

# Correlating Charge-Transfer State Lifetimes with Material Energetics in Polymer:non-Fullerene Acceptor Organic Solar Cells

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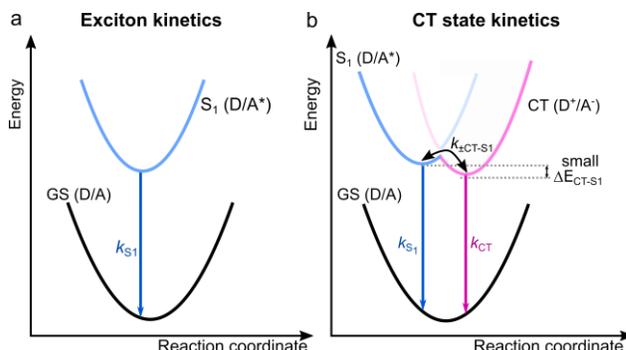
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**ABSTRACT:** Minimizing the energy offset between the lowest exciton and charge-transfer (CT) states is a widely employed strategy to suppress the energy loss ( $E_g/q - V_{OC}$ ) in polymer:non-fullerene acceptor (NFA) organic solar cells (OSCs). In this work, transient absorption spectroscopy is employed to determine CT state lifetimes in a series of low energy loss polymer:NFA blends. The CT state lifetime is observed to show an inverse energy gap law dependence and decreases as the energy loss is reduced. This behavior is assigned to increased mixing/hybridization between these CT states and shorter-lived singlet excitons of the lower gap component as the energy offset  $\Delta E_{CT-S1}$  is reduced. This study highlights how achieving longer exciton and CT state lifetimes has the potential for further enhancement of OSC efficiencies.

State-of-the-art organic solar cells (OSCs) utilize a bulk heterojunction blend between electron donor (D) and electron acceptor (A) materials.<sup>1-3</sup> Under illumination, excitons can dissociate at the D:A interface to form charge-transfer (CT) states, apparent from observations of light absorption into, and light emission from these states.<sup>4,5</sup> These CT states are key determinants of OSC device efficiencies;<sup>6-8</sup> for example the CT state energy ( $E_{CT}$ ) has been correlated with the open-circuit voltage ( $V_{OC}$ ) and voltage losses.<sup>9</sup> Reducing the energy offset driving charge separation, i.e. the energy offset between the singlet exciton ( $S_1$ ) and CT state ( $\Delta E_{CT-S1}$ ) can suppress energy losses, leading to a higher  $V_{OC}$ , but may also have a detrimental effect on the yield of free charges.<sup>10,11</sup> In many D:A devices, monomolecular CT state (i.e. geminate) recombination has been reported to be a key limitation on photocurrent generation.<sup>12-15</sup> The lifetime of CT states is a key consideration for device function, influencing how efficiently these states separate into free charges, as well as the kinetics of free carrier recombination via these states.<sup>16</sup> However, relatively few studies have addressed the parameters determining CT state lifetimes in such blends. In the

study herein, we address this issue, focusing in particular upon how the CT state lifetime is influenced by material energetics.



**Figure 1.** Potential energy surface diagrams illustrating (a) singlet exciton  $S_1$  decay to ground ( $k_{S1}$ ) in an organic semiconductor and (b) the impact of additional CT states in D:A blends. For low offset  $\Delta E_{CT-S1}$  blends,  $S_1$  and CT states can be expected to be in rapid thermal equilibrium as illustrated by  $k_{\pm CT-S1}$ , and/or hybridization. Both  $k_{S1}$  and  $k_{CT}$  include possible decay to lower energy triplet states, which is assumed to be irreversible, and both radiative and (predominately) non-radiative processes.

Analysis of non-radiative voltage losses in small molecule:fullerene OSCs has indicated that the CT state lifetime can follow the energy gap law, increasing with decreasing CT state energy.<sup>17,18</sup> Such energy gap law behavior for CT decay has been assumed in several analyses of voltage losses in OSCs.<sup>19-21</sup> Direct confirmation of this behavior has been provided by Collado-Fregoso et al., who observed from transient studies an increase of CT state lifetime with increasing CT state energy.<sup>22</sup> However the Collado-Fregoso study was limited to a series of relatively large driving force small molecule:fullerene blends.<sup>22</sup> The OSC field is now increasingly focusing on lower energy offset, high-efficiency blends employing

non-fullerene acceptors (NFAs).<sup>23</sup> Several recent studies have indicated that a low energy offset between exciton and CT state may result in the hybridization/mixing of these states,<sup>19,20,24</sup> This hybridization/mixing can be expected to impact substantially upon CT state lifetime, such that it may no longer exhibit energy gap law behavior. In the study herein, we investigate this possibility, determining the impact of material energetics upon CT state lifetimes in low-offset, high-performance polymer:NFA OSCs.

Figure 1 illustrates the impact of blend formation on the lifetime of singlet exciton and CT states in low-offset polymer:NFA blends. The impact of hybridization and/or thermal equilibrium between these states on the CT decay dynamics is likely to depend strongly on the relative magnitudes of  $k_{S1}$  and  $k_{CT}$ . For thermal mixing between these states, the equilibrium will shift towards  $S_1$  as  $\Delta E_{CT-S1}$  becomes smaller, as such  $k_{S1}$  will increasingly limit the CT state lifetime if  $k_{S1} > k_{CT}$ . For quantum mechanical hybridization, the behavior is more complex.<sup>19</sup> However in either case, if  $k_{S1} > k_{CT}$ , mixing between these states would give an opposite trend to the energy gap law behavior reported previously,<sup>22</sup> therefore requiring re-evaluation of the impact of CT state lifetime on non-radiative voltage losses in low-offset OSCs.

In this work, transient absorption spectroscopy (TAS) is employed to determine CT decay dynamics for seven polymer:NFA blend films (in Table 1) prepared following the same processing methods as for optimized devices (See Table S1 for full performance). CT state dynamics were monitored following direct excitation of the lower gap component (NFAs in most cases). TAS studies of polymer:NFA blends enable the separate tracking of both exciton and CT state dynamics, distinguishable by their distinct photoinduced absorption (PIA) peaks, as we and others have reported previously.<sup>25-27</sup> The blends studied herein were selected as they all exhibit both rapid ( $< 15$  ps) charge state generation from excitons and clearly observable, monomolecular CT state decay, as discussed below. These include both relatively amorphous (e.g.: PBDB-T:EH-IDTBR) and relatively crystalline (e.g.: PTQ10:IDIC) blends.

**Table 1.** Polymer:NFA blends with their open-circuit voltages ( $V_{oc}$ ) and energy losses ( $E_g/q - V_{oc}$ ) in devices and their CT state lifetimes.

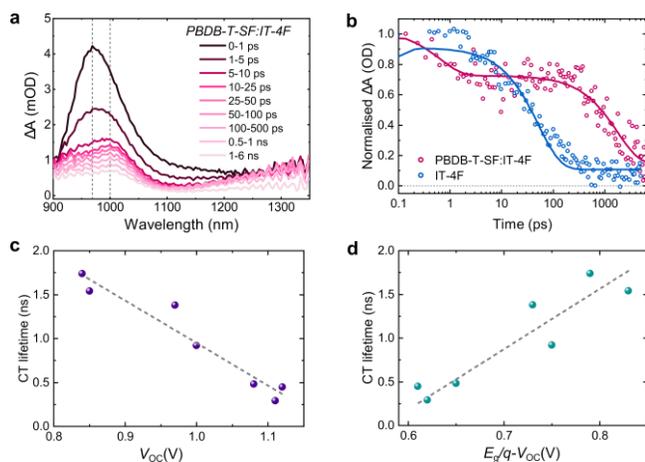
Blend systems	$V_{oc}$ (V)	$E_g/q - V_{oc}$ (V)	CT state lifetime (ns)
PffBT4T-2OD:FBR	1.11	0.62	$0.27 \pm 0.03$
PffBT4T-2OD:IDFBR	1.12	0.61	$0.45 \pm 0.08$
PffBT4T-2OD:EH-IDTBR	1.08	0.65	$0.48 \pm 0.03$
PBDB-T:EH-IDTBR	1.00	0.75	$0.92 \pm 0.20$
PTQ10:IDIC	0.97	0.73	$1.38 \pm 0.06$
PffBT4T-2OD:ITIC	0.85	0.85	$1.54 \pm 0.13$
PBDB-T-SF:IT-4F	0.84	0.84	$1.74 \pm 0.33$

As an illustrative example, the TA spectra and kinetics for one of the blends, PBDB-T-SF:IT-4F, and its corresponding pristine IT-4F film, are illustrated in Figure 2a and 2b. The pristine IT-4F film exhibits a PIA peak at 970 nm assigned to IT-4F  $S_1$  states (Figure S1). Under low intensity excitation, this PIA signal exhibits a monomolecular decay with a lifetime of  $51 \pm 3$  ps (blue circles in Figure 2b), assigned to the IT-4F exciton decay  $k_{S1}$ . In the blend film, this PIA signal exhibits a peak shift from 970 nm to 1000 nm within 10 ps, assigned to hole transfer from IT-4F excitons to PBDB-T-SF (Figure 2a). This ultrafast, and efficient, hole transfer was further confirmed by photoluminescence quenching studies (Figure S1). The 1000 nm PIA is thus assigned to CT/polaron absorption. Under low intensity excitation, this feature exhibits a monomolecular intensity-independent decay phase, indicative of geminate CT state recombination. Fitting the kinetics at 1000 nm yields a fast ( $< 10$  ps) and a slow ( $> 100$  ps) phase, attributed to

exciton dissociation and CT decay respectively. The CT state lifetime was extracted from the slow phase to be  $1.74 \pm 0.33$  ns, which is more than 30-fold longer than the IT-4F exciton lifetime. It is worth noting that the CT state decay in the PBDB-T-SF:IT-4F blend film shows a significant drop in the amplitude, although this device shows a reasonably high short-circuit current density of  $19 \text{ mA cm}^{-2}$ , suggesting the presence of field-dependent geminate recombination, as reported previously in similar low energy offset polymer:NFA blends.<sup>27,28</sup> However, we note here that, the contribution of both CT dissociation and, under the excitation fluences employed the encounter of an electron and a hole from the same CT state into the overall CT state lifetimes is likely to be minimal.<sup>29</sup> Extending the same characterization method to all blends, analogous data were obtained for CT state lifetimes in blend films (see Figure S2), we measured CT state lifetimes ranging from 0.27 ns to 1.74 ns. In most cases, these were longer than the dominant exciton lifetimes in the corresponding neat films of the lower gap component, as observed for PBDB-T-SF:IT-4F detailed above. For the shortest CT state lifetime of 0.27 ns, observed for PffBT4T-2OD:FBR, this lifetime was observed to be similar to the exciton lifetime observed for neat PffBT4T-2OD films.

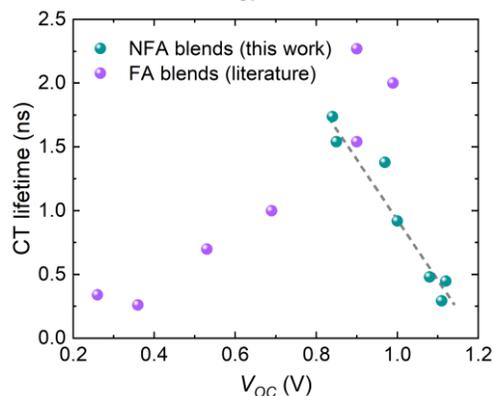
We now turn to correlation of the CT state lifetime with blend energetics. For low energy offset polymer:NFA blends, determination of CT state energies is particularly challenging, partly as for many blends the CT state emission cannot be clearly resolved from the exciton emission.<sup>30-32</sup> On the other hand, it is well established that CT state energetics exhibit an approximately linear correlation with  $V_{oc}$ .<sup>33</sup> As such, we employ  $V_{oc}$  as a convenient assay of the trend in CT state energy among blends. Figure 2c plots measured CT state lifetimes versus  $V_{oc}$ , showing that as  $V_{oc}$  increases from 0.84 V to 1.12 V, the CT state lifetime decreases from 1.74 ns to 0.27 ns. This observation of decreasing CT state lifetime with increasing  $V_{oc}$ , and therefore increasing CT state energy, is the inverse of that observed for the energy dependence of the CT state lifetime reported in the previous Collado-Fregoso study.<sup>22</sup> It is apparent that for this blend series, the CT state lifetimes do not exhibit energy gap law behavior, but rather the opposite trend, with the lifetime decreasing as the state energy increases.

To explore the origin of this ‘inverse energy gap law’ behavior further, we also plot in Figure 2d the CT state lifetime against  $E_g/q - V_{oc}$ , with this energy difference being a proxy for the energy offset  $\Delta E_{CT-S1}$  (it should be noted that  $E_g/q - V_{oc}$  is larger than  $\Delta E_{CT-S1}$ , most notably as  $V_{oc}$  is a free energy and includes the entropy increase associated with the generation of separated charge carriers, as well as energetic losses associated with charge trapping).<sup>10</sup> It is apparent from Figure 2d that a smaller energy offset leads to a faster decay of the CT state, with the lowest energy offset blend (PffBT4T-2OD:FBR) exhibiting a CT state decay with a similar time constant to the PffBT4T-2OD exciton observed in a neat PffBT4T-2OD film. This dependence is indicative of increased mixing/hybridization CT and exciton states as their energy difference is reduced, as further discussed below.



**Figure 2.** Transient absorption (a) spectra for a PBDB-T-SF:IT-4F film and (b) kinetics for IT-4F excitons in neat IT-4F films and CT states in PBDB-T-SF:IT-4F films, probed at 970 nm and 1000 nm, respectively. CT state lifetimes plotted (c) against the open-circuit voltage,  $V_{OC}$  and (d) against the energy loss,  $E_g/q - V_{OC}$ , where  $q$  is the elementary charge.

Before discussing the implications of the trend of CT state lifetime with energetics observed in Figure 2, we further extended our plot of CT state lifetime against  $V_{OC}$  to include literature data. As shown in Figure 3, the CT state lifetime shows a strong correlation with the device photovoltage, with opposite trends observed in different  $V_{OC}$  regimes. In the low  $V_{OC}$  regime ( $<$  circa 0.9 V), the CT state lifetime increases as  $V_{OC}$  increases, as reported previously,<sup>22</sup> whereas in the high  $V_{OC}$  regime ( $>$  circa 0.9 V), the CT state lifetime decreases as  $V_{OC}$  increases. The increase in CT state lifetime with increasing  $V_{OC}$  observed in the low voltage regime can be understood in terms of an energy gap law dependence for  $k_{CT}$ , as reported previously.<sup>22</sup> Conversely the inverse behavior in the high  $V_{OC}$  regime can be attributed to the concomitant reduction in the energy offset  $\Delta E_{CT-S_1}$  with increasing  $V_{OC}$  (see Figure 2c). This results in increased mixing/hybridization of the CT and exciton states, with the CT state decay becoming increasingly dominated by decay via the exciton decay pathway  $k_{S_1}$ . This conclusion is consistent with a recent analysis of the impact of exciton lifetime on device performance by Classen et al.<sup>34</sup> It is also consistent with reports that lowering the energy offset between CT and  $S_1$  states enhances the radiative efficiency (i.e.: a high electroluminescence yield) in NFA OSCs,<sup>35</sup> as the  $k_{S_1}$  will typically comprise a higher proportion of radiative recombination than  $k_{CT}$ .



**Figure 3.** CT state lifetimes plotted against  $V_{OC}$ , including the data from this work (green dots) and literature data taken from the reference (purple dots<sup>22</sup>).

The conclusion herein, that the reduction in CT state lifetimes observed for lower offset systems, is likely to be correlated with the relatively short exciton lifetimes observed for the specific NFAs

studied herein. PCBM has a singlet exciton lifetime in solid films  $>$  1 ns, such that mixing of CT and PCBM exciton states would be unlikely to accelerate CT state decay. As such, in blends where the lower gap component has long exciton lifetimes, mixing of exciton states with CT states is less likely to cause a strong acceleration of CT state decay. Hence, the strong reduction of CT state lifetime as  $\Delta E_{CT-S_1}$  is decreased is likely to be particularly pronounced for blends where the lower gap component has a relatively short exciton lifetime, as for the polymer:NFA blends studied herein. As such, short exciton lifetimes, as observed for some NFAs, impacts not only on the efficiency of exciton separation but also on the kinetics of recombination processes proceeding via CT states. This emphasizes the importance of maximizing the lifetime of NFA singlet excitons as well as material energetics for the further enhancement of the OSC performance.<sup>34</sup>

In summary, we have elucidated the role of energy offset on the kinetics of CT states in low driving force polymer:NFA blends. While CT state lifetimes in large offset systems follow an energy gap law with material energetics, low offset polymer:NFA blends exhibit an opposite trend, attributed to mixing/hybridization between these CT states and shorter-lived  $S_1$  states. Although suppressing the offset is beneficial to reduce voltage losses, it can impose a limit on the CT state lifetime when the exciton lifetime is short. Designing acceptor materials with long exciton lifetimes in the solid state and eliminating the fast geminate recombination of CT states in low-offset systems can thus pave the way for further efficiency increases in organic solar cells.

## ASSOCIATED CONTENT

(Word Style “Section\_Content”). **Supporting Information.**

The Supporting Information is available free of charge on the ACS Publications website.

Chemical names, structures and photovoltaic device performance, steady-state absorbance and photoluminescence spectra for the pristine and blend materials, transient absorption spectra and kinetics for the pristine and blend materials. (PDF)

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### Notes

No competing financial interests have been declared by the authors.

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