

Overcoming the fill-factor limit of organic solar cells

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Although organic solar cells have surpassed 20% power conversion efficiency, a persistent trade-off between open-circuit voltage and fill factor (FF) prevents them from closing the gap with inorganic technologies. Here we investigate this trade-off across a wide range of devices and identify an FF limit arising from field-dependent free-charge generation. This limit becomes more severe as voltage losses are minimized, thereby imposing an open-circuit voltage–FF trade-off. To quantitatively describe this limit, we develop an analytical model for field-dependent charge generation, revealing that the underlying cause is the field-sensitive charge-transfer process between excitons and charge-transfer states. This sensitivity originates from the field-induced charge-transfer state energy variations, mainly caused by the Stark effect. Guided by this physics-based model, we highlight that a long exciton lifetime is one of the practical and effective methods to overcome the FF limit.

Organic solar cells (OSCs) have recently achieved impressive power conversion efficiencies (PCEs) of >20% (refs. 1,2). These advances mainly benefit from the mitigated competition between open-circuit voltage (V_{oc}) and short-circuit current density (J_{sc})^{3–7} in OSCs based on non-fullerene acceptors (NFAs). Despite a thorough understanding and notable improvement in V_{oc} and J_{sc} , the third parameter, the fill factor (FF), which is equally important for PCE, is not well understood for OSCs. A critical question is whether there is a trade-off between the V_{oc} and FF^{8–10}.

The FF in solar cells is complex, as it is influenced by multiple processes and quasi-equilibrium states that vary with voltage. Some

technologies show an improved FF with a higher V_{oc} , attributed to decreased free-charge (FC) recombination losses, as observed in solution-processed perovskite solar cells^{11–13}. In silicon solar cells, by contrast, the FF– V_{oc} trade-off arises from the interplay between contact recombination and contact resistance^{14–16}. Therefore, the FF– V_{oc} relationship is a critical area of research across all solar cell technologies, especially for high-efficiency ones that are close to a Pareto frontier.

The trade-off between the FF and V_{oc} in OSCs was not a primary concern until the recent development of NFAs. During the fullerene period, the voltage loss was a major limiting factor for the PCE of

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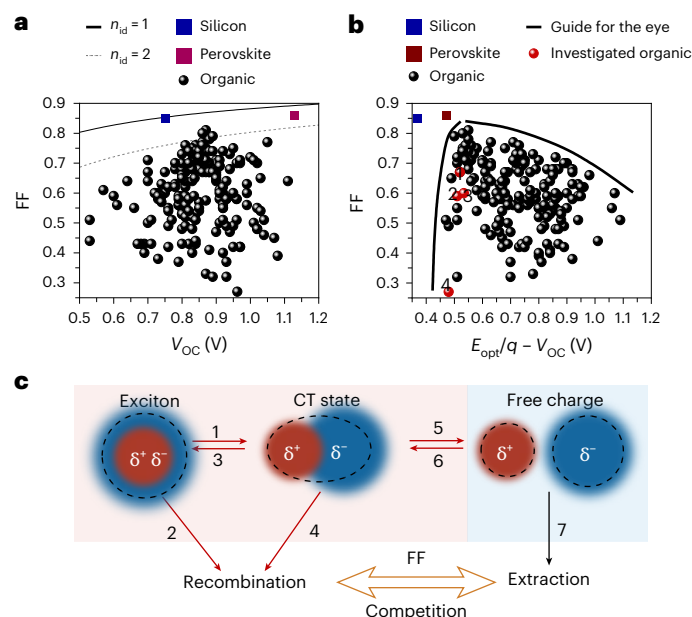


Fig. 1 | FF of OSCs. **a**, Intrinsic FF limit as a function of V_{OC} for single-junction solar cells is illustrated by the solid curve ($n_{id} = 1$) and the dashed curve ($n_{id} = 2$). The blue square represents a silicon solar cell. The red square represents a perovskite solar cell. The black dots represent OSCs. **b**, V_{OC} axis is converted to voltage losses for a bandgap-independent comparison. The red points (1–4) are PM6:Y11, PM6:Y1, PM6:IEICO-4F and PTO2:Y1, respectively, selected for investigation. **c**, Illustration of the interaction between charge carriers and Exs in OSCs. FC recombination via the formation of CT states (6) or Exs (3) followed by decay to the ground state ((2) and (4)) competes with charge separation following the path (1)–(5)–(7) that leads to photocurrent generation.

OSCs^{17,18}. The recent advancements in NFAs have substantially reduced the voltage losses, but the improved V_{OC} has introduced concerns regarding the FF^{8–10}. Whether there is an intrinsic FF limit in OSCs and how it correlates with voltage losses are fundamental questions for exploiting the full potential of organic semiconductors.

To understand the fundamental limits of the FF in OSCs, we systematically investigate its correlation with voltage losses across a diverse set of devices. For OSCs with V_{OC} losses ranging from 0.5 eV to 1.1 eV and FF values spanning from 0.27 to 0.80, we find that the FF of OSCs is not only dependent on the transport properties that determine the FF in conventional semiconductors but is also influenced by geminate recombination losses. Focusing on representative low-voltage-loss NFA systems, we probe FC generation and recombination under different voltages, and develop an analytical model that establishes field-dependent FC generation as a pivotal factor for FF losses. We attribute the field dependence to the field-induced variations of charge-transfer (CT) state energies caused by the Stark effect. Our findings converge on a critical conclusion: suppressing exciton (Ex) decay is the key to overcome the FF limitations and unlocking the full potential of OSCs.

FF and V_{OC}

For solar cells with ideal extraction and without resistive losses, the analytical relationship between the FF and the V_{OC} can be described by¹⁹

$$FF = \frac{v_{OC} - \ln[v_{OC} + 0.72]}{v_{OC} + 1} \quad (1)$$

and

$$v_{OC} = \frac{V_{OC}}{n_{id} k_B T}, \quad (2)$$

Table 1 | Comparison of FF

	PM6:Y11	PM6:Y1	PM6:IEICO-4F	PTO2:Y1
Reconstructed FF	0.66	0.61	0.56	0.58
Measured FF	0.67	0.59	0.60	0.27

Reconstructed FF is based on measured transport and recombination parameters using equations (5)–(7) given in Supplementary Fig. 3. The device based on PTO2:Y1, with low FC generation, shows a lower measured FF compared with the reconstructed FF.

where v_{OC} is the normalized V_{OC} and n_{id} is the ideality factor (Supplementary Note 1 provides the full parameter definitions). For $n_{id} = 1$ (Fig. 1a, solid curve), the FF increases with increasing V_{OC} , suggesting that a higher V_{OC} would yield a higher FF. We summarize the V_{OC} and FF data of diverse solar cells from the literature (Fig. 1a). Silicon and perovskite solar cells are close to the theoretical limit due to their efficient charge generation and collection. By contrast, OSCs show dispersed data, with V_{OC} from 0.52 to 1.11 V and FF from 0.27 to 0.80, reflecting their diverse optical, energetic and transport properties.

To compare devices with different optical gaps, we converted V_{OC} to voltage loss ($E_{opt}/q - V_{OC}$) and replotted the data in Fig. 1b (extraction of E_{opt} is shown in Supplementary Fig. 1 and the values are listed in Supplementary Data 1). Although the data remain scattered, two key trends emerge. First, the upper limit of the FF increases as voltage losses decrease, as indicated by the upper guide-to-the-eye curve. Second, at small voltage losses, a sharp drop in the FF values defines a left boundary, indicating competition between V_{OC} and FF in this region. We highlight the importance of understanding this left border, as it limits the FF (and hence the PCE) of low-voltage-loss OSCs. Overcoming the limitations that currently cause this border would make it possible to further enhance the PCE of state-of-the-art OSCs.

The upper limit can be explained by the interplay between charge recombination and extraction^{20,21}. The active-layer carrier lifetime and mobility can be combined into the electronic quality factor $Q = \mu_e \mu_h / k_2$, where μ_e and μ_h are the electron and hole mobilities, respectively, and k_2 is the bimolecular recombination coefficient^{20,22,23}. We calculated FF versus voltage loss at a fixed bandgap, external quantum efficiency (EQE) and thickness (Supplementary Fig. 2) and find that a large Q reproduces the upper limit, whereas smaller Q shifts the curve downwards as transport or recombination begins to dominate. Q , therefore, captures the scattering of the FF values from the perspective of fundamental semiconductor properties. Material- and device-level effects can be further unified by a dimensionless figure of merit α (ref. 24), but the resulting FF–voltage-loss curves do not explain the left border of the dataset (Supplementary Note 1 and Supplementary Fig. 3).

To understand the competition between FF and V_{OC} at the left border in Fig. 1b, we selected four donor–acceptor combinations (Fig. 1b, red dots) spanning V_{OC} losses of 0.53 to 0.49 V and FF from 0.65 to 0.27: PM6:Y11, PM6:Y1, PM6:IEICO-4F and PTO2:Y1 (Supplementary Fig. 4 shows the device characteristics). Single-carrier devices (Supplementary Fig. 5) show electron and hole mobilities of approximately $10^{-4} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ in all blends, consistent with previous reports in which these polymer donors and NFAs achieve PCE above 10% with FF around 70% (refs. 5,25,26). The low-voltage-loss OSC blends in this work do not have particularly low mobilities. We then obtained the recombination coefficients via the time-delayed collection field (TDCF) method and find that the bimolecular recombination coefficient is high in the low-voltage-loss cases (Supplementary Table 1), in agreement with previous reports²⁷. The high V_{OC} in these cases can be attributed to the relatively high emission quantum yield of bimolecular recombination^{3,4}. Using these parameters, we calculated the theoretical FF using equations accompanying Supplementary Fig. 3. As shown in Table 1, the reconstructed FF of PTO2:Y1 is 0.58, representing the FF expected if the device was limited only by transport and bimolecular recombination. The marked discrepancy with the measured FF of 0.27 indicates that other factors not included in Q and α affect the FF.

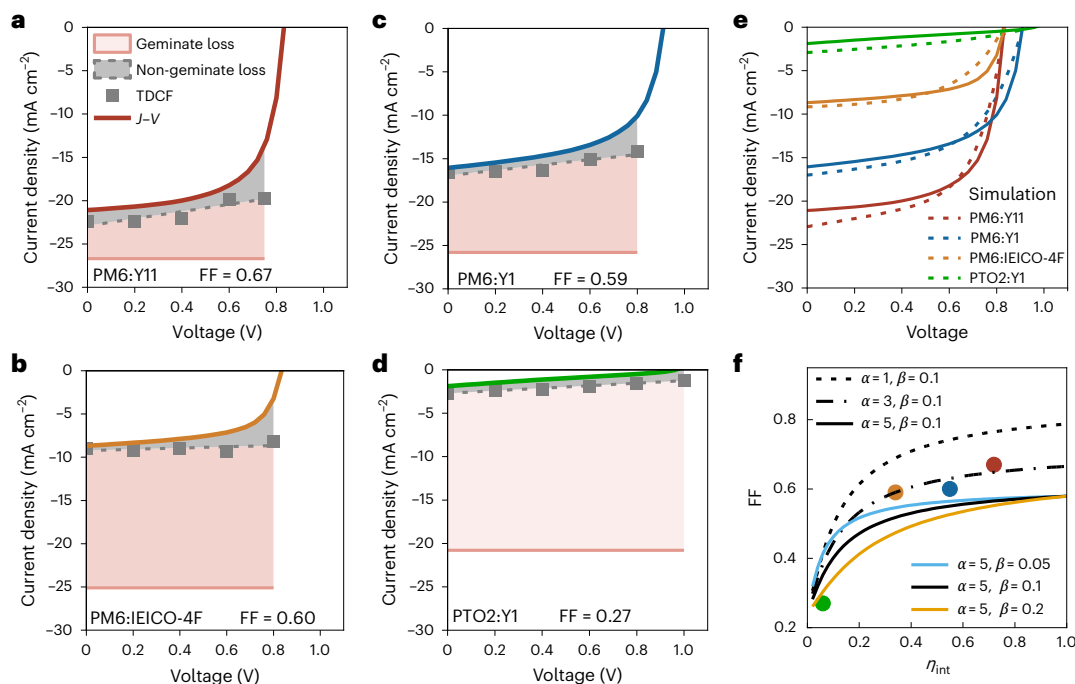


Fig. 2 | Field-dependent charge generation and its influence on FF. a–d, J – V characteristics and charge-generation analysis of PM6:Y11 (a), PM6:IEICO-4F (b), PM6:Y1 (c) and PTO2:Y1 (d). The solid curves (top) are the J – V data measured under 1-sun conditions. The grey squares represent the dissociated charges measured via TDCF. The dashed grey lines represent the charge generation current densities (J_G) obtained by linearly fitting the dissociated charges. The red lines represent photons absorbed by the device (J_{Abs}). The grey areas represent the non-geminate recombination losses. The red areas represent geminate

recombination losses. **e**, Solid curves are the measured J – V curves corresponding to the four examples. The dashed curves are simulated J – V curves that show the influence of field dependence on FF. **f**, FF limit as a function of internal FC generation efficiency at the open circuit is simulated with different parameters describing losses due to charge transport (α) and field-dependent FC generation (β). From left to right: the solid-colored circles denote PTO2:Y1, PM6:IEICO-4F, PM6:Y1 and PM6:Y11.

FF and FC generation

OSCs are based on excitonic materials in which strongly bound Exs generate FCs through intermediate donor–acceptor CT states (Fig. 1c). Geminate recombination of Exs and CT states can severely limit FC generation; in the following, we show that its field dependence imposes an FF limit in OSCs.

We performed TDCF measurements to probe FC generation in the selected systems. By adjusting the delay time and applying a large reverse bias, we measured the FCs generated at a set of pre-biases. Unlike the photocurrent, the TDCF measurements do not suffer from non-geminate losses, since photogenerated free carriers are extracted rapidly by the large reverse bias (Supplementary Fig. 6). The gap between absorbed photons (from the ellipsometry data in Supplementary Fig. 7) and the generated FCs is the loss due to geminate recombination, whereas the gap between FCs and the photocurrent reflects transport and recombination losses captured by α . The TDCF results are shown in Fig. 2a–d with pre-bias swept from 0 to near V_{oc} . The dashed grey lines mark the FC current density J_G , and the reddish area below them indicates geminate recombination. In all four systems, J_G varies with bias, showing that geminate recombination affects the FF.

To describe the influence of FC generation on the FF, we use the following analytical relationship between current density and voltage in OSCs²⁴:

$$J(V) = J_G(V) \left\{ \exp \left[\frac{q}{k_B T (1 + \alpha)} (V - V_{oc}) \right] - 1 \right\}, \quad (3)$$

where $J_G(V)$ is the FC generation current density and α is the figure of merit introduced earlier (Supplementary Note 1 provides the full parameter list). Parameterizing the field-dependent FC generation yields

$$J_G(V) = J_{Abs} \eta_{int} \left[1 + \left(\frac{1}{\eta_{int}} - 1 \right) \beta |V - V_{oc}| \right], \quad (4)$$

where η_{int} is the internal FC generation efficiency at open circuit and β (V⁻¹) is the field dependence coefficient quantifying the influence of the electric field on the geminate recombination term $(1/\eta_{int} - 1)$. Both η_{int} and β can be deduced from TDCF. Simulated current density–voltage (J – V) curves for PM6:Y11, PM6:Y1, PM6:IEICO-4F and PTO2:Y1 (Fig. 2e; Supplementary Table 2 lists the parameters) reproduce the experimental data shown in Fig. 2a–d, indicating that equation (4) captures the key parameters that determine the FF. By incorporating field-dependent FC generation, the model bridges the gap between the transport-limited prediction and the experimental reality.

We varied the model parameters (Supplementary Fig. 8) and summarize the effects on FF in Fig. 2f. PM6:Y11, PM6:Y1 and PM6:IEICO-4F fall in the regime in which transport regulates the FF. By contrast, PTO2:Y1 ($\eta_{int} = 0.06$, $\beta = 0.09$ V⁻¹) is dominated by field-dependent FC generation. The drop in FF of PTO2:Y1, thus, presents an FF limit at the left border of Fig. 1b. Shifting this boundary leftwards, that is, achieving a high FF at low voltage losses for enhanced PCE, requires understanding the physical nature of field-dependent FC generation, which we elaborate in the next section.

FF limit and geminate pairs

To determine whether the Ex-to-CT or CT-to-FC transition limits the FF, we applied pump-push photocurrent (PPPC) measurements (Supplementary Fig. 9), which used an infrared push pulse to selectively probe the bound-state dynamics^{28,29}. The geminate pairs in our blends have similar decays and binding energies to species in the neat-acceptor films, indicating that the dissociation of acceptor Exs, assisted by the built-in voltage, is the key step affecting the FF.

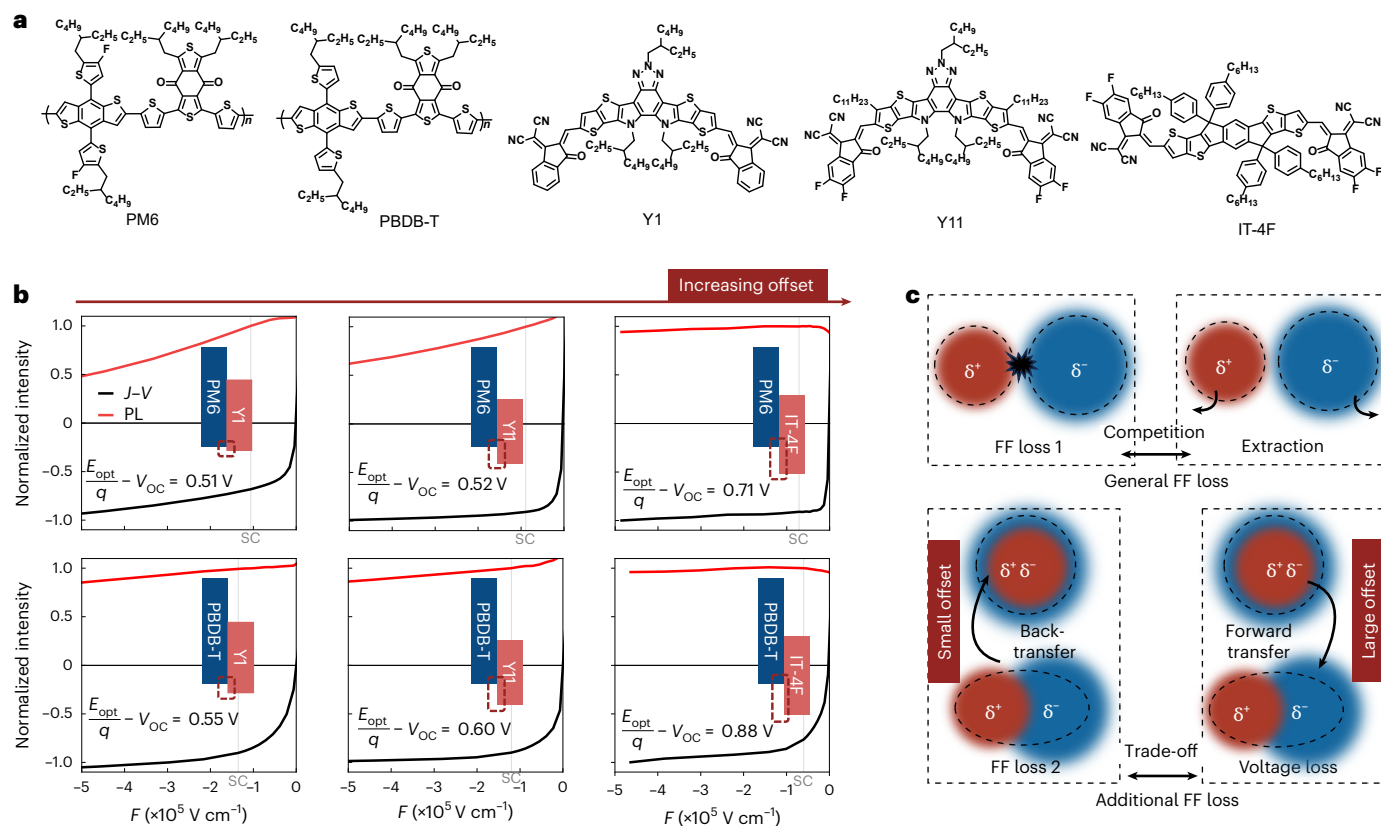


Fig. 3 | Bias-dependent PL and FF loss. a, Molecular structures of donors (PM6 and PBDB-T) and NFAs (Y1, Y11 and IT-4F). **b**, The red curves are the PL intensities obtained via integration over the acceptor emission peaks, normalized to the values at short circuit (SC). The black curves are $J-V$ values that are simultaneously measured with the PL and normalized to the maximum absolute values. The pink and blue rectangles are schematic of the relative frontier orbital energy levels of material combinations. The offset indicates the energetic offset between the

low-energy Exs and the CT state. **c**, Schematic of the loss mechanisms. Top: In general, FF loss 1 (non-geminate) arises from the competition between the recombination and extraction of FCs. Bottom: In organic photovoltaics, a specific trade-off exists governed by the energetic offset. Small offsets (left) promote back-transfer and field-dependent generation, resulting in FF loss 2. Large offsets (right) facilitate efficient separation but cause additional voltage losses.

In addition, we utilized bias-dependent photoluminescence (PL) measurements to monitor the Ex population under an applied voltage. Different from inorganic solar cells in which PL is easily quenched by charge extraction (Supplementary Fig. 10), the PL of OSCs remains strong even under SC or reverse bias because it primarily originates from geminate recombination^{30,31}. Combined with the PPPC binding energies (Supplementary Fig. 9) and the lack of a notable CT redshift in the blends (Supplementary Fig. 11), the PL intensity serves as a reliable indicator of Ex population and decay dynamics.

To reveal the factors that influence the FF, we selected two representative polymer donors and three NFAs to obtain blends with different energetic offsets (Fig. 3a) and compared the field dependence of PL intensity along with their $J-V$ characteristics (Fig. 3b). We find that a key factor affecting FF is the energetic offset between the local Ex and the CT state. Taking PM6:Y1, PM6:Y11 and PM6:IT-4F as examples, when the energetic offset increases, the field dependence of PL becomes weaker, and the FF gets higher. PM6:IT-4F (high offset) shows almost no field dependence in PL, consistent with its high FF. PM6:Y1 (low offset) and PM6:Y11 (medium offset) show obvious PL quenching under an increasing field. In PM6:Y1, the current enhancement continues in the reverse-bias region (-1 to $-5 \times 10^5 \text{ V cm}^{-1}$) along with the PL quench, because the PL is converted into the extracted current. PBDB-T-based blends with high offsets show weak PL field dependence but lower overall FF due to non-geminate losses. As illustrated in Fig. 3c, this creates a trade-off: small offsets lead to field-dependent generation and FF losses, whereas large offsets avoid this but cause voltage losses.

On the basis of PPPC and field-dependent PL spectroscopy, we believe that the electric field mainly influences the transition between Ex and CT states. Despite many discussions on field-assisted CT dissociation^{32–34} (Fig. 1c, process (5)), transient absorption studies have shown that the FC signal appears simultaneously with CT^{35,36}, indicating an efficient CT-to-FC transition and reducing the possibility of CT dissociation as the field-sensitive step. Field-assisted direct Ex separation in neat NFA domains is also unlikely, as the PL of pristine films is field independent within our investigation range (Supplementary Fig. 11i,j). The Ex-to-CT transition (probably including FC formation via long-range CT³⁷) is directly linked to the energetic offset^{9,30} and is, therefore, the most probable candidate for being affected by the electric field, responsible for the field-dependent FC generation that limits the FF in OSCs.

Recent studies have highlighted the role of delocalized excited states (the so-called i-Ex) in Y-series NFAs, which may facilitate charge generation^{35,38}. The term ‘Ex’ in our definition encompasses these delocalized states. Although delocalization aids in reducing the binding energy, our observation of field-independent PL in pristine NFA films (Supplementary Fig. 11) suggests that i-Ex states alone cannot efficiently generate FCs without the donor–acceptor interface. Therefore, the Ex-to-CT transition (whether the Ex is localized or delocalized) remains the field-sensitive bottleneck.

Physical framework for overcoming the FF limit

To further quantitatively elucidate the field-dependent Ex dissociation, we develop an analytical model. The transition rates are described using the Marcus theory^{39,40}, and the electrostatic potential is incorporated

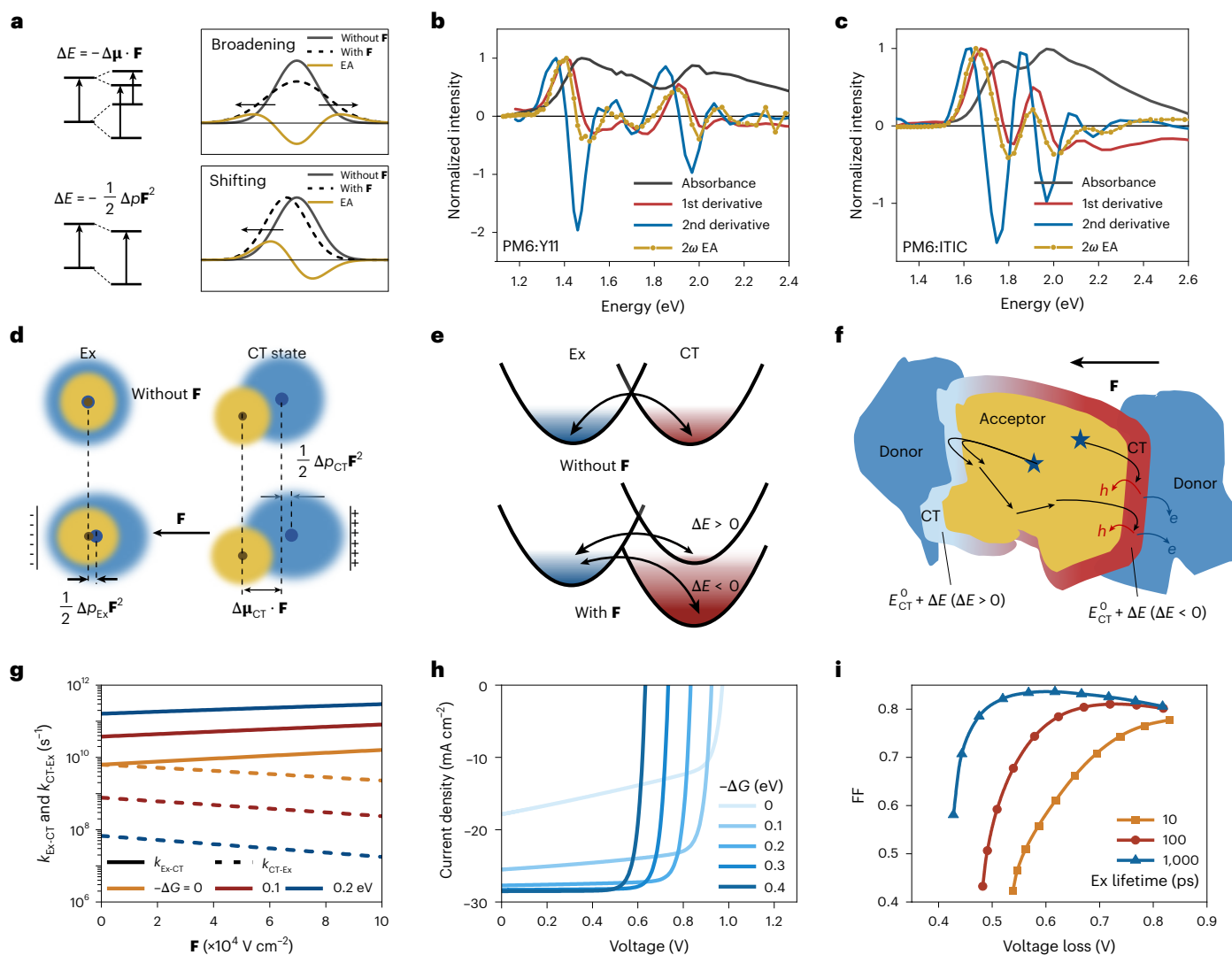


Fig. 4 | Electric-field effects on state energies and charge carrier generation.

a, Absorption change induced by an electric field. Spectral broadening (top graph) due to a dipole moment difference in an isotropic sample causes an EA shape that is approximately the second derivative of the absorption spectrum. Spectral shift (bottom graph) due to a polarizability difference leads to the EA shape, which is approximately the first derivative of the absorption spectrum. **b, c**, EA spectra (normalized $\Delta T/T$) of PM6:Y11 (**b**) and PM6:ITIC (**c**), plotted alongside their absorbance and the first and second derivatives of absorbance. **d**, The top graphs illustrate how the electron and hole charge distributions change as an Ex forms an interfacial CT state. The bottom graphs show the same process in the presence of an external electric field. The distances indicated by arrows represent field-induced energy shifts arising from changes in the dipole moment and polarizability. **e**, CT state formation rate as described by Marcus theory. **f**, Ex dynamics in a blend under an applied electric field. The field either

raises or lowers the CT state energy at acceptor–domain boundaries, depending on the boundary orientation. Exs diffusing against the electric field encounter boundaries at which the CT state energy is reduced; they readily form CT states that subsequently split into electron–hole pairs. Exs diffusing along the electric field encounter boundaries at which the CT state energy is increased; they either scatter from the heterojunction or transiently form CT states. By subsequent random diffusion, they can reach a boundary with reduced CT state energy, where they form CT states and dissociate into electron–hole pairs. **g**, Simulated transition rate coefficient between the Ex and CT states depending on the electric field based on the first-order Stark effect. **h**, Simulated $J-V$ curves with an Ex lifetime of 1,000 ps. $\Delta G = \Delta E_{CT-Ex} = E_{CT} - E_{Ex}$. **i**, Simulated trade-off between the FF and V_{OC} at different Ex lifetimes. Dots with the same colour have energy offsets varying from 0 eV to 0.45 eV.

using perturbation theory to account for the first- and second-order Stark effects. The energy of electronic states is shifted by the electric field through their static dipole moments (first order) and electronic polarizability (second order), which can alter the electron transfer between the Ex and CT states. The field-induced energy difference between the excited and ground states can be described as

$$\Delta E = -\Delta\mu F - \frac{1}{2}\Delta p F^2, \quad (5)$$

where $\Delta\mu$ is the difference in dipole moment between the excited and ground states of the considered excitation, Δp is the corresponding difference in polarizability and F is the electric field.

Incorporating this into the Marcus rate equation yields a field-dependent forward rate:

$$k_{Ex-CT}(F) \propto \exp\left(-\frac{(\lambda + \Delta G_{CT-Ex} + \Delta E_{CT}(F) - \Delta E_{Ex}(F))^2}{4\lambda k_B T}\right), \quad (6)$$

where λ is the reorganization energy³⁰ and ΔG_{CT-Ex} is the zero-field driving force for the Ex-to-CT transition (Supplementary Note 2). Importantly, the electric field can either increase or decrease the activation barrier for the Ex-CT transition.

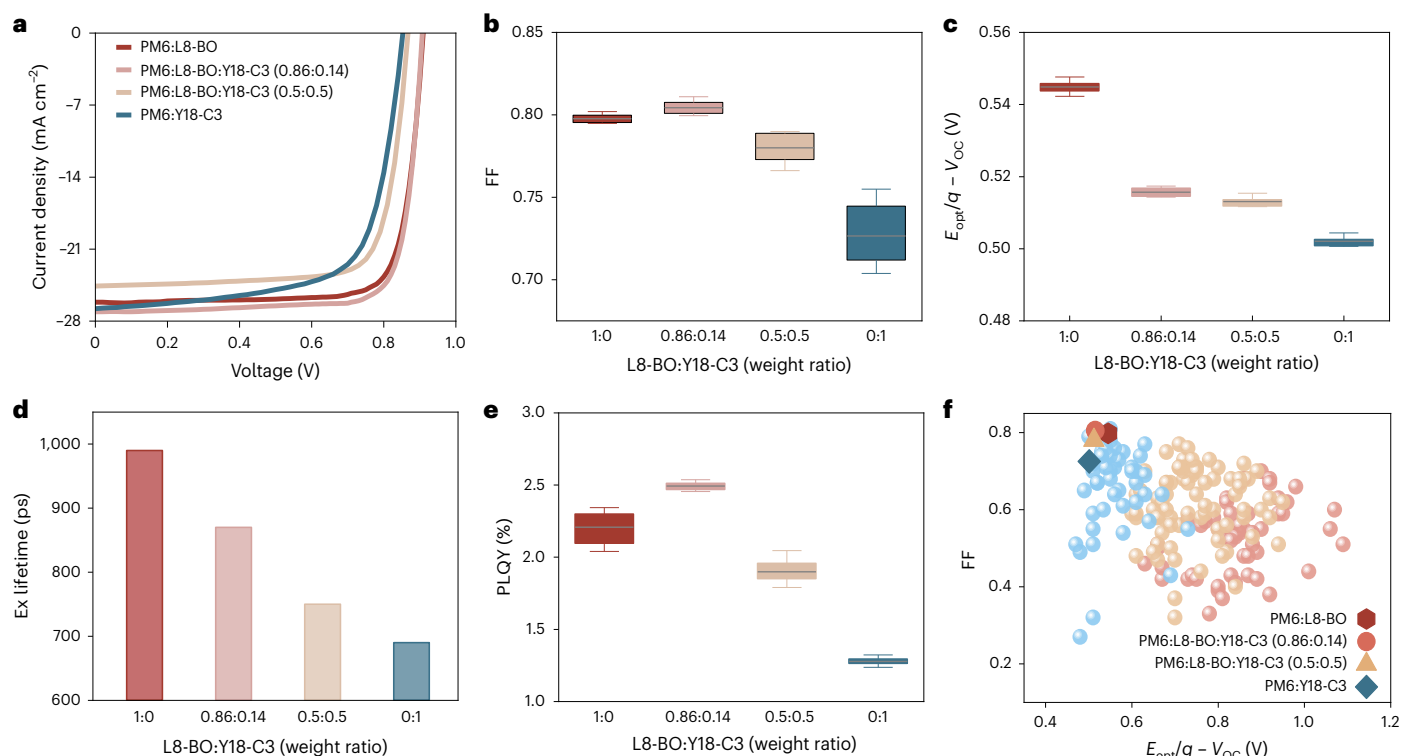


Fig. 5 | Demonstration of balanced FF and V_{oc} in ternary blends. **a**, J - V characteristics of OSCs. **b**, **c**, FF (**b**) and voltage losses (**c**) of OSCs based on PM6 and different acceptor ratios ($n = 8$ independent devices per ratio). **d**, **e**, Ex lifetime (**d**) and PLQY (**e**) of films based on acceptors with different blending ratios (two independent films and six measurements per ratio). For the box charts shown

in **b**, **c** and **e**, the box bounds indicate 25%–75% interquartile range, the middle line indicates a mean value and the whiskers mean $1.5 \times$ the interquartile range. **f**, Blue dots (left) are based on NFA reports after 2019, when Y6 was developed. Yellow dots (middle) are NFAs reported between 2015 and 2019, when ITIC and its derivatives dominated the field. Red dots (right) are NFA reports before 2015.

Figure 4a graphically demonstrates two distinct electroabsorption (EA) spectra showing how the electric field affects electronic transitions via the Stark effect. For the first-order effect, the energy levels split in disordered systems with randomly oriented molecules—the field raises the transition energy for molecules aligned against it and lowers it for those aligned with it; the corresponding EA signal resembles the second derivative of the absorption band. For the second-order effect, the energy levels shift with F^2 , and the corresponding EA signal resembles the first derivative. Figure 4b,c shows the second-harmonic EA of PM6:Y11 and PM6:ITIC alongside the absorbance and its first and second derivatives. The EA position and relative peak ratio in both blends lie between the first and second derivatives, whereas the first derivative dominates in the Ex optical transition. The CTEA is weak due to the indistinguishable CT states in small-offset systems.

We subsequently calculated the energy shifts. For the first-order Stark effect, the Ex dipole is ignored due to its small electron–hole separation, whereas the separation in interfacial CT states in NFA blends is ~ 3.5 nm (refs. 41,42). With an internal field of -1 V/100 nm (10^5 V cm^{-1}), this yields a linear Stark shift of ~ 35 meV, on the order of thermal energy and comparable with the donor–acceptor highest occupied molecular orbital offset. For the second-order effect, polarizability values for Exs in donors and NFAs range from 100 to 1,000 Å³, giving shifts of only 0.0035 to 0.035 meV, with the CT shift being similarly small^{43,44}.

Figure 4d visually explains the changes in Ex and CT state energies under an applied electric field. The second-order effect simply shifts the CT energy down, whereas the first-order term decreases the energy of CT states oriented against the field and increases that of states oriented along it (Fig. 4e). As shown in Fig. 4f, these field-modified energies enhance the average formation rate of CT states, which preferentially form at boundaries oriented against the field, establishing an optimal energetic landscape for dissociation into free carriers.

Next, we calculate the CT formation rate coefficients $k_{\text{Ex-CT}}(F)$ and $k_{\text{CT-Ex}}(F)$ as functions of the electric field, focusing on the first-order Stark effect for simplicity. Because the local Ex is more localized, we will not consider $\Delta E_{\text{Ex}}(F)$ in the following. Figure 4g shows the results with the dipole aligned along the field. Integrating these rates into a device-level drift–diffusion simulation (Supplementary Note 3) reproduces the experimental J - V curves with reduced FF at decreasing energy offsets (Supplementary Fig. 12). Because Ex dissociation competes with Ex decay, a slow Ex decay can mitigate this field dependence: as shown in Fig. 4h, a slower Ex decay rate preserves a high FF even at small voltage losses, revealing a pathway to simultaneously improve FF and V_{oc} by increasing the Ex lifetime.

To provide quantitative guidance for future material design, we further simulated the theoretical efficiency limits of single-junction OSCs as a function of Ex lifetime (Fig. 4i). By assuming minimized non-radiative voltage losses and optimized charge transport, our model predicts that extending the Ex lifetime beyond 1 ns at the current state-of-the-art energetic landscape sufficiently mitigates the FF limit to enable FFs exceeding 82%.

To validate the feasibility of our strategy, we constructed a guest–host system as a proof of concept (Fig. 5a–c and Supplementary Figs. 13–15). PM6:L8-BO, one of the most effective binary systems, delivers FF = 79.5% owing to the long L8-BO Ex lifetime (990 ps), but its voltage loss (0.545 V) leaves room for improvement. We introduced Y18-C3, an acceptor with a smaller energy offset, as the third component. The PM6:Y18-C3 binary shows a small voltage loss (0.502 V) but a low FF (68.8%) due to the short Y18-C3 Ex lifetime (690 ps). The FF and voltage loss show tunability when Y18-C3 is mixed with L8-BO. At the optimal 0.86:0.14 ratio, the ternary simultaneously achieves FF = 81.1% and voltage loss of 0.516 V, yielding PCE = 20.1%. This improvement correlates with Ex dynamics and PL quantum yield (PLQY) as L8-BO prolongs the

Ex lifetime from 690 ps in Y18-C3 to 750 ps and 870 ps in the mixtures (0.5:0.5 and 0.86:0.14, respectively), and boosts the PLQY (Fig. 5d,e). To rule out morphological origins, we characterized the blends using atomic force microscopy and grazing-incidence wide-angle X-ray scattering (Supplementary Figs. 16 and 17). The ternary films exhibit surface roughness and crystal packing parameters highly comparable with the high-performance binary control, confirming that Ex dynamics rather than phase separation drive the improved FF. The general applicability of the guest–host strategy is validated in multiple ternary systems with different donor polymers, varied guest–host acceptor pairs and fullerene-based blends (Supplementary Figs. 18–22).

The finding that suppressing Ex decay is crucial for high FF aligns with the developmental trajectory of OSCs (Fig. 5f): post-2019 Y6-based NFAs (blue) show higher FF and lower voltage loss than 2015–2019 ITIC-based reports (yellow) and pre-2015 reports (red), consistent with the longer Ex lifetimes of Y6 and L8-BO compared with the ITIC family in our time-correlated single-photon counting (TCSPC) measurements (Supplementary Figs. 23 and 24 and Supplementary Table 3). Combining our finding that extending Ex lifetime is essential to overcome the FF limit with the established requirement of high PLQY for low voltage loss^{3,4}, we conclude that suppressing non-radiative Ex decay is the key to unlocking the full potential of OSCs.

Conclusion

Our work establishes a comprehensive framework for understanding the V_{oc} –FF trade-off in OSCs. The framework reveals an FF limit imposed by field-dependent CT competing with Ex decay, via quantitatively correlating the FF and voltage loss with fundamental physical parameters. In particular, experimental and theoretical results show that enhancing the Ex lifetime is a key strategy for mitigating the FF limit and increasing the efficiency of OSCs. Using a guest–host strategy to manipulate both Ex decay and voltage loss, we demonstrate OSC devices that simultaneously achieve high FF and high performance. Beyond this proof of concept, our quantitative FF model offers general guidance for material design and device engineering, providing new opportunities to overcome long-standing efficiency bottlenecks in OSCs.

Online content

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Methods

Material preparation

The electron transport material (ZnO N-10, nanoparticle solution) was purchased from Avantama AG and used without additional treatment. The hole transport material (molybdenum oxide, powder) was purchased from Sigma-Aldrich and used without additional treatment. Among the active-layer materials, PM6, PTO2 and IEICO-4F were purchased from Solar Materials. Y1 and Y11 were synthesized at Central South University.

Fabrication of photovoltaic devices

Prepatterned indium tin oxide substrates were cleaned with detergent followed by two 20-min ultrasonic steps in acetone and isopropanol. Subsequently, a 10-min ultraviolet–ozone treatment was applied. A layer of zinc oxide (N-10, Avantama) of approximately 30-nm thickness was spin coated in air at 3,600 rpm and annealed at 120 °C for 5 min, after which the samples were moved into a glovebox. The active layer was spin coated from the solution, and the rotation speed was adjusted to yield an active-layer thickness of around 100 nm. Chloroform was used as the solvent for PM6:Y11, PM6:Y1 and PTO2:Y1. Chlorobenzene was used for PM6:IEICO-4F. The ratio and total concentration were 1:1.2 and 18 mg ml⁻¹ for PM6:Y11, 1:1 and 16 mg ml⁻¹ for PM6:Y1 and PTO2:Y1, and 1:1 and 20 mg ml⁻¹ for PM6:IEICO-4F. The solution was kept on a hotplate at 60 °C for 12 h before spin coating and was kept on a plate during spin coating. Immediately after spin coating, an annealing step at 100 °C for 10 min was applied. The substrate was then placed on a mask and transferred to a vacuum chamber. Molybdenum oxide (12 nm) and Ag (200 nm) were thermally evaporated in a vacuum of approximately 10⁻⁶ mbar. PM6:L8-BO and PM6:L8-BO:Y18-C3 were fabricated with a conventional structure of indium tin oxide/2PACz/active layer/PNDIT-F3N/Ag. A monolayer of 2PACz (0.27 mg ml⁻¹ in ethanol) was first deposited onto the indium-tin-oxide-coated substrates at 3,000 rpm for 30 s, followed by thermal annealing at 100 °C for 10 min in a nitrogen atmosphere within the glovebox. The blend of PM6 and acceptors was dissolved in chloroform containing 10 mg ml⁻¹ of DCBB as the additive, and the active-layer solution was prepared at a total concentration of 16.1 mg ml⁻¹, stirred for 2 h and then spin coated onto the 2PACz layer. The thermal annealing treatment of the active layer was performed at 100 °C for 10 min. Subsequently, a thin layer of PNDIT-F3N (1 mg ml⁻¹ in a methanol solution with 0.5 vol% acetic acid) was spin coated onto the active layer at the rate of 3,000 rpm for 30 s before the deposition of 150-nm Ag.

J–V characterization

Devices were encapsulated in a glovebox and measured in air. The active area of the tested solar cell was 4 mm². The J–V curves (measured in the forward direction, that is, from negative to positive bias, with a scan step of 0.04 V) were collected by using a Keithley 2400 source meter under AM1.5 illumination provided by a solar simulator (LSH-7320 ABA LED solar simulator) with an intensity of 1,000 W m⁻² after spectral mismatch correction. The light intensity for the J–V measurements was calibrated using a reference Si cell (VLSI standards SN 10510-0524 certified by the National Renewable Energy Laboratory).

Photovoltaic EQE measurements

EQE measurements were conducted using an integrated system (QE-R3011). The system was calibrated using a silicon reference detector.

PL and field-dependent PL measurements

Spectrally resolved PL measurements were performed on an Andor Shamrock 303i spectrograph equipped with an Andor Newton electron-multiplying charge-coupled device (DU970N-UVB). During the measurement, the charge-coupled device detector was cooled to –45 °C. The wavelength of the system was calibrated by using a mercury lamp. The intensity of the spectra was calibrated using a

standard halogen lamp (AvaLight-HAL-S-Mini, Avantes). The PL was excited by a Thorlabs collimated laser-diode-pumped DPSS laser module (CPS532), and a 550-nm long-pass filter was used to protect the detector. The PL excitation and detection were performed using an objective, a 552-nm long-pass dichroic mirror placed at 45°, and optical fibres. The samples were positioned where the light spot area was larger than the device pixel size. The laser intensity was controlled by a neutral-density filter wheel to ensure that the photocurrent was equal to that of 1 sun. A Keithley 2400 source meter was connected to the photovoltaic devices to apply a voltage bias and record the current response.

EQE_{EL} measurement

The EQE_{EL} value was recorded using a custom-built system with a Hamamatsu silicon photodiode 1010B. A Keithley 2400 meter was used for supplying bias voltages and recording the injected current, and a Keithley 485 device was used for collecting the photocurrent generated from the emitted photons of the samples.

TDCF measurement

TDCF investigations were performed using an Agilent Technologies DSO5054A oscilloscope using a 50-Ω input resistor and a Tektronix AFG3101 function generator. The samples were excited by radiation from the optical parametric amplifier TOPAS-C (LIGHT CONVERSION) pumped by a femtosecond Ti:sapphire Integra-C laser (Quantronix) generating 130-fs-duration pulses at a 430-Hz repetition rate. A linear optical parametric amplifier TOPAS-C was used to generate an excitation pulse emitting at 515 nm.

Ellipsometry measurement and analysis

Ellipsometry was performed using a Mueller Matrix Ellipsometer (RC2, J.A. Woollam Co.). CompleteEASE was used to globally fit the Mueller matrix data with the B-spline and general oscillator models for the optical properties of the samples. The refractive index and extinction coefficient were used for transfer matrix optical modelling (<https://github.com/erichoke/Stanford/tree/master>).

TCSPC measurement

TCSPC measurements were performed using a system from Edinburgh Instruments with a microchannel plate photomultiplier tube (Hamamatsu). An excitation pulse laser at 405 nm was generated using a pulsed picosecond diode (Hamamatsu).

PPPC measurements

Femtosecond pulses (800 nm, 35 fs) were generated using a 4-kHz Ti:sapphire regenerative amplifier (Astrella, Coherent). These pulses were routed onto two optical parametric amplifiers (TOPAS Prime, Coherent). The 1,400-nm output from one TOPAS passed through a frequency-doubling barium borate crystal to generate the pump at 700 nm. The pump pulse was then directed onto a mechanical delay stage to vary the time delay between the pump and push beams. The 2,000-nm output from the other TOPAS served as the push. The push was mechanically modulated at 1.1 kHz. Both pump and push pulses were aligned to a single spot on the device pixel. During the measurements, the devices were connected to a lock-in amplifier (MFLI, Zurich Instruments) and measured under short-circuit conditions. The reference current *I* was measured at a pump frequency of 4 kHz, and the push-induced current ΔI was measured at a push frequency of 1.1 kHz. The devices were placed in a cryostat (HFS600E-PB4, Linkam) with a liquid-nitrogen cooling module (LNP96, Linkam) to control the temperature.

EA spectroscopy

EA spectroscopy was conducted to measure the EA signals. The setup is equipped with a light source (Xenon Arc Lamp 1,000 W, Newport),

monochromator (Zolix), optical chopper (Thorlabs), calibrated silicon and germanium photodetectors (Thorlabs), low-noise current preamplifier (Stanford Research Systems, SR570), lock-in amplifier (Stanford Research Systems, SR830) and a function generator (SRS DS360). A monochromatic beam is transmitted through the semi-transparent device (not encapsulated) and detected by using the silicon and germanium photodetectors. During the measurement, the samples were housed inside a vacuum cryostat (Oxford Instruments) at a base pressure of around 10^{-5} – 10^{-6} torr. Although measuring the device transmittance, the optical chopper provides a synchronous reference signal (190 Hz) to the lock-in amplifier. The transmitted light intensity (I) was detected by a silicon photodetector, which generated a current signal and fed it into the lock-in amplifier. This measured transmittance (T) was used to calculate the derivatives. To measure the electric-field-induced change in transmittance (ΔT), a function generator was used to modulate the internal electric field in the organic layer by superimposing a sinusoidal voltage at a frequency of 1 kHz on a negative d.c. voltage. The applied electric field is around 10^5 V cm $^{-1}$. The modulated signal from the detector was amplified using the current preamplifier by choosing a suitable gain or sensitivity. The lock-in amplifier was connected to demodulate the signal, phase referenced to the function generator at the different harmonics of the modulation fundamental frequency. The harmonic number in the lock-in amplifier can be adjusted to measure the first- and second-harmonic EA signals. The measured ΔT needs to be scaled by a factor of $\sqrt{2}$ to convert the root-mean-square value (r.m.s.) to the peak value.

The equations governing the EA change for the second harmonic are given by

$$\Delta A_{2\omega}(E) = - \left[\frac{1}{4} \frac{\partial A(E)}{\partial E} (\Delta p) + \frac{1}{12} \frac{\partial^2 A(E)}{\partial E^2} (\Delta \mu)^2 \right] \frac{V_{a.c.}^2}{d^2} \sin \left(2\omega t + \frac{\pi}{2} \right), \quad (7)$$

$$\frac{\Delta T}{T} = 10^{-\Delta A} - 1 \approx -\ln[10]\Delta A \approx -2.3\Delta A, \text{ if } \Delta A \approx 0, \quad (8)$$

where A is the absorbance, ω is the frequency of the applied voltage, ΔA is the absorbance difference (after minus before), E is the energy of photoexcitation, Δp is the polarizability difference, $\Delta \mu$ is the electric dipole moment difference, $V_{a.c.}$ is the voltage amplitude of alternating current and $V_{d.c.}$ is the voltage amplitude of direct current. Details of the derivation of equation (7) can be found in a previous report⁴⁵.

Data availability

All data supporting the findings of this study are available within the article and its Supplementary Information. Source data are available via Zenodo at <https://doi.org/10.5281/zenodo.20082078> (ref. 46). Additional data are available from the corresponding authors upon reasonable request.

Code availability

The drift–diffusion simulation and analytical model code used in this study is available via GitHub at https://github.com/HuotianZhang/DriftFusionOPV_FieldDependent, or is available from the corresponding authors upon reasonable request.

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Author contributions

H.Z. and F.G. conceived of the ideas. H.Z. designed the project, prepared the devices and samples and performed the J – V characterizations, photovoltaic EQE measurements, PL and field-dependent PL measurements, EQE_{EL} measurements, ellipsometry measurements and analysis. J.Y. and W.L. synthesized the NFAs under the supervision of Y.Z. T.W. and H.Z. performed the PPPC measurements under the supervision of A.A.B. Y.N. fabricated the ternary-related devices under the supervision of J.Y. R.J. performed the TDCF measurements under the supervision of V.G. Y. Liu performed the TCSPC measurements under the supervision of X.Z. N.J. and H.Z. performed the TCSPC verification and analysis. Y. Li prepared the samples for PPPC measurement. N.T. performed the initial CT calculation. S.O. performed the EA measurements under the supervision of S.-W.T. M.S.S. verified the EA under the supervision of S.S. H.Z. developed the theoretical model with the help of T.K., V.C., D.N., N.T. and M.P. D.N. supervised the simulation. W.L., V.G., J.Y., T.K., F.G. and H.Z. edited the figures. F.G. supervised the project. H.Z. and F.G. wrote the paper. All authors discussed the results and commented on the final paper.

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Competing interests

The authors declare no competing interests.

Additional information

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