

RESEARCH ARTICLE

Aggregation Regulated Ultrafast Singlet Fission Pathways in TIPS-Pentacene Films

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Singlet fission (SF) is a spin-conserving process converting 1 singlet exciton into 2 triplet excitons. This exciton multiplication mechanism offers an attractive route to solar cells that circumvent the single-junction Shockley–Queisser limit. However, it remains unclear how intermolecular coupling, which is subject to the aggregation extent in thin-film morphology, controls SF pathways and dynamics. The prototype molecule 6,13-bis(triisopropylsilylethynyl)-pentacene (TIPS-pentacene) has been extensively studied to investigate SF mechanisms. However, previous literature reports have presented divergent SF mechanisms and pathways in TIPS-pentacene films. In this study, solvent vapor annealing treatment is used to deliberately adjust the aggregation extent in TIPS-pentacene films. This enables us to reproduce various SF pathways reported in the literature under the same experimental conditions, with the only variation being the level of aggregation. These results shed light on the crucial role that molecular aggregation plays in modulating both the SF mechanism and pathway and reconciles the previously contentious SF mechanisms and pathways reported in TIPS-pentacene films. Our study offers substantial insights into the understanding of the SF mechanism and provides a potential avenue for future control of SF pathways in accordance with specific application requirements.

Introduction

Singlet fission (SF) is an intriguing photophysical process that has attracted substantial attention in recent years due to its potential to boost the efficiency of solar cells beyond the Shockley–Queisser limit [1–4]. In SF, a high-energy excited singlet state undergoes a spin-allowed transition to generate 2 low-energy triplet states, effectively doubling the yield of exciton generated per absorbed photon [5–7]. SF has been observed in various acene-based organic semiconductors, such as pentacene and its derivatives [8–13]. Among these derivatives, 6,13-bis(triisopropylsilylethynyl)-pentacene (TIPS-pentacene) emerges as a remarkable prototype for investigating the SF mechanism and pathways [14–19]. Apart from its high solubility and ease of processing, the bulky triisopropylsilylethynyl substituents introduce steric hindrance that increases the spacing between adjacent TIPS-pentacene molecules, facilitating the SF process. In addition, the exoergic nature of SF in TIPS-pentacene allows for the temporal separation of multiple reaction steps, facilitating the measurement of consecutive processes [20,21].

Extensive studies have been conducted on TIPS-pentacene films for SF investigations, using various ultrafast spectroscopic techniques [22–27]. However, the SF mechanism and pathways in TIPS-pentacene film remain a subject of ongoing debate. Currently, it is widely accepted that the photoexcited singlet

state undergoes a conversion to a multiexciton or correlated triplet pair state ¹(TT) characterized by a singlet spin configuration. Subsequently, this correlated triplet pair state dissociates into 2 free triplet states [27–30]. Nevertheless, recent studies have reported the existence of a long-lived bound triplet pair state in the near-infrared spectral region. Instead of undergoing ultrafast dissociation into free triplet excitons, this bound triplet pair experiences prolonged nonradiative decay back to the ground state directly [7,31,32]. The efficiency of SF is highly influenced by the aggregation extent or molecular coupling within the film. Some studies suggested that stronger intermolecular coupling can enhance the transportation of triplet states [33–36]. However, other research demonstrated that excessive molecular coupling can expedite the spin-allowed electronic relaxation process, thereby limiting the overall yield of triplet excitons [31,37,38]. TIPS-pentacene exhibits the formation of multiple polymorphs in films, each possessing distinct spectroscopic characteristics [29,39–41]. Notably, there is substantial variation in film morphology documented in the existing literature using different film treatment methods. Specifically, studies observed that the absorption spectra of TIPS-pentacene films fabricated via spin coating closely resemble that of the solution, with the 0–0 transition occurring at approximately 650 nm, indicating an amorphous phase morphology [11,32,42–45]. Conversely, studies using solvent vapor annealing (SVA) treatment

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have observed a red-shifted absorption at 700 nm, which is commonly associated with the spectral signature of aggregation formation that induces a crystalline phase morphology in TIPS-pentacene film [7,41,46]. So far, the relationship between the extent of aggregation in the investigated TIPS-pentacene films and the proposed variations in SF mechanisms remains unclear. Therefore, further research is warranted to enhance our understanding of how aggregation controls the SF mechanism and pathways in TIPS-pentacene films, particularly its impact on the formation and dynamics of triplet and triplet pair states.

In this study, we meticulously manipulate the extent of aggregation in TIPS-pentacene films through SVA for varying durations. Femtosecond transient absorption (TA) spectroscopy is used to systematically characterize the spectral and temporal dynamics of the excited states involved. By varying the aggregation extent in TIPS-pentacene films via SVA treatment, we successfully reproduced various SF pathways reported in the literature under identical experimental conditions. Global analysis was performed on the obtained TA spectra to extract the key spectral and kinetic signatures. The analysis results revealed the various SF pathways in TIPS-pentacene films, which were modulated by the extent of aggregation. Our study provides compelling evidence that the extent of aggregation plays a critical role in shaping the SF pathway and dynamics. These outcomes will substantially contribute to a more comprehensive understanding of the general SF mechanism in other chromophores and pave the way for the application of SF in multiple directions.

Materials and Methods

Materials

TIPS-pentacene powder was purchased from Aladdin Biochemical Technology Co. Ltd. (purity > 98%). The chlorobenzene solution was obtained from Macklin Biochemical Co. Ltd. (purity > 99.5%), the dichloromethane solution was acquired from Zhiyuan Chemical Reagent Co. Ltd. (purity > 99.9%), and the isopropyl alcohol (IPA) solution was obtained from Guanshi Reagent Technology Co. Ltd. (purity > 99.7%). All chemicals were utilized without further purification, unless otherwise specified. The glass substrates were cleaned in a 1% Hellmanex III solution for 10 min and then in an IPA solution for 10 min, followed by a 30-min treatment in a ultraviolet (UV)-ozone cleaner. TIPS-pentacene in a 1:1 mixed solution of chlorobenzene and dichloromethane (concentration of 20 mg·ml⁻¹) was stirred for 8 h and subsequently passed through a filter with 220-nm pores directly into a clean sample vial. The solution was subsequently spin-coated onto the precleaned glass substrate at a speed of 5,000 rpm for a duration of 30 s. The as-prepared TIPS-pentacene films were then placed in an IPA vapor atmosphere for various durations of 3, 6, 9, and 12 min for subsequent spectral characterizations. For comparison, the pristine TIPS-pentacene film from the same batch was placed on a hot plate and completely annealed for 30 min.

Steady-state characterization

The steady-state absorption spectra of TIPS-pentacene film were measured by a fiber optic spectrometer from Flight Technology Co. Ltd. The morphology of TIPS-pentacene films was measured by scanning electron microscopy (SEM) with a Hitach SU8220 SEM (Hitach; Tokyo, Japan). To provide the conductive layer necessary for imaging, a thin layer of gold was the first supported

on the surface of TIPS-pentacene films with automatic gold and carbon plating instrument (Cressington 108auto).

TA spectroscopy

TA spectra were measured using the TA spectrometer, which was equipped with a Spectra-Physics Solstice Ace regenerative amplifier (~100 fs pulses with 1 kHz repetition rate) for the probing beam and with the TOPAS light convertor for the pumping beam. The TA was also modified to suppress all the coherent artifacts appearing in the subpicosecond time scale. We used optical pumping at 650 nm and optical probing exploiting white light supercontinuum generated in a sapphire plate. All measurements were performed in air and at room temperature using a cross-linear-polarized geometry [the pumping and probing beams were polarized out-of plane (vertical) and in-plane (horizontal) of incidence, respectively]. The measurements were carried out at environmental condition.

Results and Discussion

Steady-state characterization

SVA was used to treat the TIPS-pentacene films to systematically tune the extent of molecular aggregation. As depicted in Fig. 1A, the pristine spin-coated TIPS-pentacene films were placed in an IPA atmosphere at room temperature for varied durations, with a time gradient of 3 min. Figure 1B illustrates the observed changes with increasing SVA treatment duration. The film color turns darker, and the SEM images demonstrate an improvement in crystallinity. The crystalline signature with a strip texture reminiscent of the thermally annealed TIPS-pentacene film (Fig. S1) gradually appear as the SVA treatment duration increases, indicating that SVA treatment reorganizes the molecules resulting in the enhanced molecular aggregation within the TIPS-pentacene film [14].

The steady-state absorption spectra of corresponding films also exhibit apparent alterations (Fig. 1C). The pristine TIPS-pentacene film exhibits spectral features resembling that of the solution. The broadband vibronic transitions ranging from 500 to 750 nm correspond to the $S_0 \rightarrow S_1$ transition, and electronic transitions at 440, 420, and 350 nm correspond to the upper excited state transitions of $S_0 \rightarrow S_3$, $S_0 \rightarrow S_4$, and $S_0 \rightarrow S_8$ respectively [32,44,47]. These spectral and structural features are indicative of amorphous phase films [14]. With increasing SVA treatment duration, a substantial redshifted aggregate feature is observed in the absorption spectra. Specifically, the absorption band at 700 nm exhibits growth, while other bands diminish and broaden. In addition, the baseline at shorter wavelengths experiences a substantial increase, which aligns with the increased Rayleigh scattering attributed to larger aggregated particles [48]. These spectral modifications provide a distinctive indication that the SVA treatment increases the extent of aggregation and, thus, the intermolecular coupling in the TIPS-pentacene film, facilitating an amorphous to crystalline film transition [14].

TA spectra

Excited-state absorption

Femtosecond TA spectra were carried out to investigate the excited state dynamics in TIPS-pentacene films under excitation at 650 nm, which corresponds to the lowest vibrational transition of $S_0 \rightarrow S_1$ in TIPS-pentacene. The top panel of Fig. 2 presents the pseudo-color 2-dimensional (2D) plots of TA

spectra for SVA-treated TIPS-pentacene films. In this study, our primary focus is to examine the influence of SVA-treatment-induced aggregation on the SF mechanism. For reference, TA spectra of the as-prepared spin-coated film (SVA, 0 min) and the thermally annealed film have been provided in the Supplementary Materials (Fig. S2). The negative ΔA signal in TA spectra corresponds to ground-state bleaching (GSB), and the positive ΔA signal corresponds to excited state absorption (ESA) or photoinduced absorption. As shown in Fig. 2, in the amorphous phase films with SVA duration of 0 to 6 min, a pronounced ESA signal is observed at 508 nm. This signal undergoes a redshift of 15 to 523 nm in the crystalline phase films with SVA durations exceeding 6 min. In addition, there are notable ESA at 455 and 575 nm in the amorphous phase films, which become less prominent in the crystalline phase films. These spectral characteristics are attributed to the absorption

originating from excited states with different multiplicities [43,49,50]. Specifically, the ESA signal at 508 or 523 nm is attributed to the absorption from triplet states generated through the SF process ($T_1 \rightarrow T_n$), while the ultrafast ESA signal between 430 and 490 nm corresponds to the singlet excited state transition ($S_1 \rightarrow S_n$) [11,24,42,50]. Furthermore, in the TA spectra of the crystalline phase films with SVA duration over 6 min, a near-infrared ESA band appears, spanning the spectral range from 730 nm to greater than 870 nm. This band was attributed to the presence of a long-lived correlated triplet pair state [7,32,44,51]. To differentiate it from the intermediate triplet pair state ${}^1(TT)$ that dissociates into T_1 , we denote the long-lived triplet pair as $(TT)_L$. Previously, literature has demonstrated the existence of 2 variants of triplet pairs. One undergoes an ultrafast conversion into 2 triplet states, designated as $(TT)_a$, while the other persists for a longer duration and undergoes

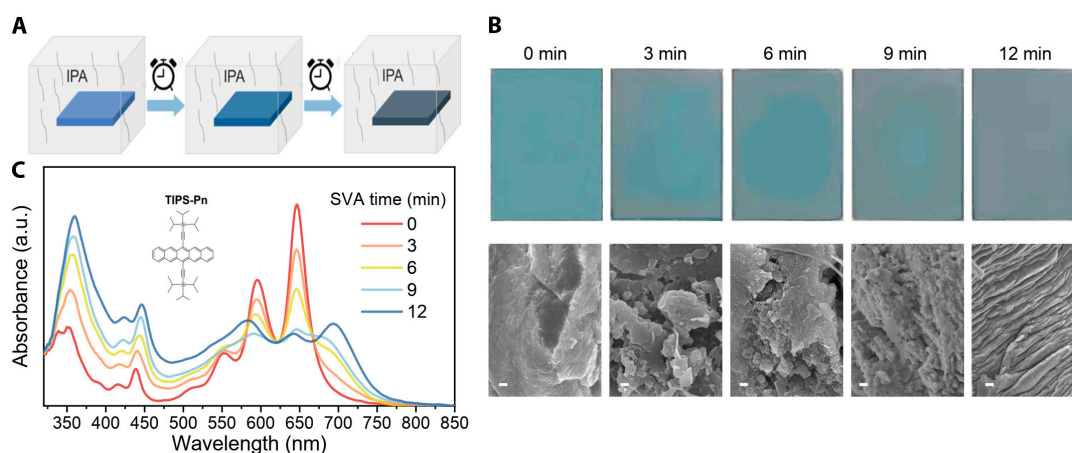


Fig. 1. (A) Schematics of SVA process. (B) Photographs of TIPS-pentacene films with various SVA treatment durations (top) and their corresponding SEM images (bottom). Scale bars, 2 mm. (C) Steady-state absorption spectra of SVA-treated TIPS-pentacene films. a.u., arbitrary units.

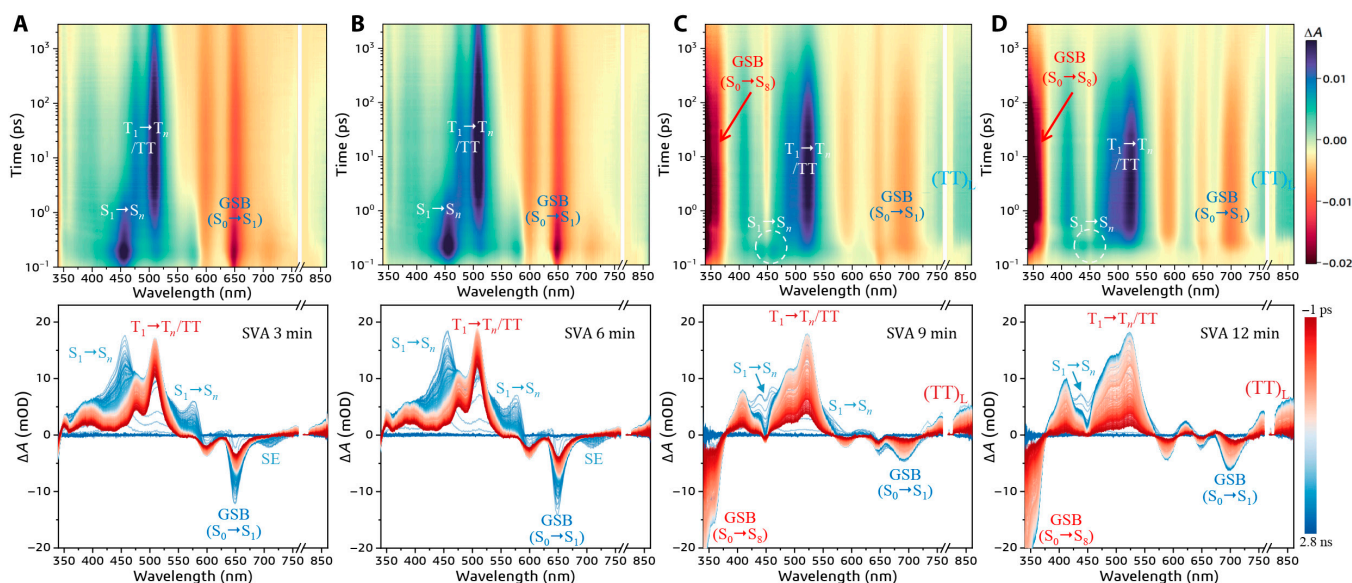
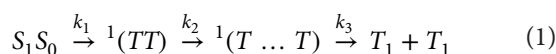
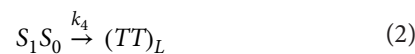


Fig. 2. 2D pseudo-color maps (top) and TA spectra (bottom) of TIPS-pentacene films with SVA treatment durations of (A) 3 min, (B) 6 min, (C) 9 min, and (D) 12 min, under excitation wavelength at 650 nm.

direct recombination back to the ground state, denoted as $(TT)_b$ [32,44]. Our observed long-lived triplet pair $(TT)_L$ corresponds to the $(TT)_b$ in sense. The kinetic traces at representative probe wavelengths in each sample and corresponding exponential fitting results are presented in Fig. S3 and Table S1. Both TA spectra and kinetics in these TIPS-pentacene films exhibit a high level of complexity, which arises from the substantial spectral overlap and the involvement of multiple intermediate states. To unravel the intricate SF mechanism and pathways, we utilize the global target fitting approach to obtain species-associated spectra and dynamics. For the amorphous phase films with SVA durations of 0 to 6 min, the following sequential kinetic model is used:



where S_1S_0 represents an individual molecule in its primary excited singlet state (S_1) and its adjacent molecule in its ground state (S_0), ${}^1(TT)$ denotes the intermediate correlated triplet pair, ${}^1(T \dots T)$ represents a spatially separated yet still spin-correlated triplet pair, and T_1 refers to an independent triplet state [21]. The species-associated spectra and kinetics reconstructed from the global fits are presented in Fig. 3A and B. The rate constants obtained for the SVA treatment of 3 and 6 min are very similar, with the values of $1/k_1 = 0.3$ (0.27) ps, $1/k_2 = 2.8$ (3.5) ps, and $1/k_3 = 276$ (263) ps. The values in parentheses correspond to the 6-min SVA treatment. The obtained absorption spectra of the intermediate triplet pairs, ${}^1(TT)$ and ${}^1(T \dots T)$, closely resemble that of the free triplet T_1 (Fig. 3A and B). In addition, as depicted in Fig. S4, the normalization of the extracted spectra of ${}^1(TT)$, ${}^1(T \dots T)$, and T_1 provides compelling evidence that as the correlation between the 2 triplet states intensifies, the absorption spectra of the correlated triplet pair state become broader and less structured. As mentioned above, in crystalline phase films with SVA durations of 9 to 12 min, alongside the formation of T_1 , a long-lived bound triplet pair is also generated, which undergoes direct relaxation back to the ground state. Therefore, a parallel kinetic pathway (Eq. 2) is introduced in addition to Eq. 1 for the global fitting analysis of the crystalline phase films.



It is also worth mentioning that the SF generated triplet state T_1 is much shorter lived in the crystalline phase films compared to the amorphous films. This can be attributed to the enhanced molecular packing in the crystalline phase, which promotes the triplet-triplet annihilation (TTA) process and accelerates the decay of triplets. In our previous study, we conducted a comprehensive investigation of the TTA process for both triplet and triplet pair states in TIPS-pentacene films [7]. The dynamics of both triplet and triplet pair state were found to be highly complex, requiring the consideration of the diffusion-limited TTA kinetic model for an accurate depiction. In this work, we only focus on TA spectra within the early 5 ps to extract the insights into the ultrafast SF mechanism and pathway while excluding the complexities arising from higher-order recombination dynamics associated with the TTA process. The obtained species-associated spectra and kinetics from the global fitting of the crystalline phase films are depicted in Fig. 3C and D. The rate constants obtained for the SVA treatment of 9 and 12 min are also similar, with the values of $1/k_{S_1} = 0.08$ (0.06) ps, $1/k_1 = 0.41$ (0.40) ps, $1/k_3 = 0.81$ (0.83) ps, and $1/k_4 = 0.1$ (0.07) ps. The values in parentheses correspond to the 12-min SVA treatment. Here, k_{S_1} represents the overall decay rate of S_1S_0 , taking into account both pathways described by Eqs. 1 and 2. The intermediate triplet pair conversion is too fast to be resolved in the crystalline phase films. Therefore, for our analysis, we only consider one intermediate ${}^1(TT)$ and disregard the term k_2 in Eq. 1. The extracted triplet-associated spectra exhibit a considerably broader and more featureless nature when compared to the amorphous phase films. This observation is consistent with aforementioned findings in the amorphous phase films, which indicate that the broader and structureless triplet pair absorption spectra can be attributed to the stronger coupling between the triplet states [21].

Comparing the TA kinetics in TIPS-pentacene films with different SVA treatment durations, a consistent trend was observed in the kinetics of triplet state. It was found that an increase in the extent of aggregation results in a corresponding

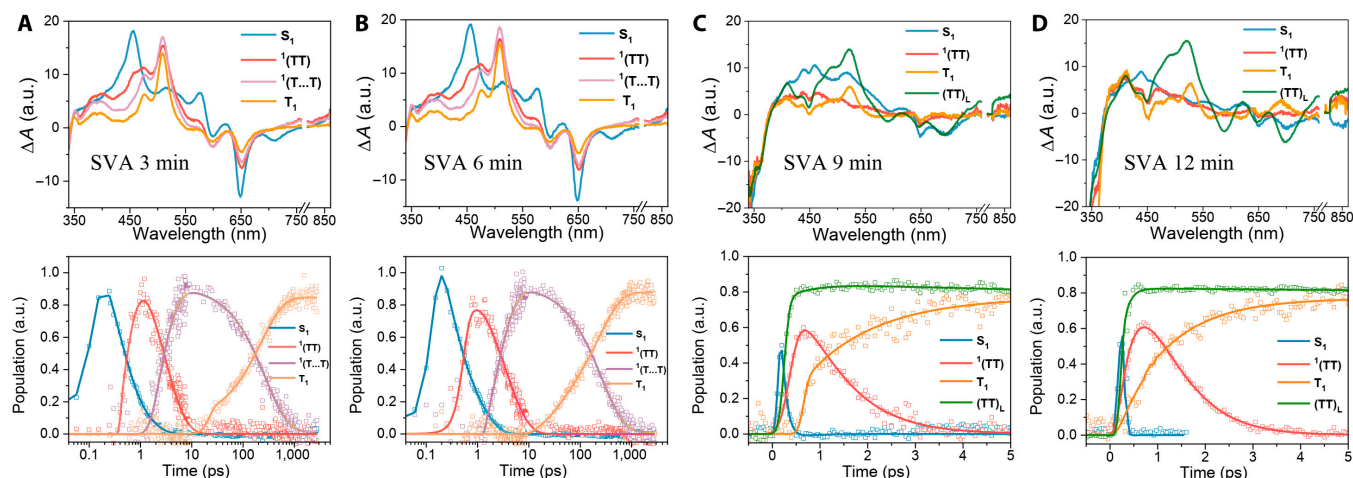


Fig. 3. Species-associated spectra (top) and kinetics (bottom) reconstructed from the global fit on the TA spectra of TIPS-pentacene films with different SVA treatment durations of (A) 3 min, (B) 6 min, (C) 9 min, and (D) 12 min, under excitation wavelength at 650 nm.

acceleration of both the formation and decay of T_1 , as illustrated in Fig. 4A. It suggests that a greater extent of aggregation enhances the formation of the triplet state through SF, while simultaneously reducing the lifetime of the triplet state due to the increased strength of intermolecular coupling, which promotes the TTA process. In the case of the long-lived triplet pair $(TT)_L$ (Fig. 4B), increased aggregation also accelerates its decay time, while the formation time did not show a noticeable influence. This indicates that the behavior of long-lived triplet pair, in relation to aggregation, show distinct difference from that of the free triplet state. These findings suggest that by meticulously controlling the extent of aggregation, it is possible to modulate the SF pathway and the resulting products.

Ground-state bleaching

In TA spectroscopy, the GSB signal originates from a decrease in the absorption of the probe beam due to the depletion of the ground-state population under pump excitation. Since both singlet and triplet excited states are supposed to bleach the ground state in the same manner, the GSB is expected to continue increasing after the pulse duration of pump beam. However, previous literature has indicated that the GSB signal does not always exhibit a rise time corresponding to SF process, which was usually attributed to the overlap with positive ESA signal or the delocalization of ground state [52,53]. In our study, in the amorphous phase films with SVA durations of 0 to 6 min, we observed the primary GSB signal corresponding to the $S_0 \rightarrow S_1$ transition (~ 650 nm) that exhibits an ultrafast decay consistent with the decay of S_1 ESA signal, as shown in Fig. S3. In addition, the GSB shoulder at ~ 600 nm shows a “rise” component that is most probably due to the overlap with the adjacent positive S_1 ESA decay. In contrast, in the crystalline phase films with SVA duration of 9 to 12 min, besides the GSB of $S_0 \rightarrow S_1$ transition (~ 700 nm), we also observed a distinct GSB signal in the UV region below 350 nm. This particular GSB signal has not been previously reported because of limitations in the probe range and the high absorption in thick samples. Comparing with the steady-state absorption spectrum, we attribute it to the GSB corresponding to $S_0 \rightarrow S_8$ transition [47]. Notably, the kinetics of this GSB signal in the upper transition range demonstrate remarkable differences when compared to the GSB signal associated with the lowest transition ($S_0 \rightarrow S_1$), as shown in Fig. S3. Hence, it is essential to delve into the intrinsic disparity between the GSB signals in the lowest ($S_0 \rightarrow S_1$) and upper ($S_0 \rightarrow S_n, n \geq 2$) transition ranges.

When the pump wavelength is in the lowest transition region ($S_0 \rightarrow S_1$), the population of the ground state (S_0) decreases while populating excited state S_1 . Therefore, the GSB signal encompasses the entire absorption spectrum covering the transitions of $S_0 \rightarrow S_1$ and $S_0 \rightarrow S_n$ ($n \geq 2$). The GSB signal of the $S_0 \rightarrow S_1$ transition arises from 2 contributing factors. First, it is a result of the depletion of the S_0 population due to the pump excitation. Second, it is influenced by the column screening effect caused by the formation of the S_1 exciton, which further reduces the oscillator strength of the $S_0 \rightarrow S_1$ transition. Therefore, the GSB signal of the $S_0 \rightarrow S_1$ transition are associated with the dynamics of both S_0 and S_1 states. In contrast, the GSB signal of the $S_0 \rightarrow S_n$ ($n \geq 2$) transition is only related to the population of the S_0 state since the upper excited states S_n are not directly populated under exciting S_1 state. In addition, since the recovery of S_0 state is associated with all excited-state relaxations, the GSB signal of the $S_0 \rightarrow S_n$ ($n \geq 2$) transition is inherently more suitable for analyzing the excited-state dynamics related to SF. Nevertheless, it is challenging to detect GSB signal associated with the $S_0 \rightarrow S_n$ ($n \geq 2$) transition. This difficulty arises from either the limited probe wavelength in the UV range in typical TA experiments or the high absorption coefficient in the UV region for most samples, which results in no transmission light available for detection. In this study, we extended the probe range down to 330 nm and decreased the thickness of the film to ensure the visibility of the GSB signal of $S_0 \rightarrow S_8$ transition centers at around 335 nm in crystalline phase TIPS-pentacene film. As elucidated previously, the crystalline phase films display intricate ESA kinetics in the early delay time, which involves ultrafast decay of singlet excited state S_1 , ultrafast formation of correlated triplet pair $^1(TT)$, and long-lived bound triplet pair $(TT)_L$, as well as the formation of free exciton T_1 . Therefore, we decompose the kinetics of GSB signal of upper transition $S_0 \rightarrow S_8$ using a set of dynamics corresponding to the dynamics of S_1 , $^1(TT)$, T_1 , and long-lived $(TT)_L$, as shown in Fig. 5B and C. Assuming that both triplet T_1 and long-lived triplet pair $(TT)_L$ generated via SF are supposed to bleach the ground state in the same manner, we can evaluate the efficiency of these 2 SF pathways by comparing the amplitude of the decomposed kinetics in the GSB of the upper transition. The analysis reveals an efficiency ratio of approximately 2:1 for SF pathway of generating T_1 and long-lived $(TT)_L$ in both crystalline phase films with SVA durations of 9 and 12 min.

On the basis of the above analysis, the aggregation modulation via SVA treatment in TIPS-pentacene film enables us to

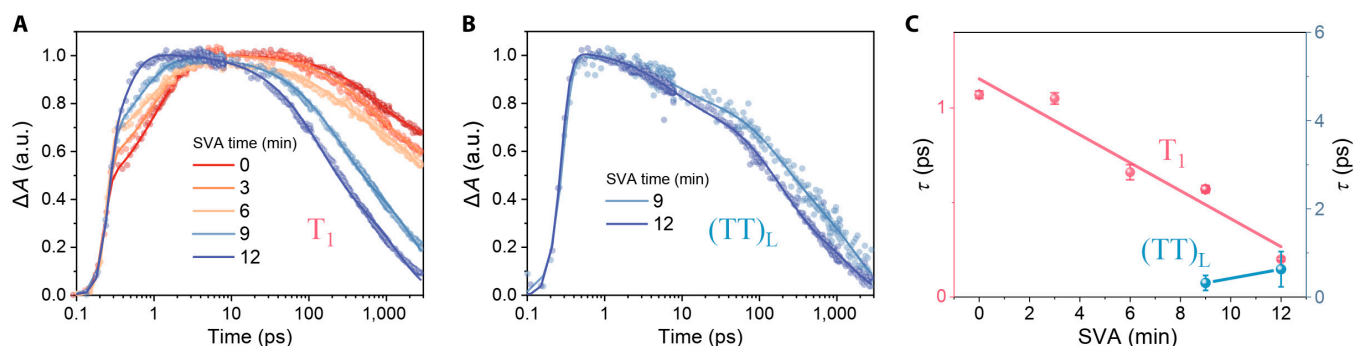


Fig. 4. (A) Normalized TA kinetic traces of (A) T_1 ($\lambda_{\text{probe}} = 508$ nm for SVA of 0 to 6 min, 523 nm for SVA of 9 to 12 min), (B) long-lived triplet pair $(TT)_L$ ($\lambda_{\text{probe}} = 845$ nm), and (C) fitted rise times in TIPS-pentacene film with various SVA treatment durations.

reproduce various SF pathways reported in the literature. This enables us to reconcile the previously contentious SF mechanisms and pathways by considering the substantial influence of molecular aggregation on the modulation of the SF process. Specifically, as shown in Fig. 6, in amorphous phase films with lower levels of aggregation, weaker molecular couplings give rise to a relatively slow formation of triplet states, occurring through multiple triplet pair intermediate states on the picosecond time scale. However, this weak coupling proves advantageous in prolonging the lifetime of free triplet states due to the reduced TTA process. In contrast, crystalline phase films characterized by stronger aggregation exhibit substantially faster formation of triplet states on the femtosecond time scale. However, the decay of triplet states is also accelerated because of the enhanced TTA resulting from strong molecular interactions [7]. Furthermore, the pronounced aggregation induces a parallel SF pathway through the generation of a long-lived bound triplet pair state that does not dissociate into free triplets but instead directly relaxes back to the ground state [40]. The existence of long-lived triplet states plays a pivotal role in extending the lifetime of excitons, which is crucial for the transfer and separation processes of excitons in photovoltaic applications. Notably, the TA spectra of TIPS-pentacene nanoparticles obtained in the mixed solvents of tetrahydrofuran and water

exhibited a strong resemblance to Fig. 2 in our study [43]. Thus, we believe that the findings derived in our study could be applicable to other aggregated systems obtained under different experimental conditions.

Conclusion

To sum up, in this study, the extent of aggregation in TIPS-pentacene film was deliberately adjusted using SVA treatment. By exclusively modulating the film aggregation extent while maintaining the same experimental conditions, we successfully replicated various SF pathways that had been previously documented in the literature. These pathways encompass the formation of free triplet states via multiple intermediate triplet pair states in less aggregated amorphous phase films, as well as the additional generation of long-lived triplet pair states in more aggregated crystalline phase films. Our results highlight the crucial role of molecular aggregation in modulating both the SF mechanism and pathways. By reconciling the previously controversial SF mechanisms and pathways reported in TIPS-pentacene films, our study offers substantial insights into the understanding of the SF mechanism and provides a potential avenue for accurate control of SF products based on specific application requirements.

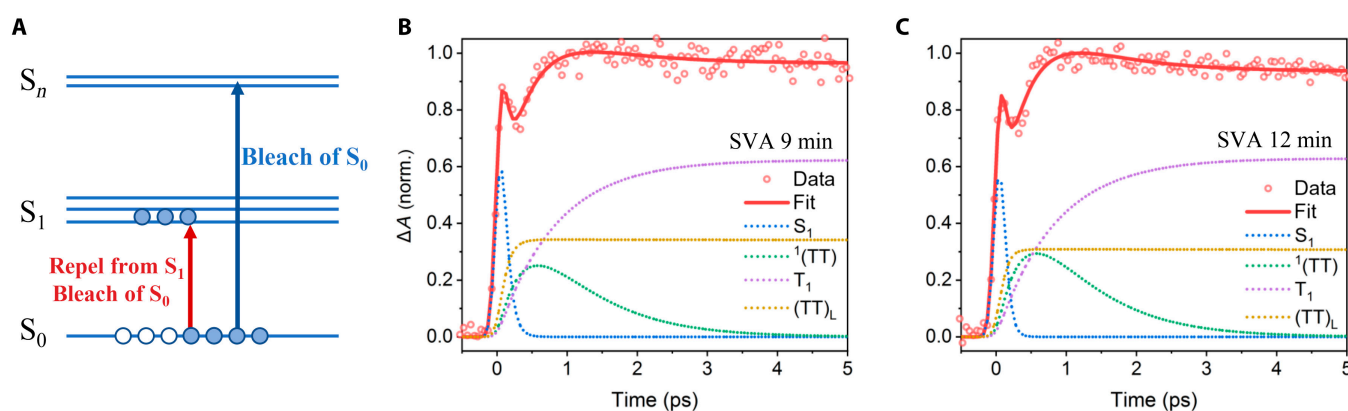


Fig. 5. (A) Schematics for GSB signal from upper and lower transitions. Normalized GSB signal at 350 nm and corresponding decomposed kinetics at upper transition levels of $S_0 \rightarrow S_8$ for crystalline phase films with SVA treatment duration of (B) 9 min and (C) 12 min.

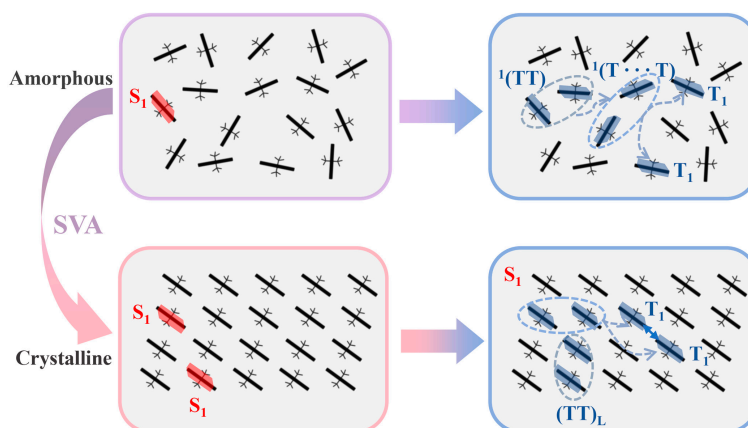


Fig. 6. Schematics of SF mechanisms and pathways in SVA-treated TIPS-pentacene films with varied aggregation extent.

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Author contributions: L.M. and T.H. designed the experiment. H.G., J.L., Z.Z. and W.K. conducted the experiments. F.Z., Y.Z. and G.L. did the data analysis. H.G., T.H. and L.M. wrote the article, with feedback from all the authors.

Competing interests: The authors declare that they have no competing interests.

Data Availability

The data are available from the authors upon a reasonable request.

Supplementary Material

Figs. S1 to S4
Table S1

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