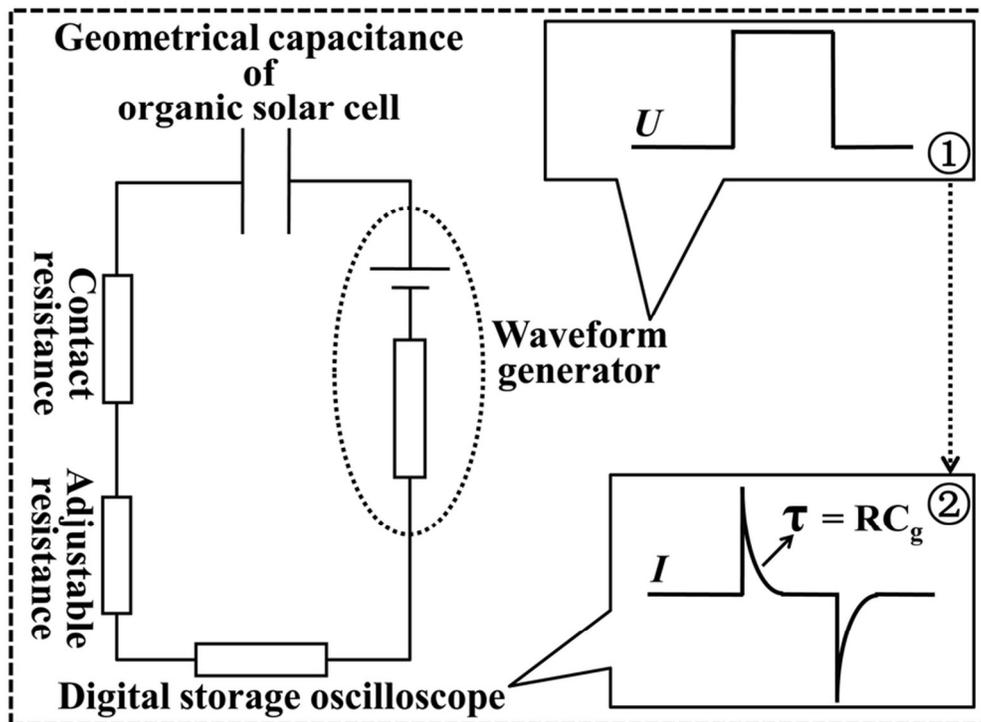
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