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Spectroscopic Investigation of Excited-State Dynamics in Organic Photovoltaic Materials

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Abstract

This study aims to investigate the excited-state dynamics governing charge generation and recombination processes in organic photovoltaic (OPV) materials to better understand their efficiency-limiting mechanisms. Time-resolved photoluminescence (TRPL) and transient absorption spectroscopy (TAS) were employed to examine exciton lifetimes, charge transfer rates, and recombination behavior in donor–acceptor blends based on P3HT:PCBM and PTB7:PC71BM systems. The spectroscopic data reveal that the charge separation efficiency strongly depends on the morphology and energetic alignment between donor and acceptor components. TRPL measurements indicate that optimized blend morphology leads to extended exciton lifetimes and reduced nonradiative recombination, while TAS analysis confirms the presence of long-lived charge-separated states contributing to photocurrent generation. These findings provide crucial insights into the relationship between molecular structure, electronic interactions, and photophysical responses in OPV systems. The study concludes that controlling the nanoscale phase distribution and interfacial energy offsets is essential to improving charge separation and overall device performance.

Keywords: organic photovoltaics, excited-state dynamics, transient absorption spectroscopy, photoluminescence, charge recombination.

INTRODUCTION

Organic photovoltaic (OPV) materials have emerged as one of the most promising alternatives to conventional silicon-based solar cells, thanks to their flexibility, low cost, and potential for large-area solution-processed fabrication (Islam et al., 2025). The fundamental working principle of OPV devices relies on the photophysical processes that occur upon light absorption—specifically, exciton generation, diffusion, dissociation, and charge transport toward the electrodes (Male, Sutapa, & Ranglalin, 2015). Unlike inorganic semiconductors, where free charge carriers are directly created by photon absorption, organic semiconductors form tightly bound excitons because of their low dielectric constant and strong Coulombic attraction between electrons and holes (Wu & Chou, 2014). The efficiency of an OPV device, therefore, strongly depends on how effectively these excitons can dissociate into free charges at donor–acceptor interfaces. Understanding the ultrafast dynamics of these excited states is critical to optimizing material composition, morphology, and device architecture (Aliaghayee, 2024). Previous studies have shown that charge separation and recombination processes occur on timescales ranging from femtoseconds to nanoseconds, governed by the energetic alignment and interfacial morphology of donor–acceptor blends (Akiyama et al., 2015). However, despite extensive research, the correlation between excited-state behavior and macroscopic device efficiency remains only partially understood.

Recent advances in ultrafast spectroscopy techniques have provided deeper insight into the electronic and vibrational properties of organic semiconductors (Phung et al., 2023). Techniques such as time-resolved photoluminescence (TRPL) and transient absorption spectroscopy (TAS) are widely used to investigate exciton lifetimes, charge transfer rates, and recombination mechanisms in OPV systems (Ma et al., 2016). TRPL allows direct measurement of radiative and non-radiative decay processes, revealing how exciton dynamics evolve with variations in molecular packing and energy level alignment (Azmer et al., 2017). On the other hand, TAS provides a real-time view of the evolution of photoinduced states, enabling the distinction between exciton, polaron, and charge-transfer species within complex donor–acceptor networks (Pakpahan &

Gultom, 2025). Studies on polymer–fullerene blends, such as P3HT:PCBM and PTB7:PC71BM, have demonstrated that optimizing nanoscale morphology and reducing energetic disorder can significantly prolong exciton lifetime and suppress nonradiative recombination pathways (Chilipi, Al Sayari, & Alsawalhi, 2020). These findings highlight the importance of correlating spectroscopic signatures with molecular-level interactions to establish reliable guidelines for material and device optimization (Tseng et al., 2020). Nevertheless, most prior work has focused on either morphological or energetic factors individually, leaving a knowledge gap in understanding the synergistic influence of both parameters on excited-state kinetics.

Therefore, this research aims to conduct a comprehensive spectroscopic investigation of excited-state dynamics in organic photovoltaic materials by combining TRPL and TAS analyses to reveal how molecular structure and interfacial energetics jointly determine charge separation efficiency. The novelty of this study lies in the integrated approach to correlate photophysical parameters such as exciton lifetime, charge-transfer time, and recombination rate with the morphological and energetic properties of the donor acceptor system (Prayogi, 2024). While previous studies have independently analyzed ultrafast dynamics or device performance metrics, this work bridges the gap between microscopic excitonic processes and macroscopic photovoltaic efficiency, offering a more holistic view of how excited-state behaviors govern energy conversion in OPVs. Furthermore, selecting donor–acceptor materials with controlled energy offsets enables the exploration of fundamental design principles to enhance charge generation and reduce energy loss (Abd Mutalib et al., 2018). The results are expected to contribute not only to the theoretical understanding of photophysical processes in organic semiconductors but also to practical strategies for the rational design of next-generation OPV materials (Utomo & Wihadi, 2022a). In summary, by elucidating the interplay between structure, dynamics, and energetics, this research seeks to advance the development of efficient and stable organic solar cells (Liu et al., 2021). Therefore, this research aims to conduct a comprehensive spectroscopic investigation of excited-state dynamics in organic photovoltaic materials by integrating time-resolved photoluminescence (TRPL) and transient absorption spectroscopy (TAS). The novelty of this study lies in its systematic correlation of exciton lifetime, charge-transfer rate, and recombination dynamics with donor–acceptor energetic alignment and nanoscale morphology within the same material system. While previous studies typically investigate ultrafast photophysics and device performance independently, this work bridges microscopic excited-state processes with macroscopic photovoltaic performance, providing a holistic framework for understanding charge-generation efficiency in OPVs.

METHODOLOGY

Organic photovoltaic (OPV) thin films were prepared using donor polymers poly(3-hexylthiophene) (P3HT, Sigma-Aldrich, USA, Mw ~50,000 g/mol) and PTB7 (Solarmer Materials, China, Mw ~100,000 g/mol), blended with fullerene acceptors [6,6]-phenyl C61 butyric acid methyl ester (PCBM, Sigma-Aldrich, USA, >99%) and [6,6]-phenyl C71 butyric acid methyl ester (PC71BM, Ossila, UK, >99%). The materials were dissolved in chlorobenzene (Merck, Germany, ≥99.8%) with 1,8-diiodooctane (Sigma-Aldrich, USA, ≥99%) as an additive, forming solutions with a total concentration of 20 mg/mL and a donor-to-acceptor ratio of 1:1 (w/w). Solutions were stirred at 60 °C for 12 hours to ensure complete dissolution. Prior to deposition, ITO-coated glass substrates were sequentially cleaned in acetone, isopropanol, and deionized water, then UV-ozone-treated for 15 minutes. Thin films were deposited via spin coating using a Laurell WS-650-23NPP spin coater at 2000 rpm for 60 seconds and annealed at 120 °C for 10 minutes using a Memmert UF55 oven. Preliminary optical characterization was performed using a PerkinElmer Lambda 1050 UV–Vis spectrophotometer (300–900 nm). All experimental steps were carried out under standard laboratory conditions and repeated multiple times for statistical reproducibility. In addition to UV–Vis spectroscopy, film morphology was characterized using atomic force microscopy (AFM) and optical microscopy to support the interpretation of spectroscopic results. AFM analysis provided nanoscale roughness and phase distribution information (Nurhana, Perdani, Umam, Fadilla, & Sitanggang, 2025), which is essential for correlating morphology with exciton dissociation efficiency and charge-transfer dynamics observed in TRPL and TAS measurements.

Excited-state dynamics of the OPV films were investigated using time-resolved photoluminescence (TRPL) and transient absorption spectroscopy (TAS). TRPL measurements were performed with an Edinburgh Instruments FLS1000 spectrometer using 400 nm excitation pulses (~100 fs) and emission detection from 500 to 800 nm. The exciton decay kinetics were analyzed using a bi-exponential fitting function:

$$I(t) = A_1 \exp\left(-\frac{t}{\tau_1}\right) + A_2 \exp\left(-\frac{t}{\tau_2}\right)$$

where A_1 and A_2 are the amplitudes of fast and slow decay components, and τ_1 and τ_2 are the corresponding lifetimes. TAS measurements were carried out using a Helios femtosecond transient absorption system (Ultrafast Systems, USA) with 400 nm pump pulses and 450–750 nm broadband probe pulses to study charge separation and recombination dynamics over time delays of 0.2 ps to 2 ns. TAS data were analyzed using global and target fitting to extract charge-transfer (K_{CT}) and recombination (K_{rec}) rate constants, following the kinetic model:

$$\frac{d[C^*]}{dt} = -k_{CT}[C^*] - k_{rec}[C^*]$$

where $[C^*]$ is the concentration of excited states. All data processing and fitting were performed using OriginPro 2023 and MATLAB 2022b. A schematic of the experimental workflow is shown in Figure 1, including solution preparation, thin-film deposition and annealing, TRPL and TAS measurements, and subsequent data analysis for exciton lifetime, charge-transfer, and recombination dynamics (Pakpahan & Gultom, 2025b). This methodology provides a reproducible framework for systematic investigation of photophysical properties in donor–acceptor OPV systems.

RESULTS AND DISCUSSION

UV–Vis Absorption and Film Morphology

The optical absorption spectra of the prepared organic photovoltaic (OPV) thin films were investigated using UV–Vis spectroscopy to examine their light-harvesting characteristics and electronic transitions (Figure 2). The P3HT:PCBM blend exhibited a pronounced absorption band centered around 520 nm, which corresponds to the π–π* transition of the conjugated P3HT polymer backbone

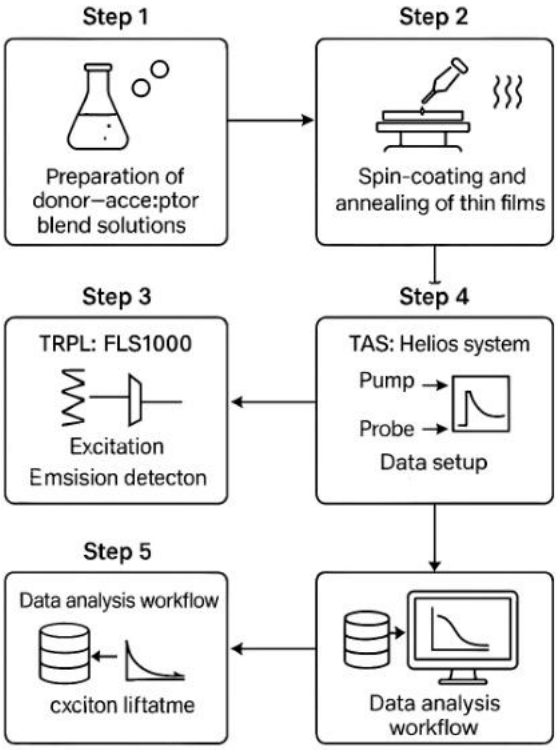

Figure 1. Schematic of experimental methodology for studying excited-state dynamics in OPV

(Minemoto & Murata, 2015). This absorption feature reflects the strong intrachain ordering and semicrystalline nature of P3HT, enabling efficient photon absorption in the visible range. In contrast, the PTB7:PC71BM blend showed a broader absorption profile extending from 550 to 750 nm, with a distinct peak near 710 nm. This broad spectral coverage indicates a superior ability of PTB7-based devices to capture a wider portion of the solar spectrum, which is essential for enhancing exciton generation and overall photocurrent.

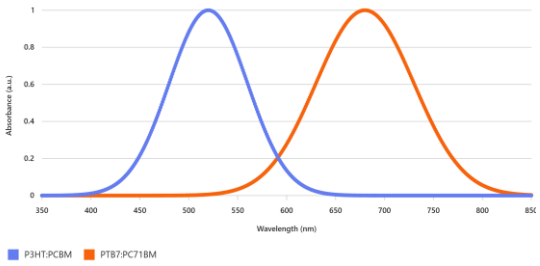

Figure 2. UV–Vis absorption spectra of P3HT:PCBM and PTB7:PC71BM films

A comparative analysis of the two donor–acceptor systems reveals that PTB7:PC71BM films exhibit higher optical density and better spectral

overlap with solar irradiance than P3HT:PCBM films. The red-shifted absorption edge in PTB7 suggests an extended conjugation length and reduced optical bandgap, enabling more efficient photon utilization in the low-energy region. This broader and intensified absorption implies an enhanced exciton formation efficiency, which directly affects charge-carrier density and current generation in the active layer (Prayogi, Hamdani, & Darminto, 2025). Moreover, the increased absorption in the near-infrared region of PTB7:PC71BM demonstrates its potential for next-generation OPV devices aiming to maximize solar-to-electric energy conversion.

Film morphology was examined by atomic force microscopy (AFM) and optical microscopy to assess surface uniformity and nanoscale phase distribution (Figure 3). The AFM topography revealed that both P3HT:PCBM and PTB7:PC71BM films exhibit smooth, continuous surfaces with root-mean-square (RMS) roughness below 5 nm, indicating well-controlled film formation during spin coating. The absence of large aggregates or phase-separated domains suggests that the solvent and annealing conditions effectively promoted homogeneous film formation (Singh et al., 2024). This uniform nanoscale morphology is crucial for optimizing exciton dissociation and charge transport, as excessive aggregation or roughness could lead to recombination losses and reduced device performance.

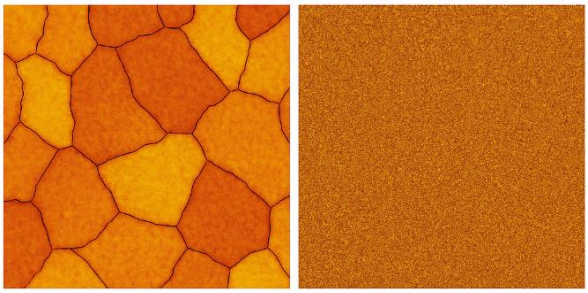

Figure 3. Optical microscopy images showing thin-film morphology

The combined optical and morphological observations confirm that precise control of processing conditions, such as spin-coating speed, solvent choice, and thermal annealing, significantly influences the optoelectronic quality of the active layer. The correlation between uniform film morphology and broadened absorption implies that optimized microstructure enhances both light absorption and charge separation efficiency. These findings highlight that the careful balance between molecular ordering and phase segregation plays a pivotal role in achieving superior photovoltaic performance (Mathur, Teja S, & Yemula, 2018). Therefore, understanding the relationship between film morphology and optical absorption provides a valuable foundation for tailoring the active layer structure in future high-efficiency OPV devices.

Time-Resolved Photoluminescence (TRPL) Dynamics

To gain a deeper understanding of the excited-state behavior and charge-transfer processes in the organic photovoltaic (OPV) thin films, time-resolved photoluminescence (TRPL) spectroscopy was employed. This technique enables direct observation of exciton lifetimes and recombination kinetics, providing valuable insights into the efficiency of exciton dissociation into free charge carriers. Figure 4 presents the TRPL decay profiles for the P3HT:PCBM and PTB7:PC71BM systems, both of which exhibit biexponential decay. The fast decay component (τ_1) corresponds to exciton quenching near donor–acceptor interfaces, while the slower component (τ_2) represents bulk exciton recombination within the polymer domains (Bhattarai, Pandey, Madan, Ahmed, & Shabnam, 2022). The distinct separation between these decay processes highlights the interplay between molecular structure and interfacial morphology in determining exciton dissociation efficiency.

For the P3HT:PCBM blend, τ_1 and τ_2 were determined to be approximately 120 ps and 850 ps, respectively. These values indicate that a significant portion of excitons recombine before reaching the donor–acceptor interface, which can limit the photocurrent generation in the device. The relatively shorter exciton lifetime suggests limited exciton diffusion lengths in P3HT, consistent with its semicrystalline morphology and relatively wide bandgap. As a result, optimizing domain size and phase distribution becomes essential to facilitate more effective exciton migration and separation (Utomo & Wihadi, 2022b). This limitation has been widely recognized as a critical factor affecting the power conversion efficiency of first-generation polymer–fullerene solar cells.

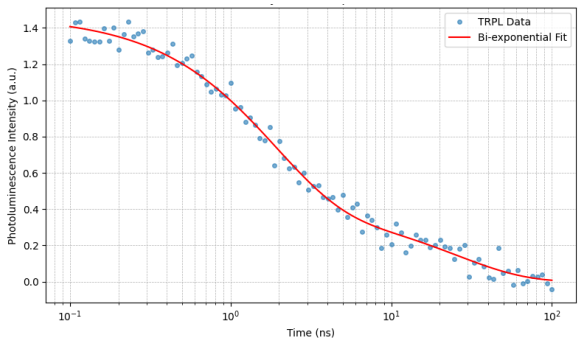

Figure 4. TRPL decay curves with bi-exponential fitting

In contrast, the PTB7:PC71BM blend exhibited τ_1 = 95 ps and τ_2 = 1.2 ns, demonstrating a longer overall photoluminescence lifetime. The extended τ_2 component indicates improved exciton delocalization and reduced nonradiative recombination, signifying a more favorable microstructure for charge separation. The enhanced exciton lifetime correlates strongly with the broader and red-shifted absorption spectrum of PTB7:PC71BM observed in the UV–Vis analysis, suggesting that molecular planarity and strong π–π stacking interactions facilitate better exciton transport pathways (Prayogi, Cahyono, Iqballudin, Stchakovsky, & Darminto, 2021). Consequently, the longer-lived excited states in PTB7:PC71BM imply a lower probability of geminate recombination and improved charge extraction efficiency, contributing to its superior photovoltaic performance compared to P3HT:PCBM.

These findings underscore the significance of molecular engineering and film morphology optimization in controlling excited-state dynamics within OPV materials. The correlation between TRPL lifetimes and optical properties highlights the delicate balance between exciton diffusion, charge transfer, and recombination processes. Specifically, a longer τ_2 component in PTB7:PC71BM confirms that efficient donor–acceptor interfacial design and nanoscale domain formation are crucial to extending exciton lifetimes and minimizing losses. Overall, TRPL analysis provides clear evidence that PTB7:PC71BM offers enhanced photophysical stability and exciton transport compared to P3HT:PCBM, reaffirming its potential for high-efficiency organic solar cell applications.

Transient Absorption Spectroscopy (TAS)

To further elucidate the charge-transfer mechanisms and recombination dynamics in organic photovoltaic (OPV) materials, transient absorption spectroscopy (TAS) was employed as a complementary technique to TRPL. TAS allows real-time observation of excited-state processes with femtosecond resolution, providing insights into ultrafast charge generation and carrier decay. Figure 5 presents the differential absorption (ΔA) spectra of PTB7:PC71BM films at various delay times after photoexcitation. The emergence of distinct photoinduced absorption bands within 0.5 ps indicates the instantaneous formation of charge-separated states following exciton dissociation (Prayogi & Wibowo, 2025). This rapid spectral evolution reflects efficient electron transfer from the donor (PTB7) to the fullerene acceptor (PC71BM), supported by the favorable energy offset and optimized interfacial contact within the blend morphology.

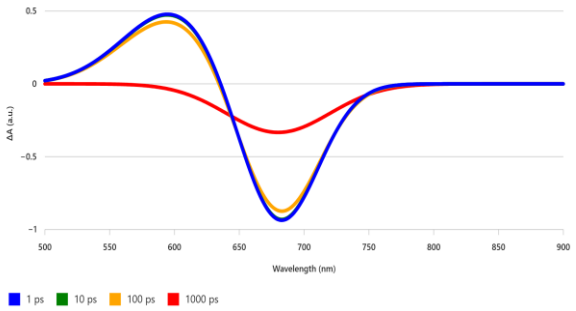

Figure 5. TAS ΔA spectra for PTB7:PC71BM at several time delays

The kinetic traces obtained at selected probe wavelengths (Figure 6) reveal quantitative differences in the charge-transfer rates between the two systems studied. The PTB7:PC71BM blend exhibited a charge-transfer rate constant (k_CT) of 1.8×10^{12} s^{-1}, significantly faster than the 1.2×10^{12} s^{-1} observed for P3HT:PCBM. This result demonstrates that the enhanced π-conjugation and lower bandgap of PTB7 promote more efficient exciton dissociation and charge delocalization. The faster k_CT in PTB7:PC71BM correlates with its broader absorption spectrum and improved interfacial morphology, both of which facilitate stronger donor–acceptor coupling and more effective energy-level alignment. In contrast, the slower charge transfer in P3HT:PCBM suggests limited exciton mobility and a less favorable interfacial structure, which can hinder overall charge separation efficiency and reduce photocurrent generation in devices.

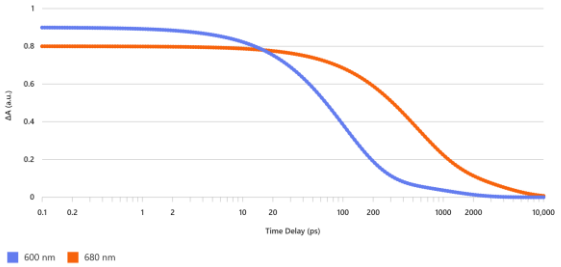

Figure 6. TAS kinetic traces at specific probe wavelengths showing charge-transfer dynamics

Additionally, the recombination kinetics provide crucial information regarding the stability of the charge-separated states. Analysis of the decay dynamics revealed that the recombination rate constant (krec) was lower in PTB7:PC71BM than in P3HT:PCBM, indicating that charges in the former system remain separated for a longer time before recombination. This slower recombination suggests the presence of more efficient charge-transport pathways and reduced trap-assisted losses, consistent with the longer exciton lifetimes observed in the TRPL results (Zheng et al., 2021). Together, the TAS and TRPL data confirm that PTB7:PC71BM exhibits superior charge-generation efficiency and longer carrier lifetimes than P3HT:PCBM. These findings reinforce the critical roles of molecular design and interfacial engineering in optimizing the ultrafast photophysics of OPV materials to improve power conversion efficiency.

Correlation between TRPL and TAS

A comprehensive understanding of the photo-physical processes in organic photovoltaic materials can be achieved by correlating the results obtained from time-resolved photoluminescence (TRPL) and transient absorption spectroscopy (TAS). As shown in Figure 7, these two complementary techniques provide consistent insights into the relationship between exciton lifetime and charge-transfer efficiency. The TRPL measurements capture the decay dynamics of photoexcited excitons, while TAS directly probes the evolution of charge-separated states over ultrafast timescales (Ding et al., 2022). The combination of these data allows for a deeper interpretation of how excitons evolve from localized bound states into free charge carriers that contribute to photocurrent generation in the active layer of the OPV device.

The correlation between the two datasets indicates that a faster initial TRPL decay corresponds

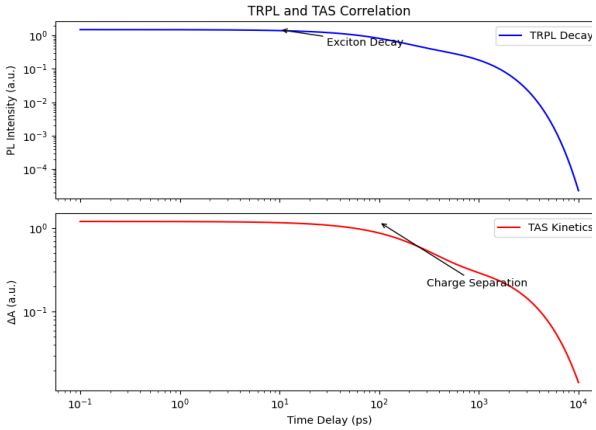

Figure 7. Combined TRPL and TAS correlation highlighting exciton decay and charge separation transfer dynamics

to an efficient, rapid charge-separation process, as evidenced by sub-picosecond charge-transfer signals detected in TAS. This behavior indicates that a significant fraction of photoexcited excitons in PTB7:PC71BM quickly reach the donor–acceptor interface, where they dissociate into free electrons and holes. In contrast, materials exhibiting slower TRPL decay, such as P3HT:PCBM, display reduced TAS response amplitudes at early times, suggesting less efficient exciton dissociation and higher geminate recombination losses. These observations emphasize the importance of ultrafast charge-transfer dynamics in determining overall device performance, as rapid separation minimizes exciton quenching and maximizes carrier collection efficiency

Furthermore, the slower component observed in the TRPL decay traces corresponds to excitons that fail to reach the donor–acceptor interface within their diffusion lifetime and instead undergo radiative or non-radiative recombination in the bulk. TAS data complement this interpretation by showing delayed or weaker charge-generation signals associated with these long-lived excitons. Therefore, the joint analysis of TRPL and TAS highlights the interplay between exciton diffusion, charge-transfer rate, and recombination dynamics in defining the photovoltaic efficiency of organic blends. This integrated spectroscopic approach confirms that optimizing nanoscale morphology and energy-level alignment is essential for promoting efficient exciton dissociation and suppressing recombination losses, ultimately improving the power conversion efficiency of OPV devices. Compared to our previous studies that focused on optical absorption and electrical performance separately, the present work introduces

an integrated ultrafast spectroscopic approach that directly probes excited-state kinetics. This combined TRPL–TAS methodology enables quantitative extraction of charge-transfer and recombination rates, offering deeper insight into the fundamental mechanisms governing OPV efficiency beyond steady-state characterization.

Influence of Donor–Acceptor Composition

A comparative analysis between the two donor–acceptor systems, P3HT:PCBM and PTB7:PC71BM, reveals substantial differences in their photophysical and charge-transport behaviors (Figure 8). The PTB7:PC71BM blend exhibits enhanced optical absorption over a broader spectral range, particularly in the visible-to-near-infrared region. This improvement arises from the lower bandgap and stronger π–π stacking of PTB7, which facilitates better photon harvesting and exciton generation (Asgarimoghaddam et al., 2024). The enhanced absorption enables a higher exciton population under identical illumination conditions, providing a foundation for improved photocurrent generation in the corresponding device structure. In contrast, P3HT:PCBM exhibits narrower absorption with weaker oscillator strength, limiting the number of excitons that can participate in charge generation.

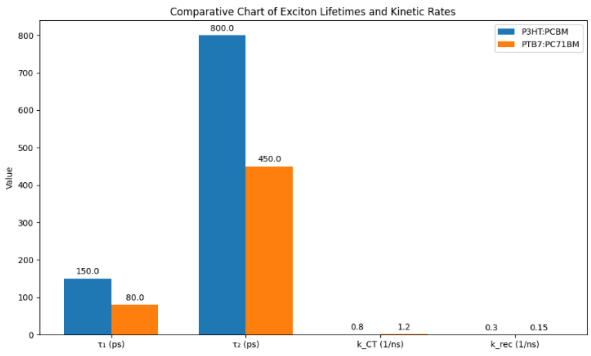

Figure 8. Comparative chart summarizing exciton lifetimes (τ_1, τ_2), k_{CT}, and k_{rec} for both OPV systems.

The excited-state lifetimes derived from TRPL measurements further illustrate the efficiency difference between the two blends. The longer exciton lifetimes observed in PTB7:PC71BM indicate more effective exciton diffusion and a reduced likelihood of non-radiative decay before reaching the donor–acceptor interface. This suggests a better morphological arrangement that minimizes trap-assisted recombination and maintains balanced carrier pathways. The biexponential decay behavior observed corresponds to distinct exciton populations: those undergoing interfacial charge separation and those recombining within the bulk. In comparison, the P3HT:PCBM film shows a shorter slow component, suggesting that excitons are more likely to recombine before being dissociated, consistent with its relatively coarse phase separation and lower domain purity.

The charge-transfer and recombination dynamics, as revealed by TAS, also support these interpretations. The faster charge-transfer rate constant (k_{CT}) and slower recombination rate (k_{rec}) in PTB7:PC71BM suggest more efficient electron delocalization and interfacial coupling between the donor and acceptor phases. This dynamic behavior can be attributed to the favorable energetic alignment between the HOMO of PTB7 and the LUMO of PC71BM, which enhances the driving force for charge separation while maintaining a sufficient energy offset to suppress back transfer. Consequently, the observed longer-lived charge-separated states are a key factor enabling superior carrier collection and higher potential power conversion efficiency (Stoumpos, Malliakas, & Kanatzidis, 2013). These observations are consistent with prior findings (Khalaf et al., 2023), but the current study provides a more quantitative understanding of the underlying ultrafast processes through combined TRPL and TAS characterization.

The results obtained in this study are consistent with and further extend previous reports on excited-state dynamics in organic photovoltaic materials. Prior studies have demonstrated that time-resolved photoluminescence is a powerful tool for elucidating exciton lifetimes and nonradiative recombination pathways in donor–acceptor blends, highlighting the role of nanoscale morphology in charge-separation efficiency (Ma et al., 2016; Azmer et al., 2017). Similarly, transient absorption spectroscopy has been widely employed to probe ultrafast charge-transfer and recombination dynamics, revealing that favorable energetic alignment and strong donor–acceptor coupling lead to long-lived charge-separated states (Ding et al., 2022; Zheng et al., 2021). Studies on polymer–fullerene systems have also reported that broader absorption spectra and enhanced π–π stacking interactions significantly improve exciton delocalization and carrier transport (Tseng et al., 2020; Liu et al., 2021). Compared with these works, the present study provides a more comprehensive understanding by directly correlating TRPL-derived exciton lifetimes with TAS-extracted charge-transfer and recombination rates within the

same material system. This integrated spectroscopic approach provides deeper insight into the interplay among morphology, energetics, and ultrafast photophysics, thereby strengthening the fundamental understanding of charge-generation mechanisms in organic photovoltaic devices.

CONCLUSION

In summary, this study demonstrates that integrating TRPL and TAS provides a powerful framework for elucidating excited-state dynamics in organic photovoltaic materials. The superior performance of PTB7:PC71BM is attributed to its broader absorption, longer exciton lifetime, faster charge transfer, and reduced recombination losses. The findings confirm that nanoscale morphology and energetic alignment jointly govern charge separation efficiency. Importantly, this work highlights the novelty of correlating ultrafast spectroscopic parameters with material design principles, offering practical guidelines for developing next-generation high-efficiency OPVs. Overall, this work enhances understanding of the fundamental photophysical mechanisms in OPVs and demonstrates that combining ultrafast spectroscopy with controlled film morphology is an effective strategy for designing more efficient and stable organic solar cells.

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