Effect of the Pauli exclusion principle on the singlet exciton yield in conjugated polymers

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Optical devices fabricated using conjugated polymer systems give rise to singlet exciton yields which are high compared to the statistically predicted estimate of 25% obtained using simple recombination schemes. In this study we evaluate the singlet exciton yield in conjugated polymers systems by fitting to a model that incorporates the Pauli exclusion principle. The rate equations which describe the exciton dynamics include quantum dynamical components (both density and spin-dependent) which arise during the spin-allowed conversion of composite intra-molecular excitons into loosely bound charge-transfer (CT) electron-hole pairs. Accordingly, a crucial mechanism by which singlet excitons are increased at the expense of triplet excitons is incorporated in this work. Non-ideal triplet excitons which form at high densities, are rerouted via the Pauli exclusion mechanism to form loosely bound CT states which subsequently convert to singlet excitons. Our derived expression for the yield in singlet exciton incorporates the purity measure, and provides a realistic description of the carrier dynamics at high exciton densities.

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I. INTRODUCTION

The properties of singlet and triplet excitons (correlated electron-hole quasiparticles) in organic systems play primary roles in determining the electroluminescent quantum efficiencies of organic optical devices fabricated using π-conjugated polymers. Two fermions of spin $\frac{1}{2}$, give rise to four microstates: one singlet state (antisymmetric under particle exchange) and three triplet states (symmetric under particle exchange). Unlike the triplet exciton, the singlet exciton undergoes rapid radiative recombination producing light. Accordingly, the ratio of singlet to triplet exciton formation rates, $\gamma$, provides a useful quantitative measure of the electroluminescence efficiency in conjugated polymers. Based on the assumption that the formation cross sections of singlet and triplet excitons are the same, $\frac{1}{4}$ of available bi-fermion pairs (or charge carriers) will form singlet excitons with the well documented statistical yield of $\frac{1}{4} \gamma$ (25%). However several works have shown evidence of internal quantum efficiencies of 25% or more. To this end, the exact processes of spin dynamics that determine singlet exciton yield and the electroluminescence efficiencies are not fully resolved.

The conversion dynamics between the singlet and triplet excitonic states, related intermediate states which present as loosely bound electron-hole pairs, as well as the recombination lifetimes of all known states are fundamental to evaluating the yields of singlet exciton states. Several studies have examined the underlying mechanisms for the high singlet exciton yields (> 25%) observed in experimental results involving conjugated polymer systems. The decrease of a spin-dependent $\gamma$ was associated with strong spin-orbital coupling effects in an earlier work. By taking into account the optical interference effects on radiative rates, a higher singlet exciton yield (35%-45%) was obtained in conjugated polymers. The singlet exciton generation is lower in monomers compared to its generation in polymers. This may be linked to the fact that recombination dynamics is spin-independent in monomers, while it becomes spin-dependent in polymers. These differences highlight the dependence of exchange interactions and other electronic properties on the molecular configuration of the polymer chains. The energy relaxation process involving the singlet exciton is rapid unlike the slower relaxation via the low lying triplet exciton. This factor may contribute partly to differences in dynamics of the two spin states of the exciton. It appears that several underlying processes account for the variations in singlet exciton yield, which also depends on the material system under investigation. Thus far the inclusion of the Pauli-exclusion principle which becomes dominant at higher exciton densities has not been incorporated in all earlier works. The importance of the Pauli-exclusion principle at increasing exciton densities and its neglect in earlier works forms the key motivation for the current study.

While two or more ideal excitons can occupy the same quantum state, the multiple occupation of single-particle states is forbidden for several fermions by virtue of the Pauli-blocking mechanism. The rapid absorption of photons under intense illumination conditions excites multiple pairs of electrons and holes, generating closely spaced excitons. The decrease in distance between the optically generated electron-pair pairs, results in non-ideal bosonic excitons that acquire fermionic attributes. The ideal boson commutation relations which are applicable to the structureless exciton need to be modified to non-ideal commutation forms due to the operation of the Pauli exclusion principle.
While the exclusion principle operates under all conditions, it does not influence the dynamics of almost ideal excitons at very low densities.

The quantum statistical attributes of the coboson such as the composite exciton has received increased interest in recent works. These studies examined the links between entanglement and composite nature of bosons with non-ideal attributes. A key result is that quantum entanglement underpins the binding of constituent fermions to form the composite boson. While large entanglement between constituent fermions gives rise to a point-like structureless ideal boson, the less entangled electron-hole pair represents the non-ideal composite boson. Quantum entanglement of the fermions constitutes is thus critical to ensuring the bosonic attribute of the composite system remains intact.

In this work, the concept of entanglement is incorporated via the purity measure to examine quantum processes in organic materials. The purity measure is linked to the number of Schmidt modes associated with the entanglement of the paired structure of the exciton or any other species of composite boson. An exciton with zero or very small purity behaves as an ideal boson, while non-zero values of purity describe a non-ideal composite boson. A loosely bound electron-hole pair is associated with the less entangled electron and hole system with purity. In a system of many excitons, the purity quantifies the correlations between the fermions, with purity implying a highly correlated system of ideal excitons. For excitons occupying the same quantum state, the exciton can be considered a boson as long as the excitonic bosons are confined to spaces which are comparable to their sizes. In this study, we employ a composite boson model of the exciton to examine the singlet exciton yield at non-zero values of purity. The incorporation of entanglement aspects in this study will help overcome the shortcomings of conventional models used to model correlated system behavior in solids.

We consider the model of inter-chain electron-hole interaction between pairs of parallel polymers, where “charge-transfer” (CT) exciton states exist at intermediate energy levels positioned between the electron-hole continuum and the strongly bound exciton states. The singlet and triplet charge-transfer (CT) quasi-particles appear as critical precursors to the tightly bound exciton states in polymers. We extend this model by incorporating a mechanism in which tightly bound excitons convert to the more loosely bound CT intermediate states with loss of pure bosonic attributes. The conversion occurs due to operation of the Pauli exclusion principle, with the charge-transfer states possessing a higher degree of fermionic attributes.

The energetic difference between singlet and triplet exciton which arise from the opposite sign of the exchange interaction is generally high (≈ 0.7 eV) in conjugated polymers. The “charge-transfer” (CT) singlet and triplet states, on the other hand, are formed at approximately the same energy levels due to the weak exchange interactions of these states. Inter-conversion between the two types (singlet, triplet) of intermediate CT states may occur soon after their formation. Typical exciton lifetimes for organic materials such as PDHFV, PTEH and PDHFHPPV, the exciton lifetimes can range from 0.65 to 0.71 ns.

In this work, we consider that the Pauli exclusion operational times may proceed faster than the exciton lifetimes at high densities. The Pauli exclusion related conversion of one excitonic species to another becomes favorable over excitonic interactions with phonons or impurities with increased deviations in the bosonic attributes of excitons. Depending on the purity, the exciton density and the material system under study, Pauli exclusion based conversions are expected to proceed at times in the order of 0.01 to 1 ns. These processes are expectedly absent at low densities due to the ideal bosonic nature of the excitons. The results in this study are thus relevant to excitons which become non-ideal bosons as the excitonic purity is increased. In this study, we provide results of the singlet exciton yield for range of conversion times linked to the Pauli mechanism. The main results of this work is the derivation of an expression for the singlet exciton yield which incorporates the purity measure.

II. RATE EQUATIONS INCORPORATING THE PAULI EXCLUSION PRINCIPLE

In typical organic devices, the injection of charges carriers precedes the formation of excitons which diffuse for an amount of time before decaying radiatively (singlet exciton) or non-radiatively (triplet exciton). The overall external quantum efficiency of the organic devices is written as a product, \( \eta_f \eta_s \eta_p \eta_o \) where \( \eta_f \) is the efficiency of exciton formation from free carriers, \( \eta_s \) is the singlet exciton yield, \( \eta_p \) is the photoluminescence quantum efficiency and \( \eta_o \) is the fraction of of photons which are emitted without being utilized by the device. When the exciton density becomes high enough, the manifestation of the Pauli exclusion principle can be examined through \( \eta_f \) and \( \eta_o \), with incorporation of relevant device parameters. In this work, we focus on the effects of the Pauli exclusion principle on the underlying processes which determine \( \eta_s \). To evaluate \( \eta_o \), we consider a typical setup in which both singlet and triplet excitons
FIG. 1: Schematics of the energy levels \( E(i) \) of singlet \( (S_X) \) and triple \( (T_X) \) exciton states and singlet \( (S_{CT}) \) and triplet \( (T_{CT}) \) “charge-transfer” states. The conversion times within the singlet states \( (\tau_{SCT}) \) and triplet states \( (\tau_{TCT}) \) are indicated by the green arrows. The unequal times of inter-conversions between the spin CT states \( (\tau_{TS}, \tau_{ST}) \) are indicated by the blue arrows. The Pauli exclusion reversal times \( (\tau_{Tp}, \tau_{Sp}) \), which are present only at non-zero exciton purity \( p \), are denoted by the orange arrows, while the recombination times of the singlet and triplet excitons \( (\tau_{Sx}, \tau_{Tx}) \) are shown via the purple arrows. \( \Delta \) denotes the exchange energy between \( S_X \) and \( T_X \).

We consider the occurrence of Pauli exclusion reversal times \( \tau_{Tp} \) and \( \tau_{Sp} \) for the triplet and singlet exciton states respectively at non-zero exciton purity \((p_t, p_s)\). Depending on the degree of exciton purity or compositeness, the reversal times may proceed faster than the times of \( \tau_{TS}, \tau_{ST}, \tau_{Sx}, \tau_{Tx} \). The reversal times at non-zero exciton purity \( \tau_{Tp} \) and \( \tau_{Sp} \) are indicated by the upward arrows in Fig.1. Both these reversal times do not apply at low exciton densities, for which the recombination processes, \( S_{CT} \to S_X \) and \( T_{CT} \to T_X \) occur uninterrupted due to the almost ideal bosonic exciton. At high enough exciton densities where the purity assume non-trivial values, Pauli exclusion related processes become favorable compared to exciton decoherence due to interaction with phonons and/or impurities. The role of the times, \( \tau_{TS}, \tau_{ST}, \tau_{SCT}, \tau_{TCT}, \tau_{Tp} \) and \( \tau_{Sp} \) in enhancing the singlet exciton yield will be examined assuming the absence of coherence between the singlet and triplet CT states.

We first consider the simplified model with equal times of inter-conversions between the spin CT states, so that \( \tau_{TS} = \tau_{ST} = \tau_{ISC} \). The set of coupled equations relating the number of various quasiparticle states such as singlet
excitons ($N_{SX}$), triplet excitons ($N_{TX}$), singlet CT states ($N_{SCT}$) and triplet CT state ($N_{TCT}$) appear as

\[
\begin{align*}
\frac{dN_{SCT}}{dt} &= \frac{N_i}{4\tau} + \frac{N_{TCT}}{\tau_{ISC}} - N_{SCT} \left( \frac{1}{\tau_{ISC}} + \frac{1}{\tau_{SCT}} \right) + \frac{P_s N_{SX}}{\tau_{Sp}}, \\
\frac{dN_{TCT}}{dt} &= \frac{N_i}{2\tau} + \frac{N_{SCT}}{\tau_{ISC}} - N_{TCT} \left( \frac{1}{\tau_{ISC}} + \frac{1}{\tau_{TCT}} \right) + \frac{P_t N_{TX}}{\tau_{Tp}}, \\
\frac{dN_{SX}}{dt} &= \frac{N_{SCT}}{\tau_{SCT}} - \frac{N_{SX}}{\tau_{SX}} - \frac{P_s N_{SX}}{\tau_{Sp}}, \\
\frac{dN_{TX}}{dt} &= \frac{N_{TCT}}{\tau_{TCT}} - \frac{N_{TX}}{\tau_{TX}} - \frac{P_t N_{TX}}{\tau_{Tp}},
\end{align*}
\]

where $\frac{N_i}{\tau}$ is the initial number electron-hole pairs created per second. To reduce the number of coupled equations, we have used a single term ($N_{TCT}$) to denote the two possible triplet terms. In Eqs. $[1]$ to $[4]$ we have considered an exciton number dependent term which appears as

\[
\frac{p_i N_{jX}}{\tau_{jp}},
\]

where $N_{jX}$ denotes the number of singlet exciton ($N_{SX}$) or triplet excitons ($N_{TX}$). The purity measure $P_s$ ($P_t$) is associated with the singlet (triplet) non-ideal exciton. In order to obtain quantitative results in forthcoming sections, we consider that the Pauli exclusion reversal times $\tau_{Sp}$ and $\tau_{Tp}$ to be of the same order as the recombination times of the singlet and triplet excitons $\tau_{Sx}$, $\tau_{Tx}$. The form of Eq. $[5]$ is based on the lower bound of the normalization ratios associated with $(N + 1)$ and $N$ exciton which will be discussed next.

A. Pauli exclusion purity factors, $P_s$ and $P_t$

The purity measures, $P_s$ and $P_t$ in Eqs. $[1]$ $[4]$ of the non-ideal singlet and triplet excitons created from the “charge-transfer” states appear in the derived relation$^{27}$

\[
1 - P_i \cdot N \leq \frac{\chi^{N+1}}{\chi^N} \leq 1 - P_i,
\]

where $i = s$ (singlet) or $t$ (triplet), and $\chi$ is the normalization term linked to the superposed state of $N$ non-ideal excitons$^{23,27}$. The deviations from unity in the ratio, $\frac{\chi^{N+1}}{\chi^N}$ yield a measure of the non-bosonic quality of the correlated electron-hole pairs$^{23,24,27}$, establishes a link with with entanglement. The upper and lower bounds to $\frac{\chi^{N+1}}{\chi^N}$ in Eq. $[6]$ appear in terms of the purity $P_i$ of the single-fermion reduced state. The ratio $\frac{\chi^{N+1}}{\chi^N}$ can be interpreted as the effective probability of increasing the number of excitons from $N$ to $N + 1$ in a system. Excitons with ideal boson properties are described by an $N$ independent term $\chi$ so that $P_s = P_t = 0$. The lower bound in Eq. $[6]$ decreases monotonically with $N$, and vanishes at $P = \frac{1}{N}$.

The coherence between the $N$ excitons is decreased when the $(N + 1)$st exciton is added. This is due to the Pauli exclusion principle operating with the probability, $1 - \frac{\chi^{N+1}}{\chi^N}$. The importance of incorporating density dependent attributes, $P_i$ ($i = s, t$) in analyzing the singlet exciton yield in Eqs. $[1]$ to $[4]$ is highlighted by Eq. $[6]$. The density dependent attributes, $P_i$ ($i = s, t$) therefore has to be taken into account in any analysis of the singlet or triplet exciton yield under strong illumination conditions.

At high purities, the bounds in Eq. $[6]$ becomes less efficient$^{20,31,41}$ as the upper and lower bound terms differ significantly from each other. The purity $P$ alone is not sufficient to provide a reliable measure of the bounds in Eq. $[6]$. In this regard $P$ cannot be used to accurately predict the bosonic attribute of excitons. To further exploit the simplicity of Eq. $[6]$, we restrict our study to excitonic systems of small purity. For the purpose of illustrating the explicit role of the excitonic purity term, we consider an $N$ independent upper bound for the normalization ratio

\[
\frac{\chi^{N+1}}{\chi^N} \approx 1 - P_i
\]

which is applicable to a system of interacting excitons with minimal deviations from ideal bosonic attributes. This simple model is used to derive an analytical form for the exciton yield. Subsequently, we utilize a realistic numerical model which is applicable at small purity based on the relation

\[
P \approx \frac{\zeta}{N}
\]

where $0 < \zeta < 1$. We next employ Eqs. $[1]$ $[4]$ as well as Eqs. $[7]$ and $[8]$ to obtain quantitative results of the exciton yield.
B. Analytical form of the singlet exciton yield using Eq. 7

Using Eq. 7 which is applicable for system of weakly interacting bosons, and solving Eqs. 1 to 4 under the steady state conditions we obtain the singlet exciton yield \( \eta_s \) as

\[
\eta_s = \frac{N_{SX}/\tau_{SX}}{N_i/\tau} = \frac{\gamma + 3P t \alpha t + 3}{4(\beta + \gamma - \beta P_s \alpha_s - 2\gamma P_s \alpha_s - 2P_t \alpha_s - 2P_s P_t \alpha_s \alpha_t + P_t \alpha_t + 1)},
\]

where \( \beta = \tau_{SCT}/\tau_{TCT} \) and \( \alpha = \tau_{SCT}/\tau_{p} \). At \( P_s = P_t = 0 \), Eq. 8 reduces to the form

\[
\eta_s = \frac{3 + \gamma}{4(1 + \beta + \gamma)}
\]

which was earlier derived in Ref.8. In Fig. 2a, the singlet yield is obtained as a function of \( P_s = P_t = 0 \) at \( \beta = 0 \) (or \( \tau_{TCT} \gg \tau_{SCT} \)), and for equivalent Pauli reversal times to recombination times, \( \alpha_s = \alpha_p = 1 \). Fig. 2b shows the singlet yield as a function of \( \gamma = \tau_{SCT}/\tau_{TCT} \) at two different values of \( P_s = P_t = 1 \). We restrict the purity values to the range \( 0 \leq P \leq 0.15 \) in Figs. 2a, b as Eq. 9 holds valid only for small values of \( P \). The results in Figs. 2a, b highlight the importance of taking into account the Pauli exclusion mechanism for non-ideal excitons. A gradual increase in the singlet exciton yield occurs even with small increase in the purity values, \( P_s \) and \( P_t \). This increase in the singlet exciton yield is further enhanced at lower \( \gamma \). These preliminary results suggest that the spin-allowed route \( T_X \rightarrow T_{CT} \rightarrow S_{CT} \rightarrow S_X \) facilitates the increase in \( \eta_s \). These findings can be compared with those of earlier works8,19,20 where an increase in the singlet exciton yield is attributed to transfer of excitation from an intermediate triplet state rather than from the tightly bound triplet state.

The model used to derive results for Fig. 2a,b is only applicable for small deviations in bosonic attributes of the excitons. While we expect saturation in the singlet exciton yield to occur at higher \( P \) resulting in \( \eta_s \rightarrow 1 \), this cannot be predicted within the simple model used here. The usefulness of Eq. 9 is highlighted by the fact that it quantifies the mechanism by which there is increased proportion of singlet excitons through decrease in triplet exciton recombination. The triplet excitons are instead rerouted as a result of the Pauli exclusion principle to form loosely bound CT states which subsequently convert to singlet excitons. As a consequence, the fluorescence quantum yield can reach estimates of even 50% or more, which is the main result of this work.

C. Numerical solution for population of exciton species using Eq. 8

Using numerical techniques, we determine the population of all species, \( N_k \) \((k= SX, TX, TCT, SCT)\) using Eqs. 1 to 4 and substituting \( P_s \) and \( P_t \) with the \( N \)-dependent form in Eq. 8. We use typical parameters estimates for organic materials such as poly(p-phenylene vinylene), PPV37 and anthracene38 where the exciton lifetime is of the order of 1 ns39, \( \tau_{TCT} \approx 10 \text{ns} \), \( \tau_{SCT} \approx 1-10 \text{ps} \)38 and \( \tau_{ISC} \approx 0.1-1.0 \text{ns} \).36,38,37

The evolution of \( N_k \) \((k= SX, TX, TCT, SCT)\) with time, \( t \) is shown in Fig. 3. While we set \( \zeta = 0 \) in Fig. 3a, a non-zero \( \zeta = 0.15 \) (which reflects higher exciton purity) is used to obtain the results shown in Fig. 3b. All other parameters are provided in the caption of Fig. 3. In particular, we chose \( \tau_{TCT} = 10 \text{ns} \) and \( \tau = 10 \text{ns} \). Accordingly the time of evolution, \( t \) appears in units of 10 ns and the population of all species, \( N_k \) are taken relative to the initial number electron-hole pairs, \( N_i = 1 \). We note that for the chosen set of parameters, the singlet exciton population rises rapidly and reaches a steady state at \( t \approx 30 \text{ns} \). The triplet exciton which shows appreciable loss even at the beginning stage of evolution, is not apparent in Fig. 3a,b. The population of the triplet CT species appears higher than the singlet CT species, for the set of parameters employed to solve Eqs. 1 to 4. A saturation of the singlet exciton population occurs beyond large times \( t > 35 \text{ns} \) for non-zero purity \( P_s = P_t \).

In contrast to Fig. 3a,b where we used \( \beta = \tau_{SCT}/\tau_{TCT} = 0.001 \), the results in Fig. 4b are obtained using an increased \( \beta = 0.1 \). Otherwise other parameters used in Fig. 4b remain the same as those used in Fig. 3b. We note the prominent appearance of the triplet exciton with increase in \( \beta \) in Fig. 4b. Furthermore, with increase in the exciton purity (non-zero \( \zeta \)), the population of the triplet exciton exceeds that of the singlet CT species. The results obtained here indicates the possibility of converting incoming photons to singlet excitons with minimal energy dissipation with onset of the Pauli exclusion mechanism under suitable conditions. The results obtained in this study clearly demonstrates the critical role of triplet excitons when present at high densities. We make clear that all results obtained in this study are valid for small exciton purity estimates. The model employed in this study therefore stands valid for non-ideal excitons which depart slightly from possessing ideal boson behavior.
FIG. 2: (a) The singlet exciton yield, $\eta_s$ as a function of $P_i = P_t = P_i$ obtained using Eq. 9. The red solid line corresponds to $\gamma = \frac{\tau_{ISC}}{\tau_{TCT}} = 4$, while the blue dashed line corresponds to $\gamma = 0.5$. (b) The singlet exciton yield, $\eta_s$ as a function of $\gamma = \frac{\tau_{ISC}}{\tau_{TCT}}$ (Eq. 9). The red solid line corresponds to $P_i = 0.15$, while the blue dashed line corresponds to $P_i = 0.05$. For both figures, we set $\alpha_s = \alpha_p = 1$ ($\alpha_s = \frac{\tau_{SX}}{\tau_{TCT}}$, $\alpha_p = \frac{\tau_{TX}}{\tau_{TCT}}$) and $\beta = 0$.

FIG. 3: (a) $N_k$ (k=SX (red solid line), TX (red dashed line), SCT (blue solid line), TCT (blue dashed line)) as a function of time, $t$, obtained by numerically solving Eqs. 1 to 4 using Eq. 8, and setting $N_i = 1$, $\tau = 10$ ns, $\zeta = 0$. We also choose $\beta = \frac{\tau_{TCT}}{\tau_{TX}} = 0.001$, $\gamma = \frac{\tau_{ISC}}{\tau_{TCT}} = 0.5$, $\frac{\tau_{SX}}{\tau_{TCT}} = 0.7$, $\frac{\tau_{TP}}{\tau_{TCT}} = 0.5$ with $\tau_{TCT} = 10$ ns. (b) Same values for all parameters as in (a), with exception of $\zeta = 0.15$ (see Eq. 8), and $\alpha_s = \frac{\tau_{SX}}{\tau_{TP}} = 1$, $\alpha_p = \frac{\tau_{TX}}{\tau_{TP}} = 1$

III. CONCLUSION

In conclusion, we have derived an analytical relation of the singlet exciton yield that incorporates the Pauli exclusion mechanism via the purity measure, $P$. Our model considers the formation of singlet excitons via inter-conversion from loosely bound charge-transfer (CT) triplet electron-hole pair. We include a channel by which CT triplet states are formed from tightly bound excitons at high densities. Hence we have taken into account the crucial mechanism by which singlet excitons are increased by preventing the loss of triplet exciton which otherwise would occur through recombination and other dissipation processes. Non-ideal triplet excitons are rerouted via the Pauli exclusion mechanism to form loosely bound CT states which subsequently convert to singlet excitons. As a consequence, we obtain the fluorescence quantum yield that can increase beyond the well known estimate of $> 25\%$. This is the main result of this work which has importance in conjugated polymers systems. The theoretical framework presented in this work provides a possible explanation for the high exciton yields noted in earlier works and will be useful in evaluating optimal electroluminescence efficiencies in organic devices.

The idea of entanglement control by structural variations in novel quantum dot nanostructures may be employed to seek enhancement of the singlet exciton yield in polymer systems. The incorporation of the Pauli exclusion principle during diffusion of photogenerated excitons in drift–diffusion models will also assist in the prediction of realistic
operating conditions in polymer based devices with morphologies of different dimensions\textsuperscript{45–47}. To this end, future experimental investigations may consider the control of the entanglement attributes of excitons through polymer length tailoring and engineering of morphological attributes\textsuperscript{48}. The incorporation of the Pauli exclusion principle in determining the efficiency of devices composed of other material systems such as CdSe and PbSe nanocrystals\textsuperscript{49} where multiple excitons are generated, and layered transition metal dichalcogenides with enhanced excitonic properties\textsuperscript{50–54} may be explored in future works. Future experimental works should seek to investigate alternative means of targeting the triplet excitons to increase the singlet exciton yield.

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