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An Analysis of the Factors Determining the Efficiency of Photocurrent Generation in Polymer:Nonfullerene Acceptor Solar Cells

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Please submit a plain text version of your cover letter here.	Department of Chemistry Imperial College London London SW7 2AZ, UK Tel: +44 (0) 2075 949551 j.durrant@imperial.ac.uk http://www.imperial.ac.uk/people/j.durrant Prof. James Durrant, Professor of Photochemistry London, 18 May 2018. Dr. Till von Graberg Editor-in-Chief Advanced Energy Materials Weinheim Germany Dear Dr. Till von Graberg, Please consider the manuscript entitled "An Analysis of the Factors Determining the Efficiency of Photocurrent Generation in Polymer:Nonfullerene Acceptor Solar Cells", co-authored by Hyojung Cha, Ching Hong Tan, Jiaying Wu, Yifan Dong, Weimin Zhang, Hu Chen, Sridhar Rajaram, K. S. Narayan, Iain McCulloch, James R. Durrant, for publication in Advanced Energy Materials. The co-authors have all agreed to submission. The work has not been submitted for publication elsewhere. The non-fullerene electron acceptors (NFAs) have attractive interest to replace fullerene derivatives, PC61BM and PC71BM for organic bulk heterojunction solar cells. Studies of the application of NFAs to organic solar cells to date have largely focused on efficiency optimization, yielding in particular impressive open circuit voltage with

higher the lowest unoccupied molecular orbital (LUMO) levels as well as suppression of non-radiative recombination losses, albeit typically with more modest device photocurrents, although NFA typically exhibits superior light harvesting properties compared to fullerene acceptors. In this report, we focus upon these issues, and in particular, how the energy offset driving interfacial charge separation, ΔE_{CS} determines device external quantum efficiency (EQE).

Our study employs films and devices incorporating three different electron donor polymers and seven different electron acceptors including NFAs, and includes steady state and transient optical data to determine the extent to which excitons decay to ground, geminate recombination of bound charges at the donor/acceptor interface and non-geminate recombination of dissociated charges impact upon the EQE and the photocurrent of the complete devices, and thus elucidate the key parameters determining device efficiency. Both our study and other reported studies provide indicators of how to achieve this, with for example well defined phase separation, internal donor charge transfer, high local carrier mobilities and optimal interface molecular structure all being shown to favour more efficient charge dissociation. The variation of short circuit current (JSC) and EQE between blends is not primarily determined by variations in light absorption, nor by variations in exciton separation efficiency nor by variations in charge collection efficiency. However, the increase in EQE with larger interfacial energy offset is shown to result from suppression of geminate recombination losses. This variation between blends is shown to be determined primarily by the magnitude of geminate recombination losses on the circa 10 ps – 1 ns timescale, assigned, following established literature, to the recombination of at least partially bound interfacial electron-hole pairs. These results are discussed in terms of their implications for the design of donor/NFA interfaces in organic solar cells, and strategies to achieve further advances in device performance. The challenge now is built upon this success to design complimentary donor and acceptor materials to achieve efficient charge separation with minimal energy offsets, so as to enable further advances in organic solar cell performance. The understanding of charge carrier recombination dynamics and the energetic dependence on polymer:NFA-based solar cells is a significant step towards the development of NFA-based photovoltaics. Consequently, we believe our work is appropriate for publication in Advanced Energy Materials.

We would like to propose the following as referees of this manuscript:

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Yours sincerely,

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Do you or any of your co-authors have a conflict of interest to declare?	No. The authors declare no conflict of interest.
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An Analysis of the Factors Determining the Efficiency of Photocurrent Generation in Polymer:Nonfullerene Acceptor Solar Cells

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Abstract

Herein a meta-analysis of the device performance and transient spectroscopic results are undertaken for various electron donor:electron acceptor blends, employing three different electron donor polymers and seven different electron acceptors including nonfullerene acceptors (NFAs). From this analysis, we find that the primary determinant of device external quantum efficiency is the energy offset driving interfacial charge separation, ΔE_{CS} . For devices employing the donor polymer PffBT4T blended with NFA and fullerene acceptors, an energy offset $\Delta E_{CS} = 0.30$ eV is required to achieve near unity charge separation, which increases for blends with PBDTTT-EFT and P3HT to 0.36 and ~ 1.2 eV, respectively. For

blends with PffBT4T and PBDTTT-EFT, a 100 meV decrease in acceptor LUMO level is observed to result in a ~2 fold increase in EQE. Steady state and transient optical data determine that this energy offset requirement is not associated with the need to overcome the polymer exciton binding energy and thereby drive exciton separation, with all blends studied showing efficient exciton separation. Rather the increase in EQE with larger interfacial energy offset is shown to result from suppression of geminate recombination losses. These results are discussed in terms of their implications for the design of donor/NFA interfaces in organic solar cells, and strategies to achieve further advances in device performance.

1. Introduction

Nonfullerene acceptors (NFAs) are attracting extensive academic and commercial interest as potential replacements for fullerenes in organic solar cells (OSCs), motivated by reports of higher power conversion efficiencies (PCEs), lower synthesis costs and improved device stabilities.^[1-7] Over last three years, NFA-based OSC performance has improved remarkably, with several studies reporting efficiencies over 13% driven primarily through improved molecular design.^[4-7] Particular progress has been made in enhancing the open circuit voltage (V_{oc}) of OSCs, achieved by use of NFAs with higher the lowest unoccupied molecular orbital (LUMO) levels as well as suppression of non-radiative recombination losses.^[8-14] However, progress in achieving improvements in photocurrent generation has proven more challenging, despite NFAs typically exhibiting superior light harvesting properties compared to fullerene acceptors.^[15-18] Indeed, with a small number of notable exceptions,^[19,20] most OSCs employing NFAs have yielded lower external quantum efficiencies (EQEs) than equivalent fullerene-based devices. In this paper, we undertake a meta-analysis of the device performance and transient spectroscopic results for various polymer:NFA and polymer:fullerene blends, employing three different electron donor polymers and seven different electron acceptors. From this analysis, we identify the primary factor limiting the

efficiency of photocurrent generation in these blends and discuss its implications on materials design to further improve polymer:NFA blend solar cell performance.

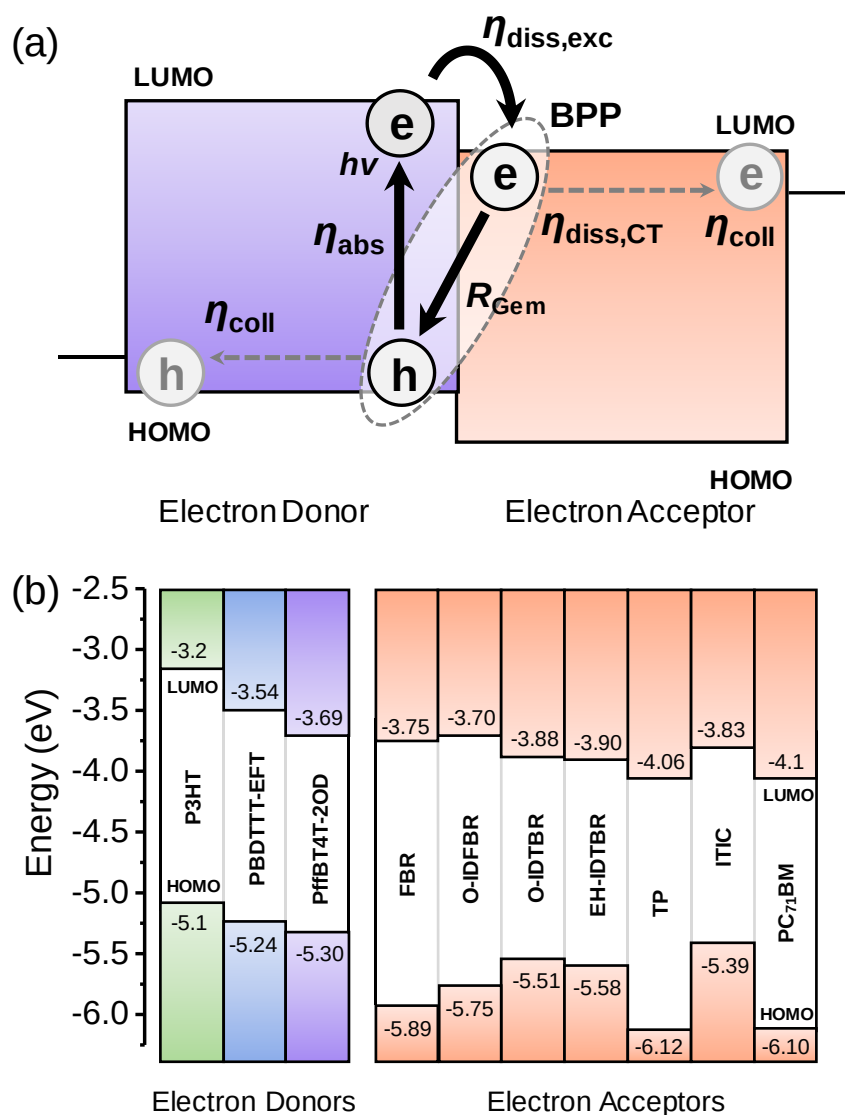


Figure 1. (a) Energy level diagram of an interface between electron donor and electron acceptor showing photoexcitation (η_{abs}) of an electron into LUMO of the electron donor, followed by exciton dissociation ($\eta_{\text{diss,exc}}$) via electron transfer into LUMO of the electron acceptor and then migration of the separated charges away from the interface ($\eta_{\text{diss,CT}}$). The electron transfer process initially generates a Coulombically bound polaron pairs (BPP) state that also requires charge dissociation before the free charge carriers can be collected (η_{coll}) by the electrodes. (b) Energy level diagram measured from thin films, where bandgap from optical spectroscopy and HOMO energy levels of conjugated polymers and LUMO energy levels of electron acceptors from using cyclic voltammetry.^[21,54]

Following literature reviews,^[21-23] the key processes of photocurrent generation in OSCs are illustrated in **Figure 1a**: exciton generation by light absorption, exciton diffusion to,

1 and dissociation at, the donor/acceptor interface, subsequent charge separation into free
2 charge carriers, and finally charge transport to, and charge collection by, the electrodes. The
3
4 parameters determining the efficiency of this photocurrent generation and particularly
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6 photoinduced charge separation in OSCs remains the subject of much debate. The efficiency
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8 of exciton diffusion to the donor/acceptor interface is primarily dependent upon blend
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10 morphology. The charge separation process has been reported to be dependent upon dielectric
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12 constant of blend, the energy offset driving charge separation, the charge carrier mobility
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14 and/or the molecular structure and electronic interactions at the donor/acceptor interface.^[24-27]
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16 Photocurrent generation can be further limited by charge collection efficiency losses resulting
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18 from non-geminate recombination during charge transport.^[28] As such, the dominant
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20 parameter(s) that determine the efficiency of photocurrent generation in NFA-based OSCs is a
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22 key consideration for further optimisation of material design and device performance.
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29 In the charge separation process, several reports have indicated that the geminate
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31 recombination of initially generated charge pairs at the donor/acceptor interface can be a
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33 significant loss pathway limiting the photocurrent generation.^[29,30] The importance of this
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35 geminate recombination, and the extent to which interfacial charge pairs (often referred to as
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37 charge transfer states when optically coupled to the ground state) are Coulombically bound or
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39 readily dissociate, remains an area of controversy.^[31-34] In this study we report herein, we find
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41 that efficiency of photocurrent generation is correlated with the magnitude of the energy
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43 offset driving charge separation, at least for the material systems studied herein, and relate
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45 this observation to the magnitude of geminate recombination losses at the donor/acceptor
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47 interface.
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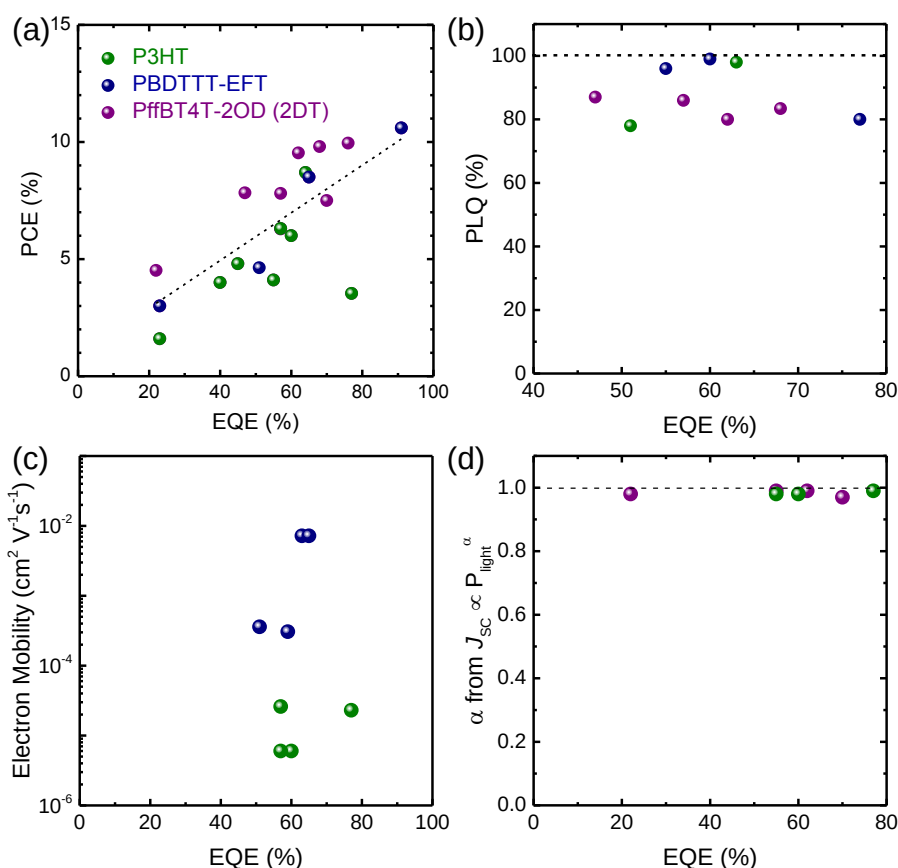


Figure 2. (a) PCEs (the dash-line using a linear fit analysis), (b) PL quenching yields, (c) electron mobilities from SCLC and (d) α from $J_{\text{SC}} \propto P_{\text{light}}^\alpha$ as a function of external quantum efficiencies (EQEs) in various NFA-based blends incorporating PffBT4T (purple), PBDTTT-EFT (blue) and P3HT (green) as an electron donor, respectively.

Results

The study reported herein employed three different donor polymers P3HT^[35-36], PBDTTT-EFT^[37] and PffBT4T^[38], blended with six different NFAs, and PCBM as a control acceptor. The highest occupied molecular orbital (HOMO) and LUMO energy levels and molecular structure of these materials are illustrated in Figure 1b and Figure S1, respectively. Details of blend layer processing and device fabrication are described in the supporting information. All devices employed an inverted device architecture, ITO/ZnO/organic blend/MoO₃/Ag. Device performance characteristics are listed in Table S1 and are similar to previously reported literature values where available.^[39] Figure S2 plots the correlations between PCE and the

corresponding underlying device parameters: short circuit photocurrent density (J_{SC}), V_{OC} , and fill factor (FF). It is apparent, that whilst high V_{OC} and FF are essential for efficient device performance, PCE is most strongly correlated with J_{SC} , with low J_{SC} being the primary cause of lower PCEs for these device series.

Variations in J_{SC} between blends can result, at least in part from variations in light harvesting efficiency, with some blends exhibiting lower optical bandgaps and more complementary donor and acceptor absorption, as have been discussed previously for several polymer:NFA solar cells.^[4-7] However, such variations in light harvesting do not appear to be the primary determinant of the variations in J_{SC} for the blends studied herein. **Figure 2a** plots the correlation between PCE and EQE, with these EQEs being determined at the peak blend absorption, which was relatively invariant between blends (peak optical absorption ranging from 70 to 75 %). It is apparent from Figure 2a and S2a that these EQE peak values which range from 20% to 90 % for the blends studied herein (see Table S2 for EQE values for each blend) are the primary determinant for the variation in J_{SC} . Hereafter, we address the origin of this variation in EQE values between blends.

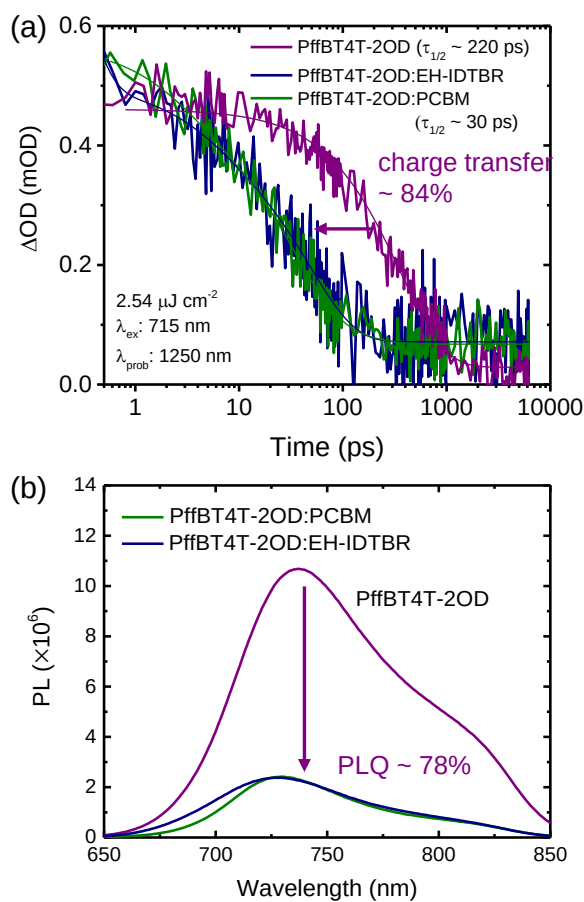


Figure 3. (a) Transient absorption kinetics of a neat PffBT4T-2OD film, and of PffBT4T-2OD blend films with EH-IDTBR and PC₇₁BM, excited at 715 nm and probing PffBT4T-2OD exciton absorption at 1250 nm, and (b) photoluminescence spectra of the same neat and blend films, excited at 600 nm.

We consider first whether the efficiency of exciton quenching at the donor/acceptor interface is a key determinant of the variation of EQE values for the blend systems studied herein. Figure 2b plots the efficiency of photoluminescence quenching (PLQ) measured in blends versus EQEs from devices, with Table S2 again giving values for each blend studied. For almost all blends studied, efficient PLQ (80 - 99%) is observed, indicative of efficient exciton separation at the donor/acceptor interface. This conclusion was further supported by ultrafast transient absorption studies of exciton decay dynamics in neat and blend films, where ultrafast (< 10 ps, and typically < 1 ps) exciton separation was observed, at least 10 fold faster than exciton decay to ground (see for example Figure 3 and also ref. 32 and 37-39). An excellent correlation was obtained between the variations in efficiency of exciton separation

determined from the PLQ and from transient absorption spectroscopy (TAS) data, confirming the validity of these measurements (see Figures 3 and S3). Furthermore, it is apparent from Figure 2b (and also from Figure S5a and Table S3 for other polymer:NFA blends) that there is no significant correlation between PLQ and EQE, indicating that variations in efficiency of exciton separation are not the primary determinant of the observed variations in EQE for the blends studied herein.

We turn now to consideration of whether variations in charge collection efficiency could be the determinant of the variation in EQE between blends. Charge collection efficiency is primarily determined by the competition between charge transport to the device electrodes and non-geminate recombination losses during this charge transport. We note that poor collection efficiencies typically result in low FFs as well as potentially lower J_{SC} . The rather small variation in FFs for the blends studied herein (40-60%), and the absence of significant correlation between FF and PCE (Figure S2c), both indicate that variations in collection efficiency between blends are not the primary determinant of variations in EQE and J_{SC} . We note that PffBT4T-2OD blends showed a more significant correlation between FF and PCE compared to blends of P3HT and PBDTTT-EFT, most probably resulting from field dependent charge generation for some blends in this series, as we have reported previously,^[16] rather than a variation in collection efficiency. Supporting this conclusion, we did not observe, where the data was available, a significant correlation between electron mobility or hole/electron mobility balance (determined from space charge limited current data) and EQE for the blends studied herein, as shown in Figure 2c (see also Figures S5b and S5c and Table S3 for electron mobility versus EQE data for other polymer:NFA blend devices). Furthermore, analysis of J_{SC} versus light intensity showed near linear behaviour ($J_{SC} \propto P^\alpha$ with α in range of 0.97 to 0.99), as shown in Figure 2d and S6. Such near linear behavior is indicative of the absence of significant non-geminate recombination losses at short circuit, and therefore efficient charge collection. We note that non-geminate recombination losses are important

determinants of FF and V_{OC} , and can limit J_{SC} for thick photoactive layers (in the study herein relatively thin photoactive layer thicknesses were employed, in the range 70 - 150 nm, with the exception of PffBT4T-2OD:PCBM).^[43,44] We also note that inefficient charge collection has been reported to limit the photocurrent generation in polymer/NFA blends which employed relatively crystalline acceptors, attributed to the unfavorable blend morphologies preventing electron transport.^[45-47] Such considerations are however beyond the scope of this study. Rather for the blends studied herein, these data suggest that variations in charge collection efficiency do not appear to be the primary determinant of the variations in EQE observed between blends.

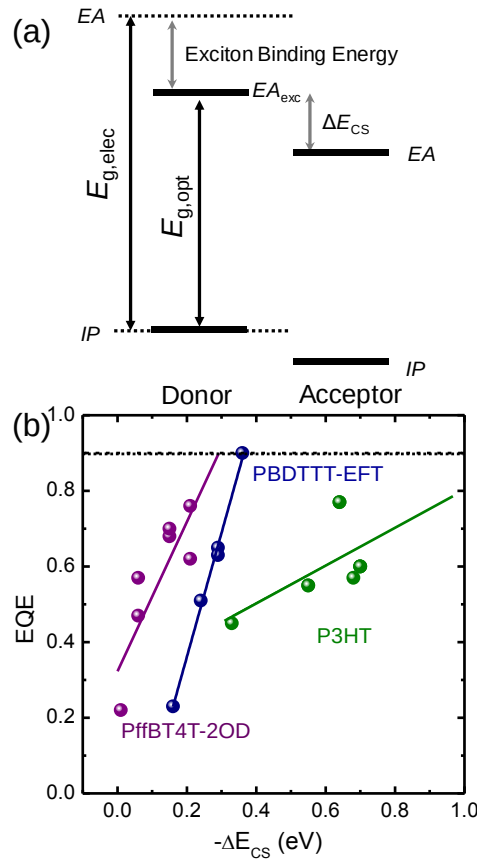


Figure 4. (a) Electronic energy level diagram depicting proposed model for exciton binding energy and energy offset $-\Delta E_{CS}$. In this diagram, $E_{g,opt}$ and $E_{g,elec}$ refer to the polymer optical and electrical bandgaps respectively, with the difference between these two values corresponding to the exciton binding energy. (b) EQEs as a function of energy offset $-\Delta E_{CS}$ in various NFA-based blends incorporating PffBT4T (purple), PBDTTT-EFT (blue) and P3HT (green) as an electron donor, respectively. The solid lines have been obtained by using a linear fit analysis and the dashed line in (a) at EQE = 0.9 corresponds to near unity internal charge generation efficiency.

We turn now to the consideration of the extent to which the observed variation in EQE values correlates with variations in energy offsets driving charge transfer at the donor/NFA interface. **Figure 4** shows a plot of the correlation between EQE values determined at the peak polymer absorption and the energetic driving force for charge separation from polymer excitons, $\Delta E_{CS} = (IP_D - EA_A) - E_{g,opt}$ where $E_{g,opt}$ is the polymer singlet exciton energy E_s estimated from its optical bandgap and the energy of the dissociated charges is determined from both the ionization potential of donor (IP_D) and the electron affinity of acceptor (EA_A) which are measured by cyclic voltammetry. Details of these energy level measurements are given in Figure S7. We note that our calculation of the energy offset ΔE_{CS} already accounts for the exciton binding energy, as the exciton energy employed in calculating ΔE_{CS} is determined optically from the energy required for exciton formation. As such $\Delta E_{CS} = 0$ corresponds essentially to an electronic donor/acceptor LUMO energy offset just sufficient to match the exciton binding energy, as shown in Figure 4a.

The determination of absolute values for the interfacial energy level offset ΔE_{CS} are highly measurement and analysis technique dependent. For example, we determine $E_{g,opt}$ from the mid-point of absorption and emission spectra for neat films of each material (see Figure S9). An alternative assay is from extrapolation of the absorption onset ($E_{g,abs}$ see Figure S9). $E_{g,opt}$ is less than $E_{g,abs}$ by 50-100 meV, attributed due to exciton relaxation following photoexcitation. It is possible that such relaxation may not be complete prior to electron transfer, such that in some cases (where for example exciton separation occurs in < 1 ps) $E_{g,abs}$ may be a more appropriate assay than $E_{g,opt}$. Our assay of the energetics of free charges is based upon cyclic voltammetry data of the onset of neat film oxidation/reduction $IP_D - EA_A$ (and so neglects variations in these values between neat and blend films)^[45]. An alternative assay of the energy of charge pairs is measurement of charge transfer state energy, E_{CT} . E_{CT} differs from $IP_D - EA_A$ as it includes the CT state coulomb binding energy, and is also likely

to differ in polarisation/relaxation energies; its use would therefore formally result in the determination of different energy offset to that addressed herein. A detailed consideration of these different approaches to assaying such interfacial energy offset in organic BHJ's is beyond the scope of this study. As a limited analysis of this issue, we compared data for measurement of ΔE_{CS} and $\Delta E_{CT} = E_{CT} - E_{g, opt}$, where the E_{CT} was determined from the midpoint of EQE and CT state electroluminescence spectra. Typical data for one blend, PffBT4T-2OD:EH-IDTBR, are shown in Figure S8. For this system, ΔE_{CT} was determined to be 50 meV, approximately 170 meV smaller than ΔE_{CS} . For other blends, we also typically observed that ΔE_{CT} energy offsets were smaller than ΔE_{CS} values. This suggests that our experimental approach may result in a modest overestimate of interfacial energy offset, although the larger values for ΔE_{CS} may also reflect more fundamental differences between the definitions of ΔE_{CS} and ΔE_{CT} , as discussed above. In anycase, given these uncertainties in absolute determination of energy offsets, we focus primarily here only relative values of this offset between blends.

For the blends studied herein, clear correlations are observed between EQE and $-\Delta E_{CS}$ for blends of three donor polymer with different acceptors, as shown in Figure 4b, consistent with the correlations between J_{SC} and $-\Delta E_{CS}$ illustrated in Figure S10. These correlations are clearer for the PffBT4T and PBDTTT-EFT blend systems compared with P3HT. The poorer correlation employing P3HT as the donor polymer can be most probably attributed to larger variations in the blend morphology with different acceptors, as we discuss further below (we note that due to overlap of P3HT emission and acceptor absorption, exciton energy transfer may also in part explain the poorer correlation). It is also apparent that the P3HT blend series appears to require larger ΔE_{CS} values to achieve efficient EQEs, which we also discuss further below. For both the PffBT4T and PBDTTT-EFT series, a 0.1 eV decrease in acceptor E_{A} , and therefore increase in ΔE_{CS} , results in an approximately two-fold increase in EQE. It can thus be concluded that, at least for the PffBT4T and PBDTTT-EFT blend series, the acceptor

electron affinity, and therefore the energy offset ΔE_{CS} , is the key determinant of the efficiency of EQE and photocurrent generation.

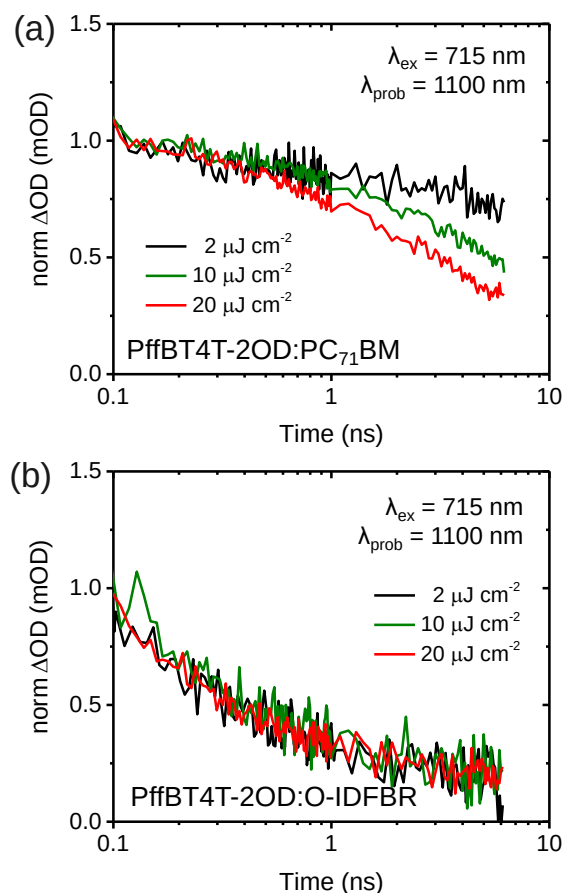


Figure 5. Transient absorption decay dynamics for neat polymer film, (a) PffBT4T-2OD:PC₇₁BM blend film and (b) PffBT4T-2OD:O-IDFBR blend film excited at 715 nm and probed at 1100 nm with various excitation densities from 2 to 20 $\mu\text{J cm}^{-2}$.

In order to investigate further the correlation between EQE and ΔE_{CS} , we turn to transient absorption studies which show the charge generation efficiency and lifetime in the blends studied herein. Transient absorption data were collected following the decay dynamics of polymer polaron photoinduced absorption in the near IR as a function of excitation density on timescales from 200 fs – 6 ns, following procedures reported previously. Typical data for two of the blends studied herein are shown in **Figure 5** (see also references 13 and 35 for more of the blends studied herein). As discussed previously, excitation density dependent decay dynamics, observed at higher excitation densities, are indicative of non-geminate (bimolecular

recombination) whilst excitation density independent decays are indicative of geminate (monomolecular) recombination. For all blends, the excitation density was lowered until any polaron absorption decay on the timescale studied was observed to be excitation density independent, thereby removing any contribution of non-geminate recombination from the transient data. In some cases, geminate recombination could not be clearly temporally or spectrally distinguished from exciton decay, preventing direct determination of the magnitude of geminate recombination losses for some blends (see for example reference 13, 35). As such we employ the amplitude of polymer polaron photoinduced absorption observed at 1 ns, after both exciton decay and geminate recombination is largely complete but prior to the onset of non-geminate recombination, as a direct assay of the yield of spatially separated charges in each blend.

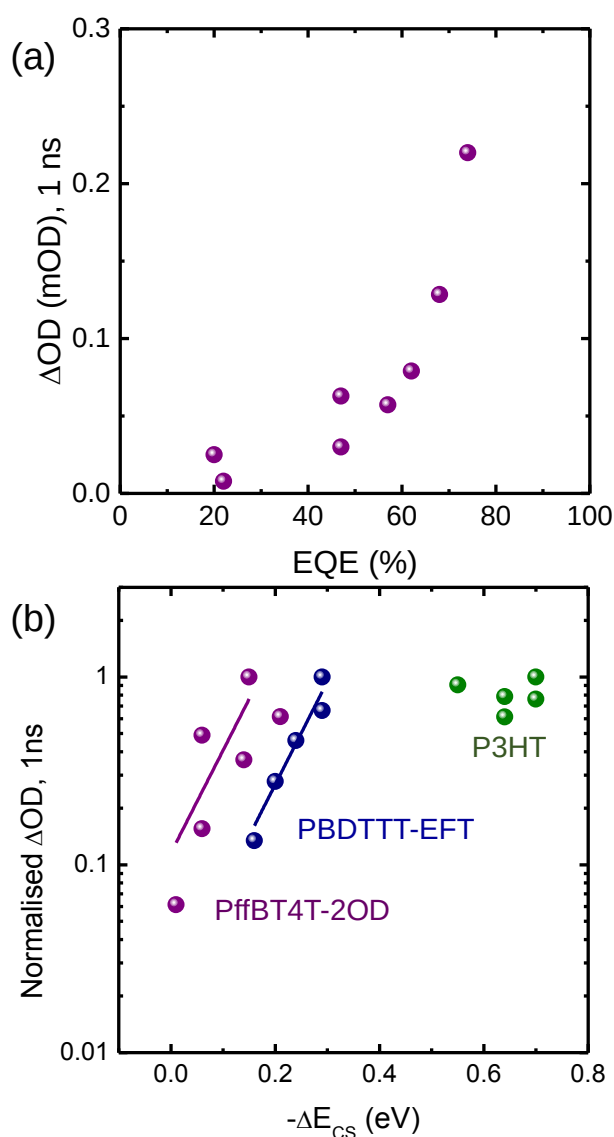


Figure 6. (a) Correlation between ΔOD assay of polaron yield measured by transient absorption spectroscopy on thin blend films and EQE measured in complete devices for a series of PffBT4T:electron acceptor blends, and (b) Transient absorption assay of the yield of dissociated charges determined from the magnitude of the ΔOD signals at 1 ns delay time normalised to matched densities of absorbed photons : various NFA-based blends incorporating PffBT4T (purple), PBDTTT-EFT (blue) and P3HT (green). For each series, the largest ΔOD signal is normalized to one to facilitate comparison between series, as differences in polaron extinction coefficients between polymer series are possible. The solid lines were obtained as linear best fits to these log/linear plots.

In **Figure 6a**, we plot the transient absorption assay of the charge separation yield (ΔOD) versus device EQE. It is apparent that we observe a clear correlation between the amplitude of our transient absorption assay of polaron yield ΔOD , and EQE for the series of PffBT4T:acceptors blends, consistent with the device EQE of these blends being primarily

determined by the yield of separated charges measured at this 1 ns time delay. A similar correlation was observed for the ΔOD measured at 6 ns (at this time delay, all geminate recombination is complete, but the data is more susceptible to contributions from non-geminate recombination losses), confirming the reliability of our analysis procedures (Figure S11). The correlation in Figure 6a is not linear, most probably due to the overlap between the absorption of donor and acceptor at the excitation wavelengths employed (we note that due to the variations in acceptor absorption region and spectral overlap, reliable trends between ΔOD and EQE could not be determined for the acceptor excitation). Overall these data indicate that for the blend systems studied herein, the primary determinant of the variation of EQE is the yield of charges observed on the nanosecond timescale. Given that these blends all exhibit similarly efficient exciton separation, this variation in the yield of dissociated charges can be assigned to the difference in the magnitude of geminate recombination loss of interfacial charge pairs generated by exciton separation.

In order to probe the origin of this variation in the charge generation efficiency between the blends, we investigate the correlation between our TAS assay of charge generation yield ΔOD (1 ns) versus the energy offset driving charge separation, $-\Delta E_{CS}$, as shown in Figure 6b. It is apparent that there is an excellent correlation between the energy offset driving polaron generation and the yield of dissociated polarons. This correlation strongly suggests that for these PffBT4T and PBDTTT-EFT blend series, the charge dissociation efficiency, correlated to photocurrent generation efficiency, is driven primarily by the energy offset driving charge separation, ΔE_{CS} .

Discussion

The study herein addresses the factors determining the efficiency of photocurrent generation in blend films employing three distinct donor polymers with six NFAs and with PCBM. Overall, we find that the variation of J_{SC} and EQE between blends is not primarily determined

by variations in light absorption, nor by variations in exciton separation efficiency nor by variations in charge collection efficiency. Rather this variation between blends is shown to be determined primarily by the magnitude of geminate recombination losses on the circa 10 ps – 1 ns timescale, assigned, following established literature, to the recombination of at least partially bound interfacial electron-hole pairs. We moreover find that, at least for blends with PffBT4T and PBDTTT-EFT donor polymers, this variation in geminate recombination losses is determined primarily by the energy offset driving charge separation, and therefore for each polymer series by acceptor LUMO energy.

The importance of the geminate recombination of bound interfacial charge pairs in limiting the photocurrent generation in polymer:fullerene blends remains controversial. Some studies, including our own, have reported on its importance, while others find that some interfacial charge pairs (often referred to in such studies as charge transfer states) can readily dissociate without undergoing geminate recombination. Among the many studies of organic solar cells employing NFAs, the consideration of geminate recombination losses in limiting the device performance, particularly the device photocurrent, has been largely absent from the literature to date, apart from our own studies with a limited number of blends^[55]. In particular, we have recently reported that the suppression of geminate recombination losses can result in higher EQE values in blends of PSEHTT with two alternative NFAs, FTB and FTTB.^[48] In contrast most other studies of polymer/NFA blends and devices to date have focused rather on the importance of exciton separation and/or carrier mobility in limiting photocurrent generation, without explicitly considering the role of geminate charge recombination losses.^[40-42] The results reported herein clearly indicate that, at least for the materials studied herein, direct consideration of geminate recombination losses is critical to understand the parameters limiting the photocurrent generation in polymer:NFA blend devices.

Similarly, the importance of energy offset, or ‘excess energy’, in determining the magnitude of geminate recombination losses is also controversial. Whilst several studies,

including our own, have provided evidence of its importance, other studies have found for other material systems that other parameters are more important, such as morphology, carrier mobility and dielectric constant.^[21,45] For the study herein, we observe a clear correlation between EQE and ΔE_{CS} for blend series of two donor polymers, PffBT4T and PBDTTT-EFT, but only a weak correlation for the third donor polymer P3HT. For all the PffBT4T and PBDTTT-EFT blends studied herein, it is reasonable to expect that from previous literature^[16,38,55], the blend morphology is relatively acceptor independent – with PffBT4T being highly crystalline and PBDTTT-EFT being amorphous. In contrast, the crystallinity of P3HT in the blends studied is strongly acceptor dependent, with it being highly crystalline in annealed films blended with PCBM, but being relatively amorphous in blends with, for example, the NFA FBR.^[35] It is therefore likely that the poor correlation between the acceptor LUMO level and the EQE for blends with P3HT primarily results from the additional dependence of geminate recombination upon blend morphology and potential variations in non-geminate recombination, consistent with studies reported previously for polymer:PCBM blends.^[49-51] We note that P3HT data is included in this manuscript in part to emphasise that the observation of direct correlations between EQE and ΔE_{CS} is dependent upon the choice of materials series, with clear correlations only being observed when other factors which may also influence EQE being relatively invariant.

The correlation we observe between the acceptor LUMO level and the device EQE has important implications for NFA design. The development of NFAs with LUMO levels higher than PCBM has been a key factor behind the increases in device voltage, and reduced energy loss ($E_g^{opt}/q - V_{OC}$) obtained with NFAs.^[10] The data reported herein indicate a clear limitation to this approach, indicating that higher LUMO levels will result in lower EQEs, unless other factors are introduced to facilitate the charge dissociation. This dependence of EQE upon energy loss has been reported previously for other systems, but typically without a discussion about the importance of geminate charge recombination in determining this dependency.^[52-53]

The energy offset required for an efficient charge dissociation observed herein appears significantly lower for blends with PffBT4T and PBDTTT-EFT donor polymers than with P3HT. Extrapolation of the EQE versus ΔE_{CS} plots in Figure 3b indicate whilst PffBT4T blends require an energy offset ΔE_{CS} of only 0.3 ± 0.05 eV for an EQE of 90%, indicative of near unity internal charge generation. This increases to 0.36 ± 0.03 eV for PBDTTT-EFT blends and $\sim 1.2 \pm 0.2$ eV for P3HT blends. As discussed above, our optical/cyclic voltammetry assay of ΔE_{CS} results in modestly larger energy offset values than those determined from EQE/electroluminescence data, such that the absolute values of ΔE_{CS} determined herein maybe modest overestimates of the energy offset required to achieve near unity charge separation. Whilst the origin of the dependence of ΔE_{CS} on donor polymer is unclear, it appears likely to be related to the more push-pull or internal charge transfer character of the PffBT4T and PBDTTT-EFT donor polymers compared to P3HT. The smaller energy offset requirement for PffBT4T relative to PBDTTT-EFT may also result from the higher crystallinity of PffBT4T.

We observe that, for the PffBT4T and PBDTTT-EFT blend series, an increase in the acceptor LUMO level, corresponding to a decrease in energy offset, by 0.1 eV results in a two-fold decrease in EQE. A 0.1 eV increase in acceptor LUMO level can also be expected to result in a 0.1 V increase in device V_{OC} . As such these data provide a clear guideline to the LUMO level required for these blend series for optimum performance, all other things being equal. It is interesting to note that the acceptor yielding the highest device performance for PBDTTT-EFT is neither that with the highest LUMO (FBR) or lowest (PCBM), but rather acceptors with an intermediate LUMO level (IDTBR), selected to achieve the optimal voltage whilst still retaining a sufficiently large energy offset to enable reasonably efficient photocurrent generation.^[55]

The results herein indicate that the requirement for an electronic orbital energy offset to drive the charge separation in the polymer:NFA blends studied is primarily not limited by

the requirement of an offset to drive exciton separation, but rather by the need to suppress the subsequent geminate recombination of charges generated from exciton separation at the donor/acceptor interface. More specifically, for the blends studied herein, the efficiency of exciton separation, as assayed by PL quenching or transient absorption measurements of exciton kinetics, is independent of energy offset for almost all the blends studied (see Figure S12 for further illustration of this). In contrast, we observe that an increased energy offset correlates strongly with a suppression of geminate recombination losses. We have observed analogous correlations from optical data alone in polymer:fullerene blends.^[21,45] For all the blends studied herein (ΔE_{CS} in the range 0 - 0.9 eV), the LUMO level energy offset is larger than the exciton binding energy (we note this analysis is only approximate due to the difficulty in determining absolute values for ΔE_{CS} , as discussed above). As such our determination of an energy offset of $\Delta E_{CS} = 0.3$ eV for near unity efficiency dissociated charge generation in PffBT4T blends implies this energy offset requirement above that required to overcome the exciton binding energy. We also note that the presence of an exciton binding energy in organic semiconductors does not imply a reduction in achievable device efficiency, as the exciton binding energy also lowers the optical bandgap of the polymer, thereby enhancing light absorption. As such, it appears reasonable that the presence of an exciton binding energy in organic materials does not represent a fundamental limit to device efficiency. However, the additional requirement for a higher energy offset (i.e.: $\Delta E_{CS} > 0$ eV) to enable charges generated at the donor/acceptor interface to dissociate efficiently and thereby avoid geminate recombination losses, appears to be a more fundamental limit to the potential efficiency of OSC devices. In this regard, it is apparent that our observation that P3HT based devices require rather large ΔE_{CS} values (~ 1.2 eV) for efficient charge dissociation implies a severe, and potentially fundamental limit, to the efficiency of P3HT based solar cells. This is consistent with the more modest efficiencies achieved with this donor polymer even with acceptors selected for complimentary, lower bandgap absorption

and higher V_{OC} . In contrast, this fundamental limitation seems much less severe for PffBT4T and PBDTTT-EFT devices, consistent with the higher device efficiencies obtained with these donor polymers with a range of fullerene and NFAs.

Conclusion

It can be concluded from the results and discussion above that a clear objective for both donor and acceptor design in organic solar cells should be the reduction of the additional energy offset ΔE_{CS} required to drive the near unity quantum efficiency generation of dissociated charges. Both the study herein, and other studies, provide indicators of how to achieve this, with for example well defined phase separation, internal donor charge transfer, high local carrier mobilities and optimal interface molecular structure all being shown to favour more efficient charge dissociation. The development of NFAs such as IDTBR and ITIC is clearly a key breakthrough in driving towards more efficient OSC devices. The challenge now is built upon this success to design complimentary donor and acceptor materials to achieve efficient charge dissociation with minimal energy offsets, so as to enable further advances in OSC device performance.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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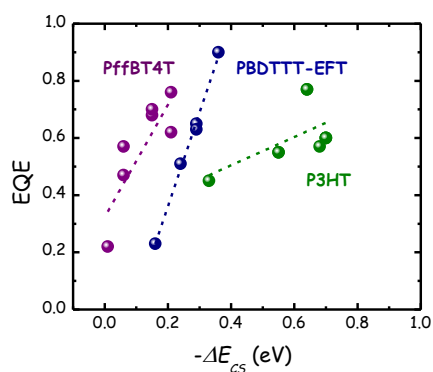
The primary factor limiting the efficiency of photocurrent generation is identified in polymer:nonfullerene acceptor (NFA) blends and discuss its implications for materials design to optimise organic solar cell performance. In this study we report that magnitude of this geminate recombination loss pathway is the key determinant of the efficiency of photocurrent generation in polymer:NFA blend solar cells.

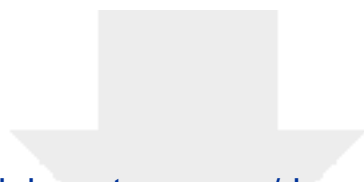
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Dr. H. Cha, C. H. Tan, J. Wu, Y. Dong, Dr. W. Zhang, Dr. H. Chen, Dr. S. Rajaram, Prof. K. S. Narayan, Prof. I. McCulloch, Prof. J. R. Durrant*

An Analysis of the Factors Determining the Efficiency of Photocurrent Generation in Polymer:Nonfullerene Acceptor Solar Cells

ToC figure

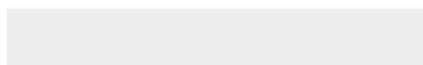
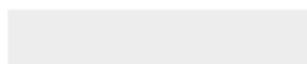




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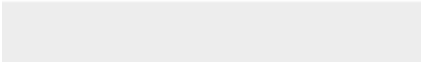
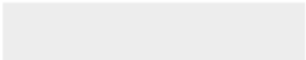




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