Interplay between RISC and TTA in exciplex-based TADF OLEDs

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ABSTRACT

Organic light emitting diodes (OLEDs) based on thermally activated delayed fluorescence (TADF) show increased efficiencies due to efficient upconversion of non-emissive triplet states to emissive singlets states via reverse intersystem crossing (RISC). To assess the influence of the characteristic efficiency-enhancing RISC process as well as possible efficiency-limiting effects in operational OLEDs, we performed temperature-dependent measurements of transient electroluminescence (trEL). With kinetic modeling, we quantify and separate the impact of different time- and temperature-dependent contributions to the EL in the established model donor:acceptor system m-MTDATA:3TPYMB. The underlying rate equations adapted for EL measurements on TADF systems include radiative and non-radiative first- and second-order effects. In this way, we are able to evaluate the non-radiative recombination and annihilation processes with respect to their efficiency-limiting effects on these OLEDs. On the one hand, we evaluate the depopulation of intermolecular exciplex triplet states via non-radiative direct triplet decay, RISC and triplet-triplet annihilation (TTA). On the other hand, we determine the contribution to EL from the formation of singlet exciplex states via polarons, RISC and TTA. Our results show that triplet depopulation by TTA outcompetes RISC and thus contributes significantly to EL while, however, limiting the overall device quantum efficiency.

INTRODUCTION

Organic light emitting diodes (OLEDs) based on thermally activated delayed fluorescence (TADF) have attracted much attention since their first report [Endo2009], due to the ability to achieve 100% internal quantum efficiency. The underlying mechanism is based on harvesting the non-emissive triplet states and up-convert them to emissive singlet states via reverse intersystem crossing (RISC) [Goushi2012a, Tao2014]. Since this mechanism is driven by thermal energy, a necessary molecular property for this purpose is a small energy gap $\Delta E_{ST}$ between the first excited singlet and triplet states, e.g. by minimizing the overlap of involved HOMO and LUMO orbitals [Endo2011]. Several promising concepts are under consideration to realize efficient TADF. Either by combining donor and acceptor molecules that can share an intermolecular exciton at their interface, also called exciplex [Goushi2012b, Uoyama2012] or alternatively by designing molecules that incorporate donor and acceptor moieties, enabling intramolecular excitons [Zhang2014, Nakanotani2013]. However, a small energy gap $\Delta E_{ST}$ is not the only requirement for efficient TADF devices [Haase2018]. The efficiency-increasing RISC rate must be large enough to outperform efficiency-reducing non-radiative recombination and annihilation processes.

This work focuses on a model system for the investigation of exciplex-based OLEDs employing the donor 4,4’,4”-Tris[(3-methylphenyl)phenylamino]triphenylamine (m-MTDATA) and the acceptor Tris(2,4,6-trimethyl-3-(pyridin-3-yl)phenyl)borane (3TPYMB). This material combination still attracts a lot of attention in literature [Yuan2018, Huang2018, Bunzmann2020], however, according to reports, it never achieved an external quantum efficiency (EQE) higher than 12.9 % [Huang2018]. Since intermolecular donor:acceptor systems are generally characterized by a small energy gap $\Delta E_{ST}$, the limitation in efficiency is surprising. To investigate the influence of the characteristic RISC process as well as possible efficiency-limiting effects on device quantum efficiency, we carried out temperature-dependent transient electroluminescence (trEL) measurements. In order to identify loss processes, we

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analyzed these transient measurements with a suitable kinetic model adapted for EL measurements on TADF devices. The underlying rate equations for singlets, triplets and polarons include second-order terms to consider annihilation processes.

We aim to address all relevant processes involved in an operating OLED device, since previous reports refer either to modeling of transient photoluminescence (trPL) on TADF materials [Haase2018, Kudriashova2018, Naito2020], transient EL of TADF devices without considering annihilation effects [Weissenseel2019] or on transient EL of non-TADF devices considering for second-order processes [Baldo2000, Zhang2010, Kasemann2011, Ruhstaller2001]. However, as shown in the following, it is especially important in TADF devices to account for annihilation effects. On the one hand, annihilation effects are often based on interactions with triplet excitons and are therefore more influential with increasing triplet density in the device. However, the RISC process positively counteracts depopulation via annihilation as it reduces the triplet density. Thus, the magnitude of the RISC rate and the effective triplet density are important factors determining the influence of efficiency-limiting processes. On the other hand, particularly in electrically driven devices, the consideration of second-order effects is important since significantly more triplet excitons are generated through injection than in optical excitation. Due to their long lifetime, triplet excitons support accumulations which result in interactions of annihilation [Murawski2013]. However, a difficulty in transient EL modelling is that the OLED first reaches an equilibrium state, determined by all present rates, before the transient EL decay is recorded at the time of switching off the applied voltage. This results in a population distribution of singlets and triplets in the operational steady-state, differing significantly from the generation ratio of 1:3. Therefore, a suitable kinetic model has to determine the population number in the operational steady-state as starting parameters for the transient EL decays.

In this paper, we show how adequate kinetic modeling of temperature-dependent transient EL measurements helps to quantify efficiency-increasing and -limiting first- and second-order processes. Ultimately, this enables us to quantify the contribution of these different processes to the steady-state EL of operational devices.

THEORETICAL

Figure 1. Kinetic model of a three-level system for transient EL. Injected charge carriers form excitons with the Langevin recombination rate $\gamma$. Excitons populate singlet and triplet states according to 1:3 spin statistics. Possible first-order processes include fluorescent rate $k_F$, (reverse) intersystem crossing rate $k_{(r)ISC}$ and non-radiative decay rate $k_T$. Second-order processes are considered with singlet-triplet annihilation rate $k_{ST}$, triplet-triplet annihilation rate $k_{TT}$ and triplet-polaron annihilation rate $k_{TP}$.
**Kinetic Model.** Figure 1 schematically illustrates the kinetic model including all of the considered processes for the analysis of electrically driven TADF OLEDs. In EL measurements, the excitation is given by electrical injection. The injected charge carriers form excitons assuming the Langevin recombination with rate $\gamma$, populating continuously the singlet states $S_1$ and triplet states $T_1$ in a 1:3 ratio according to spin statistics [Versin2011, Czerwieci2016]. Afterwards, the excitons undergo transitions with rate-dependent probabilities. After a certain time of continuous OLED operation, an equilibrium of singlet, triplet and charge carrier densities, determined by all present rates, is achieved. At this point, the OLED has reached the steady-state of EL operation. The first-order processes in this kinetic model include the radiative rate for fluorescence $k_F$ of the $S_1$ states, the non-radiative decay rate $k_T$ of the $T_1$ states and the direct (ISC) $k_{ISC}$ as well as the reverse ISC rate $k_{RISC}$ for transitions between $S_1$ and $T_1$. Density-dependent second-order processes involve all relevant annihilation effects that are considered in the literature in relation to TADF [Niwa2018, Choi2017, Huang2018, Yuan2018]. These are particularly effects that involve interactions with triplet excitons. Regarding the long lifetime of triplet excitons, the probability of annihilation processes is expected to be higher for triplet excitons than for singlet excitons [Murawski2013]. Furthermore, the triplet density in electrically driven devices is significantly higher than in optical excitation due to their generation ratio. Therefore, we consider the rates for triplet-triplet annihilation $k_{TT}$, triplet-polaron annihilation $k_{TP}$ and singlet-triplet annihilation $k_{ST}$.

Triplet-triplet annihilation (TTA) is based on the interaction of two triplet excitons. Their encounter leads to an intermediate compound or scatter state ($T_1^2$) which, according to spin statistics, can be transformed into a singlet, triplet or quintet state [Dyakonov1997, Stich2013]. Since no triplet exciton is lost for the quintet state, the rate $k_{TT}$ can effectively be transformed into 1/4 for the singlet and 3/4 for the triplet states (see Supporting Information):

$$T_1 + T_1 \rightarrow \begin{cases} 
\frac{5}{9} & T_1 + T_1 \\
\frac{3}{9} & T_1 + S_0 \\
\frac{1}{9} & S_1 + S_0 
\end{cases} \text{Supp. Inf.} \rightarrow \begin{cases} 
\frac{3}{4}k_{TT} & T_1 + S_0 \\
\frac{1}{4}k_{TT} & S_1 + S_0 
\end{cases}$$  \hspace{1cm} (1)

Triplet-polaron annihilation (TPA) is mainly a Förster-type transfer [Reineke2007] and can be described as an annihilation process of triplet excitons with the spin state of the polaron [Murawski2013], resulting in the transfer of the polaron in the ground state $n$ to an excited state $n^*$ [Zhang2010]:

$$T_1 + n \rightarrow k_{TP} S_0 + n^*$$  \hspace{1cm} (2)

Based on the long-living triplet states, singlet-triplet annihilation (STA) can also occur at high triplet densities [Kasemann2011]. STA is a spin-allowed Förster-type energy transfer, whereby the triplet exciton will be raised to a higher triplet state $T_n$ with relaxation of the singlet exciton into the ground state $S_0$ [Zhang2010]:

$$S_1 + T_1 \rightarrow k_{ST} S_0 + T_n$$  \hspace{1cm} (3)

Considering these second-order effects in combination with the linear first-order processes mentioned above, the kinetics of the excited states in an operating OLED can be described with the following rate equations. We assume a polaron density $n$ due to injected current density $j$, which recombines with the Langevin recombination rate $\gamma$ [Baldo2002, Zhang2010, Kasemann2011, Murawski2013]:

$$\frac{dn}{dt} = \frac{j}{ed} - \gamma n^2$$  \hspace{1cm} (4)
While \( j / ed \) describes the exciton generation within the emissive layer thickness \( d \) and elementary charge \( e \) [Baldo2000, Reineke2007], the second term \( \gamma n^2 \) subtracts the formed excitons. The Langevin recombination rate is given by \( \gamma = \frac{e (\mu_e + \mu_h)}{(\varepsilon_0 \varepsilon_r)} \) [Ruhstaller2001] with \( \mu_e \) and \( \mu_h \) representing the mobilities of electrons and holes in the emission layer [Tanaka2007, Yuan2018] and \( \varepsilon_0 \) and \( \varepsilon_r \) the vacuum and emission layer permittivity, respectively (see Supporting Information).

A quarter of the created excitons directly contribute to the population of the singlet state. Together with equation 1 – 4, this results in the following rate equation for the kinetics of the singlet states:

\[
\frac{dS_t}{dt} = -k_F S_1 - k_{ISC} S_1 + k_{RISC} T_1 + \frac{1}{4} k_{TT} T_1^2 - k_{ST} T_1 S_1 + \frac{1}{4} \gamma n^2 \tag{5}
\]

The triplet states are directly populated by the other three quarters of initial excitons, resulting together with the other mentioned processes in the rate equation for the kinetics of the triplet states:

\[
\frac{dT_1}{dt} = -k_T T_1 + k_{ISC} S_1 - k_{RISC} T_1 - \frac{5}{4} k_{TT} T_1^2 - 2k_{TP} T_1 n + \frac{3}{4} \gamma n^2 \tag{6}
\]

The pre-factor for TTA (\( \frac{1}{4} \) or \( \frac{5}{4} \)) is obtained from the different cases of eq. 1 (see Supporting Information). The pre-factor for TPA (2) is based on the assumption of balanced electrons and holes in the emissive layer, which is discussed in more detail below.

**METHODS**

**OLED Fabrication.** The OLEDs were fabricated on indium-tin-oxide (ITO) pre-coated glass substrates (1.62 cm\(^2\)). A 40 nm hole injection layer of poly(3,4-ethylenedioxythiophene):polystyrolsulfonate (PEDOT:PSS, 4083Ai) was spin-coated and subsequently annealed for 10 min at 150°. In a vacuum thermal evaporation chamber with base pressure \( p < 10^{-6} \) mbar, the donor m-MTDATA and the acceptor 3TPYMB as well as the calcium (Ca) and aluminum (Al) cathode were deposited by thermal evaporation. The resulting device structure is ITO/PEDOT:PSS/m-MTDATA(30nm)/m-MTDATA:3TPYMB(70nm,1:1)/3TPYMB(30nm)/Ca(5nm)/Al(120nm).

**Transient EL Measurements.** The transient EL measurements were performed in a nitrogen flow cryostat (Oxford 935). A function generator (Agilent Technologies 81150A) was used for pulsed OLED operation and the luminescence was detected with a photodiode (Hamamatsu Si photodiode S2387-66R). A current-voltage amplifier (Femto DHP-PCA-100) amplified the light-induced photocurrent, which was recorded with a digitizer card (GaGe Razor Express 1622 CompuScope). OLEDs were driven by voltage pulses of 2 ms to ensure steady-state operation. EL transients were recorded after the end of the voltage pulse. The response time of the setup is determined by the bandwidth of the current voltage amplifier of 1 MHz at a gain level of \( 10^6 \) V/A. To receive a defined sample data count, the step-size of the transient EL measurements was interpolated to this time resolution.

**Fit Method.** To fit the measured transient EL data, we used the *curve_fit* tool from the Python toolbox *SciPy* (v. 1.1.0.). The procedure differs from fitting transient PL traces after pulsed laser excitation, since the steady-state of the OLED must first be simulated based on the rate equations 4 – 6 and the injected current density. The knowledge of the population densities in the steady state is important because it determines the initial state of the transient decay. Since this population ratio is determined by an equilibrium of all present rates, it differs from the generation ratio of 1:3. For the transient decay, the current density \( j \) in equation 4 is set to zero, which is equivalent to switching off the OLED. In order to determine the rates from the transient EL decays, the iterative numerical solution of the rate equations is fitted to the measured EL transients starting from 10 \( \mu \)s after switching off the OLED (to avoid
influence of electrical artifacts). More details about the fitting process are specified in the Supporting Information.

RESULTS AND DISCUSSION

![Figure 2](image)

**Figure 2.** Temperature-dependent transient EL decays for an OLED with emission layer of m-MTDATA:3TPYMB. The traces show a temperature-activated non-linear behavior.

**Experimental Results.** Figure 2 shows transient EL measurements of the reported system in semi-logarithmic representation for temperatures from 200 K to 300 K in 20 K steps. On the one hand, we observe a temperature-activated process since the transient EL decays become faster with increasing temperature. On the other hand, the shape of the transients deviates from the expected biexponential decay by prompt and delayed fluorescence. A deviation from linear traces in semi-logarithmic presentation indicates the influence of second order processes, e.g. annihilation processes. Murawski et al. and Baldo et al. reported deviations from mono-exponential decay in transient EL measurements of phosphorescent OLEDs and both attributed this effect to TTA [Baldo2000, Murawski2013]. However, in order to also consider also the other annihilation effects mentioned, we used the rate equations 4 – 6 including STA, TPA and TTA to find the dominant second-order process responsible for the shape of the traces in Figure 2.

In order to avoid distorting the fit by too many free parameters, the radiative rate $k_F$ and the intersystem crossing rate $k_{ISC}$ are set to values from literature [Goushi2012a, Deotare2015]. Therefore, the fit determined the RISC rate $k_{RISC}$, the non-radiative decay rate $k_T$ and the annihilation rate. Before fitting the transient EL decays from Figure 2, we first identify the dominant annihilation effect that determines the shape of the transients. Therefore, we use reduced rate equations of equation 4 – 6 with one annihilation effect, respectively and investigate its influence on the numerical solutions.
To visualize the influence of the different annihilation effects on the transient EL decay, Figure 3 shows the numerical solution of the reduced rate equations 4 - 6 with the variation of one annihilation rate, respectively. Figure 3a presents simulated transient EL traces with different STA rates. In Figure 3b, the TPA rate was varied and Figure 3c shows the influence of different TTA rates. For comparison with experimental data, the gray curve always displays the experimental EL transient recorded at 200 K. The fitting method described in the method section provided only reasonable fits for the case of TTA as annihilation effect. Because STA and TPA alone cannot reproduce the experimental data, suitable values for the annihilation rates had to be set manually for illustrative purpose.

Since the influence of STA depends on the product of singlet and triplet population densities, STA does not have a relevant influence due to the smaller density and shorter lifetime of singlet excitons. At lower values of STA, the traces are still approximately linear in the logarithmic presentation. At higher orders of magnitude, the influence can be observed mainly at the beginning of the transient. Overall, this annihilation effect cannot reproduce the shape of the measured trace. Furthermore, the measured transient EL decays are current/voltage dependent (Figure S1), which contradicts the dominance of STA [Murawski2013]. The influence of the TPA rate on EL transients (Figure 3b) is particularly strong shortly after switching off the OLED, i.e. in the first microseconds. The reason for this is the finite transit time of polarons in the device active layer after the voltage turn-off, determining the time scale in which TPA can occur. After this time, the traces follow a mono-exponential decay. In OLEDs, the presence of majority charge carriers is often discussed, as they can promote TPA while minority charge carriers are used for exciton formation [Reineke2007]. However, the influence of these majority carriers would also be negligible after the transit time of a few microseconds and can therefore not be responsible for the shape of the transient at later times. In contrast, TTA is present during a longer time span of the decay. Certainly, the influence of TTA is higher at the beginning of the transient, since most triplet excitons are present there. Nevertheless, TTA influences the EL decay as long as triplet excitons are existent, therefore the influence of TTA is present over the whole decay. Figure 3c confirms that considering TTA in the rate equations 4 – 6 matches the measured transient EL trace almost perfectly.

By comparing the behaviors of the modeled traces, we can observe that TTA is the mechanism, which is responsible for the characteristic shape of the transient EL trace. On the one hand, this result is consistent with the already mentioned statements of Murawski et al. and Baldo et al., who explained deviations in EL transients from linear progressions with TTA [Baldo2000, Murawski2013]. On the other hand, we carried out excitation power-dependent PL measurements (Figure S2). These measurements give the order of the process from the slope when plotted in a double logarithmic representation. Even though under optical excitation (where the triplet density is slightly lower), TTA can be verified by a slope of almost two, which implies the necessity of two triplet excitons for a radiative decay.
**Fit Results.** The results in the following section were obtained with the introduced fit procedure, based on reduced rate equations 4 – 6 with considering the annihilation effect TTA. Therefore, the fit procedure determined the RISC rate $k_{RISC}$, the non-radiative decay rate $k_T$ and the triplet-triplet annihilation rate $k_{TT}$. Typically, transient PL measurements [Haase2018, Kudriashova2018] or sometimes also PL quantum yield measurements [Dias2016] provide fundamental rates like $k_F$, $k_{(R)ISC}$ and $k_T$. In most exciplex systems, however, the CT state does not absorb directly (Figures S3). Thus, the optical excitation first generates an intramolecular exciton on the donor or acceptor molecule. The subsequent charge transfer creates an intermolecular exciton at the interface (CT/exciplex state). In contrast to that, electrical excitation (EL measurements) populates CT states directly, which simplifies the system to a three-level system. Therefore, transient EL measurements are particularly appropriate (compared to PL) for the determination of the RISC rate in these exciplex systems.

**Figure 4.** Results of the fit procedure for 200 K. a) Fit (red) of the iterative numerical solution of equations 4 – 6 with considering TTA as the dominant quenching mechanism. b) Time-dependent population density of polarons $n$ (purple), singlet states $S_1$ (orange) and triplet states $T_1$ (gray).

**Figure 4a** shows the resulting fit (red) of the transient EL measurement at 200 K. The RISC rate was determined to be $k_{RISC} = 3.3 \cdot 10^4 \text{ s}^{-1}$ which corresponds to a RISC time of about 30 $\mu$s. The order of magnitude of this rate is consistent with reports from this or similar donor:acceptor TADF systems [Goushi2012a, Hontz2015]. At the same time, the RISC rates in these intermolecular exciplex systems are smaller than for state-of-the-art intramolecular TADF emitters. Hence, the depopulation of triplet states by RISC proceeds slower and the long-living triplet excitons accumulate, enhancing the influence of TTA [Murawski2013]. From the fit we determined a TTA rate of $k_{TT} = 1.6 \cdot 10^{13} \text{ cm}^3 \text{s}^{-1}$. Baldo et. al., Kasemann et. al. and Murawski et. al. reported TTA rates from transient EL measurements on phosphorescent OLEDs of the same orders of magnitude [Baldo2000, Kasemann2011, Murawski2013]. Niwa et. al. determined a TTA rate of about $10^{18} \text{ cm}^3 \text{s}^{-1}$ at helium temperatures by steady-state PL measurements of intramolecular TADF emitters [Niwa2018]. Since the TTA rate is highly temperature dependent (Figure S4), the fitted TTA rates agree very well with previously discussed rates. The fit curves for all temperatures (Figure S5) as well as the rates determined by the fit are available in the Supporting Information (Table S1).

To quantify the density and lifetime of the excited states, **Figure 4b** shows the time-dependent population density of polarons ($n$), first excited singlet states ($S_1$) and first excited triplet states ($T_1$) slightly before and after switching off the OLED at time $t = 0$ $\mu$s for 200 K. The simulations for the population density are based on the fit results in Figure 4a and the rate equations 4 – 6. On the one hand, we observe that the polarons disappear within a few microseconds. This observation agrees well with the results shown in Figure 3b, suggesting that TPA only affects the beginning of the EL transients as
there are negligibly few free polarons available in the device after several microseconds. On the other hand, Figure 4b shows that the steady-state triplet density \((t < 0 \, \mu s)\) is significantly higher than the singlet density (20:1). The actual triplet-to-singlet ratio when the OLED is switched off therefore does not match the electrical occupation of the states with the ratio of 3:1 according to simple spin statistics. In fluorescent OLEDs, when only considering the non-radiative decay rate, Shinar et. al. showed that the triplet density can be up to \(10^5\) times higher than the singlet density [Shinar2012]. However, they assumed a relatively high ratio of triplet to singlet lifetime of \(10^3:1\) and included no depopulation of the triplet states by annihilation processes or RISC mechanism in their estimation. Since the ratio of triplet to singlet lifetime in our system is smaller and the RISC and TTA processes both play a significant role in the depopulation of triplet states, the triplet to singlet population ratio is reduced.

**Figure 5.** Schematic bucket model (top) and temperature-dependent impact (bottom) of different (de-)population processes. a) Contribution to depopulation of triplet state \(T^*_t\) via non-radiative decay (blue), RISC (green) and TTA with creation of a singlet state (red, dotted) or a triplet state (red). b) Contributions to EL of three different processes: singlet excitons formed via polarons (gray), RISC (green) and TTA (red, dotted).

**Figure 5a** shows a scheme of the processes that contribute to the population (polarons, ISC) and depopulation (non-radiative decay, TTA, RISC) of the triplet states. To assess the influence of the processes depopulating the triplet states, we investigate the impact of respective depopulation channels (gray framed rates in Figure 5a). Therefore, the diagram in Figure 5a represents the temperature-dependent contributions of the individual channels to the depopulation of the triplet density (added up to 100%). The data sets are derived from the fit results of the measured EL transients (Table S1), which we used to calculate the steady-state with equations 4 – 6. The first-order non-radiative decay process of the triplet state (blue) slightly increases with temperature but overall plays a minor role in the depopulation of the triplet state. Therefore, TTA and RISC are primarily responsible for the depopulation of the triplet states. Our fit procedure delivers a singlet-triplet energy gap \(\Delta E_{ST} = 52 \, \text{meV}\), which is consistent with previous reports for this material system [Goushi2012a, Deotare2015, Huang2018, Bunzmann2020]. The RISC rate is assumed to increase exponentially with temperature according to a Boltzmann factor. However, the impact of RISC on depopulation of triplet states \(T^*_t\) decreases with increasing temperature. The reason for the decline is the proportion of TTA, which is also temperature-activated. An excess of injected charge carries cannot cause this effect, since the current remains almost constant for higher temperatures (Table S1). Actually, in contrast to the RISC rate, which is increasing following a Boltzmann factor \(\exp(-c_t/T)\), the TTA rate decreases
exponentially $\exp(c_2 \cdot T)$ (Figure S4). Consequently, the proportion of TTA in triplet density depopulation increases with temperature and outperforms depopulation via RISC. This observation is remarkable since at relatively high temperatures, the triplet lifetime is significantly reduced by the fast phonon-assisted decay, which normally overcomes TTA [Liu2009, Xiang2016]. The fact that the proportion of TTA nevertheless increases indicates the enormous influence of TTA in this system. TTA has already been detected in other TADF systems [Niwa2018, Scholz2020]. However, the proportion of TTA in exciplex-based OLEDs is significantly higher. On the one hand, the reason is the investigation of operational OLEDs, since a higher triplet density is present in EL in contrast to PL. On the other hand, and this is the larger influence, the moderate RISC rate in exciplex-based systems allows an accumulation of triplet excitons. As a result, at room temperature, more than 80% of the triplet depopulation is caused by TTA and only about 15% by the - for TADF specific - RISC mechanism. Since one TTA event results in the loss of at least one triplet exciton, this annihilation effect provides a strong efficiency-limiting decay channel in these OLEDs.

According to equations 1 and 5, TTA also produces a small amount of singlet excitons (red, dotted area in Figure 5a) and therefore also contributes to the EL. Figure 5b shows a scheme of the processes that populate (polarons, TTA, RISC) and depopulate (radiative decay, ISC) the singlet states. To investigate the influence of the population of the singlet state by polarons, TTA and RISC on the EL, we consider the total number of excitons present in the singlet states (gray box) initially populated by the mentioned processes (depopulation processes from the singlet states are included iteratively). In order to quantify the influence of these processes on the EL, the diagram in Figure 5b illustrates the contributions of singlet excitons generated via the different channels to the actual light emission (added up to 100%). Since more triplet excitons are lost by TTA with increasing temperature (Figure 5a), the proportion of polarons in the EL increases with temperature. As already shown in Figure 5a, the percentage of triplet excitons that are depopulated by RISC decreases with temperature. Accordingly, the number of singlet excitons created by RISC follows the same trend. The interesting fact is that TTA events, whereof only 25% generate a singlet exciton (equation 1), constitute a considerable share of the EL (red, dotted part). Since this path of producing singlet excitons requires twice as many triplet excitons as RISC, it is a less efficient way of triplet harvesting. At room temperature, however, the EL is composed almost equally of singlet excitons initially populated by polarons, RISC and TTA, questioning the role of TADF in these OLEDs.

CONCLUSION

We have established a kinetic model for electrically driven TADF OLEDs including radiative and non-radiative first- and second-order effects that reproduces transient EL measurements accurately. We developed a suitable fit procedure to analyze the kinetic processes in operational OLEDs and revealed triplet-triplet annihilation (TTA) as the dominant second-order effect. Remarkably, TTA outperforms the RISC process in terms of triplet depopulation as a result of a moderate RISC rate and a high triplet density, especially for electrical excitation. Since one TTA event results in the loss of at least one triplet exciton, this is a significant quantum efficiency-limiting loss channel for this type of OLEDs. We determined that the influence of TTA increases with temperature, which leads to TTA limiting the overall performance of OLEDs especially at room temperature. Consequently, the EL does not only consist of prompt and delayed constituents due to RISC, it also contains a considerable contribution of emissive excited states formed via TTA. Since TTA is a less efficient triplet harvesting mechanism in contrast to RISC, this result represents an important insight for research on exciplex-based TADF OLEDs. We propose to use transient EL analysis with this adapted kinetic model as a standard tool to study the properties, possible efficiency-limiting processes and contributions to EL of electrically driven TADF OLEDs.
ASSOCIATED CONTENT

Supporting Information
Supporting Information includes more details about the fit procedure, all results including fitting errors, voltage-dependent transient EL measurements, excitation power-dependent PL measurements, photoexcitation/-emission spectra, temperature dependence of TTA rate $k_{TT}$, temperature-dependent trEL measurements including fits and transformation of TTA rate in equation 1.

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Notes
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| References                                                                 |
|---------------------------------------------------------------------------|
| Baldo2000: Baldo, M. A., Adachi, C., & Forrest, S. R. (2000). Transient analysis of organic electrophosphorescence. II. Transient analysis of triplet-triplet annihilation. *Physical Review B*, 62(16), 10967. DOI: 10.1103/PhysRevB.62.10967. |
| Baldo2002: Baldo, M. A., Holmes, R. J., & Forrest, S. R. (2002). Prospects for electrically pumped organic lasers. *Physical Review B*, 66(3), 035321. DOI: 10.1103/PhysRevB.66.035321. |
| Bunzmann2020: Bunzmann, N., Weisenseel, S., Kudriashova, L., Gruene, J., Krugmann, B., Grazulevicius, J. V., Sperlich, A., & Dyakonov, V. (2020). Optically and electrically excited intermediate electronic states in donor: acceptor based OLEDs. *Materials Horizons*, 7, 1126 – 1137. DOI: 10.1039/c9mh01475f |
| Choi2017: Choi, J. M., Lee, W., Kim, K. K., & Lee, J. Y. (2017). Exciton management by co-doping of blue triplet emitter as a lifetime improving method of blue thermally activated delayed fluorescent devices. *Organic Electronics*, 45, 104-107. DOI: 10.1016/j.orgel.2017.03.003 |
| Czerwieniec2016: Czerwieniec, R., Leitl, M. J., Homeier, H. H., & Yersin, H. (2016). Cu(I) complexes—Thermally activated delayed fluorescence. Photophysical approach and material design. *Coordination Chemistry Reviews*, 325, 2-28. DOI: 10.1016/j.ccr.2016.06.016 |
| Deotare2015: Deotare, P. B., Chang, W., Hontz, E., Congreve, D. N., Shi, L., Reusswig, P. D., Modtland, B., Bahlke, M. E., Lee, C. K., Wilard, A., P., Bulović, V., Van Voorhis, T. & Baldo, M.A. (2015). Nanoscale transport of charge-transfer states in organic donor–acceptor blends. *Nature materials*, 14(11), 1130-1134. DOI: 10.1038/nmat4424 |
| Dias2016: Dias, F. B., Santos, J., Graves, D. R., Data, P., Nobuyasu, R. S., Fox, M. A., Batsanov, A. S., Palmeira, T., Berberan-Santos, M., Bryce, M. R. & Monkman, A. P. (2016). The role of local triplet excited states and D-A relative orientation in thermally activated delayed fluorescence: photophysics and devices. *Advanced Science*, 3(12), 1600080. DOI: 10.1002/advs.201600080 |
| Dyakonov1997: Dyakonov, V., Röschl, G., Schwoerer, M., & Frankevich, E. L. (1997). Evidence for triplet interchain polaron pairs and their transformations in polyphenylenevinylene. *Physical Review B*, 56(7), 3852. DOI: 10.1103/PhysRevB.56.3852 |
| Endo2009: Endo, A., Ogasawara, M., Takahashi, A., Yokoyama, D., Kato, Y., & Adachi, C. (2009). Thermally activated delayed fluorescence from Sn4+-porphyrin complexes and their application to organic light emitting diodes—A novel mechanism for electroluminescence. *Advanced Materials*, 21(47), 4802-4806. DOI: 10.1002/adma.200900983 |
| Endo2011: Endo, A., Sato, K., Yoshimura, K., Kai, T., Kwada, A., Miyazaki, H., & Adachi, C. (2011). Efficient up-conversion of triplet excitons into a singlet state and its application for organic light emitting diodes. *Applied Physics Letters*, 98(8), 42. DOI: 10.1063/1.3558906 |
| Goushi2012a: Goushi, K., Yoshida, K., Sato, K., & Adachi, C. (2012). Organic light-emitting diodes employing efficient reverse intersystem crossing for triplet-to-singlet state conversion. *Nature Photonics*, 6(4), 253-258. DOI: 10.1038/nphoton.2012.31 |
| Goushi2012b: Goushi, K., & Adachi, C. (2012). Efficient organic light-emitting diodes through up-conversion from triplet to singlet excited states of exciplexes. *Applied Physics Letters*, 101(2), 023306. DOI: 10.1063/1.4737006 |
| Haase2018: Haase, N., Danos, A., Pflumm, C., Morherr, A., Stachelek, P., Mekic, A., Brütting, W. & Monkman, A. P. (2018). Kinetic modeling of transient photoluminescence from thermally activated delayed fluorescence. *The Journal of Physical Chemistry C*, 122(51), 29173-29179. DOI: 10.1021/acs.jpcc.8b011020 |
| Hontz2015: Hontz, E., Chang, W., Congreve, D. N., Bulović, V., Baldo, M. A., & Van Voorhis, T. (2015). The role of electron–hole separation in thermally activated delayed fluorescence in donor–acceptor blends. *The Journal of Physical Chemistry C*, 119(45), 25591-25597. DOI: 10.1021/acs.jpcc.5b07340 |
Huang2018: Huang, Q., Zhao, S., Wang, P., Qin, Z., Xu, Z., Song, D., Qiao, B. & Xu, X. (2018). Investigating the evolution of exciplex states in thermally activated delayed fluorescence organic light-emitting diodes by transient measurement. *Journal of Luminescence, 201*, 38-43. DOI: 10.1016/j.jlumin.2018.03.094

Liu2009: Liu, R., Zhang, Y., Lei, Y. L., Chen, P., & Xiong, Z. H. (2009). Magnetic field dependent triplet-triplet annihilation in Alq 3-based organic light emitting diodes at different temperatures. *Journal of Applied Physics, 105*(9), 093719. DOI: 10.1063/1.3125507

Kaji2015: Kaji, H., Suzuki, H., Fukushima, T., Shizu, K., Suzuki, K., Kubo, S., Komino, T., Oiwa, H., Suzuki, F., Wakamiya, A., Murata, Y., & Adachi, C. (2015). Purely organic electroluminescent material realizing 100% conversion from electricity to light. *Nature Communications, 6*, 8476. DOI: 10.1038/ncomms9476

Kasemann2011: Kasemann, D., Brückner, R., Fröb, H., & Leo, K. (2011). Organic light-emitting diodes under high currents explored by transient electroluminescence on the nanosecond scale. *Physical Review B, 84*(11), 115208. DOI: 10.1103/PhysRevB.84.115208

Kudriashova2018: Drigo, N. A., Kudriashova, L. G., Weissenseel, S., Sperlich, A., Huckaba, A. J., Nazeeuruddin, M. K., & Dyakonov, V. (2018). Photophysics of Deep Blue Acridane-and Benzonitrile-Based Emitter Employing Thermally Activated Delayed Fluorescence. *The Journal of Physical Chemistry C, 122*(39), 22796-22801. DOI: 10.1021/acs.jpcc.8b08716

Murawski2013: Murawski, A. J., Nazeeruddin, M. K., & Dyakonov, V. (2018). Time-resolved fluorescence organic light-emitting diodes based on thermally activated delayed fluorescence. *Advances in Materials, 25*(47), 6801-6827. DOI: 10.1002/adma.201301603

Nakanotani2013: Nakanotani, H., Masui, K., Nishide, J., Shibata, T., & Adachi, C. (2013). Promising operational stability of high-efficiency organic light-emitting diodes based on thermally activated delayed fluorescence. *Scientific Reports, 3*, 2127. DOI: 10.1038/srep02127

Naito2020: Kobayashi, T., Kawate, D., Niwa, A., Nagase, T., Goushi, K., Adachi, C., & Naito, H. (2020). Intersystem Crossing Rate in Thermally Activated Delayed Fluorescence Emitters. *physica status solidi (a), 217*(3), 1900616. DOI: 10.1002/pssa.201900616

Naito2018: Niwa, A., Haseyama, S., Kobayashi, T., Nagase, T., Goushi, K., Adachi, C., & Naito, H. (2018). Triplet-triplet annihilation in a thermally activated delayed fluorescence emitter lightly doped in a host. *Applied Physics Letters, 113*(8), 083301. DOI:

Reineke2007: Reineke, S., Walzer, K., & Leo, K. (2007). Triplet-exciton quenching in organic phosphorescent light-emitting diodes with Ir-based emitters. *Physical Review B, 75*(12), 125328. DOI: 10.1103/PhysRevB.75.125328. DOI: 10.1063/1.5025870

Ruhstaller2001: Ruhstaller, B., Carter, S. A., Barth, S., Riel, H., Riess, W., & Scott, J. C. (2001). Transient and steady-state behavior of space charges in multilayer organic light-emitting diodes. *Journal of Applied Physics, 89*(8), 4575-4586. DOI: 10.1063/1.1352027

Scholz2020: Scholz, R., Kleine, P., Lygaitis, R., Popp, L., Lenk, S., Etherington, M. K., Monkman, A. P. & Reineke, S. (2020). Investigation of Thermally Activated Delayed Fluorescence from a Donor–Acceptor Compound with Time-Resolved Fluorescence and Density Functional Theory Applying an Optimally Tuned Range-Separated Hybrid Functional. *The Journal of Physical Chemistry A, 124*(8), 1535-1553. DOI: 10.1021/acs.jpca.9b11083

Shinar2012: Shinar, J. (2012). Optically detected magnetic resonance studies of luminescence-quenching processes in π-conjugated materials and organic light-emitting devices. *Laser & Photonics Reviews, 6*(6), 767-786. DOI: 10.1002/lpor.201100026

Stich2013: Stich, D., Späth, F., Kraus, H., Sperlich, A., Dyakonov, V., & Hertel, T. (2014). Triplet–triplet exciton dynamics in single-walled carbon nanotubes. *Nature Photonics, 8*(2), 139. DOI: 10.1038/nphoton.2013.316

Tanaka2007: Tanaka, D., Takeda, T., Chiba, T., Watanabe, S., & Kido, J. (2007). Novel electron-transport material containing boron atom with a high triplet excited energy level. *Chemistry Letters, 36*(2), 262-263. DOI: 10.1246/cl.2007.262
• Tao2014: Tao, Y., Yuan, K., Chen, T., Xu, P., Li, H., Chen, R., Zheng, C., Zhang, L. & Huang, W. (2014). Thermally activated delayed fluorescence materials towards the breakthrough of organoelectronics. *Advanced Materials*, 26(47), 7931-7958. DOI: 10.1002/adma.201402532
• Uoyama2012: Uoyama, H., Goushi, K., Shizu, K., Nomura, H., & Adachi, C. (2012). Highly efficient organic light-emitting diodes from delayed fluorescence. *Nature*, 492(7428), 234-238. DOI: 10.1038/nature11687
• Weissenseel2019: Weissenseel, S., Drigo, N. A., Kudriashova, L. G., Schmid, M., Morgenstem, T., Lin, K. H., Prlj, A., Corminboeuf, A. A., Sperlich, A. Brütting, W., Nazeeruddin, M. K. & Dyakonov, V. (2019). Getting the right twist: influence of donor–acceptor dihedral angle on exciton kinetics and singlet–triplet gap in deep blue thermally activated delayed fluorescence emitter. *The Journal of Physical Chemistry C*, 123(45), 27778-27784. DOI: 10.1021/acs.jpcc.8b08716
• Xiang2016: Xiang, J., Chen, Y., Jia, W., Chen, L., Lei, Y., Zhang, Q., & Xiong, Z. (2016). Realization of triplet–triplet annihilation in planar heterojunction exciplex-based organic light-emitting diodes. *Organic Electronics*, 28, 94-99. DOI: 10.1016/j.orgel.2015.10.017
• Yersin2011: Yersin, H., Rausch, A. F., Czerwieniec, R., Hofbeck, T., & Fischer, T. (2011). The triplet state of organo-transition metal compounds. Triplet harvesting and singlet harvesting for efficient OLEDs. *Coordination Chemistry Reviews*, 255(21-22), 2622-2652.
• Yuan2018: Yuan, P., Qiao, X., Yan, D., & Ma, D. (2018). Magnetic field effects on the quenching of triplet excitons in exciplex-based organic light emitting diodes. *Journal of Materials Chemistry C*, 6(21), 5721-5726. DOI: 10.1039/C8TC01260A
• Zhang2010: Zhang, Y., Whited, M., Thompson, M. E., & Forrest, S. R. (2010). Single–triplet quenching in high intensity fluorescent organic light emitting diodes. *Chemical Physics Letters*, 495(4-6), 161-165. DOI: 10.1016/j.cplett.2010.06.079
• Zhang2014: Zhang, Q., Li, B., Huang, S., Nomura, H., Tanaka, H., & Adachi, C. (2014). Efficient blue organic light-emitting diodes employing thermally activated delayed fluorescence. *Nature Photonics*, 8(4), 326. DOI: 10.1038/nphoton.2014.12

**Table of Contents Image**

Transient electroluminescence of exciplex-based TADF OLEDs is modelled to quantify the contributions of reverse intersystem crossing and triplet-triplet annihilation to steady state OLED operation.
SUPPORTING INFORMATION

for

Interplay between RISC and TTA in exciplex-based TADF OLEDs

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Fit Results and Fit Procedure

Table 1. Temperature-dependent fit results (± fit error) for m-MTDATA:3TPYMB OLEDs. The current density was determined by separate measurements and the radiative rate $k_F$ as well as the ISC rate $k_{ISC}$ were taken from literature (see below).

| $T$ [K] | $k_{TT}$ [cm$^2$s$^{-1}$] | $k_T$ [s$^{-1}$] | $k_{RISC}$ [s$^{-1}$] | $k_F$ [s$^{-1}$] | $k_{ISC}$ [s$^{-1}$] | $j$ [A cm$^{-2}$] |
|---------|-------------------|-----------------|-------------------|-----------------|-------------------|-----------------|
| 200     | 1.64e-13 ± 2.6e-15| 1.42e3 ± 7.6e1  | 3.26e4 ± 7.0e2    | 1e5             | 1e6               | 0.0120          |
| 220     | 3.96e-13 ± 1.8e-15| 1.05e3 [1]      | 4.30e4 [2]        | 1e5             | 1e6               | 0.0133          |
| 240     | 1.09e-12 ± 2.9e-15| 2.88e3 ± 6.0e1  | 5.40e4            | 1e5             | 1e6               | 0.0113          |
| 260     | 4.00e-12 ± 1.2e-14| 4.33e3 ± 9.78   | 6.56e4            | 1e5             | 1e6               | 0.0083          |
| 280     | 1.00e-11 ± 7.7e-14| 8.99e3 ± 3.4e2  | 7.74e4            | 1e5             | 1e6               | 0.0080          |
| 300     | 2.68e-11 ± 1.9e-13| 2.17e4 ± 4.4e2  | 8.94e4            | 1e5             | 1e6               | 0.0070          |

[1] Fit with boundary condition ($\Delta E_{ST} \geq 0$) → no realistic error computable
[2] $\Delta E_{ST} = 52$ meV follows from fitting $k_{RISC}$ at 220 K (see below)

The RISC rate $k_{RISC}$ was determined for 200 K because this temperature provides the longest transient and therefore the smallest fit inaccuracy. By fitting the transient EL measurement at 220 K, the singlet-triplet gap $\Delta E_{ST}$ was determined by the RISC rate at 200 K and the assumption of a Boltzmann activated process with the singlet-triplet gap $\Delta E_{ST}$. Since the rates to be fitted are not independent from each other, the knowledge of an exponentially increasing RISC rate with the Boltzmann term was used. For the transients from 240 K – 300 K, the RISC rates were calculated with the RISC rate for 200 K including the fitted energy gap $\Delta E_{ST}$ of 52 meV:

$$k_{RISC,2x0K} = k_{RISC,200K} \cdot \exp \left( \frac{1}{T_{200K}} - \frac{1}{T_{2x0K}} \right) \cdot \frac{\Delta E_{ST}}{k_B}$$

This implies that for the transients for 240 K - 300 K, there were only two free fitting parameters $k_T$ and $k_{TT}$. The errors indicated in Table 1 correspond to the fit errors. All fits (except the energy gap, noted in Table 1) were performed without boundary conditions to determine the global minimum. The

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fluorescence rate $k_F$ and the ISC rate $k_{ISC}$ were assumed to be constant with the temperature [Goushi2012a] and the orders of magnitude were taken from literature [Goushi2012a, Deotare2015]. The Langevin recombination rate was determined with $\epsilon = 2.9$ [Zhang2010, Kasemann2011], $\mu_e = 10^{-5}$ cm$^2$/Vs and $\mu_h = 10^{-4}$ cm$^2$/Vs [Tanaka2007, Yuan2018]. The fits for all temperatures are displayed in Figure S3.

**Figures**

![Figure S1](image1.png)

**Figure S1.** Voltage-dependent transient EL measurements at 300 K. The transient EL decay becomes faster with increasing applied voltage.

![Figure S2](image2.png)

**Figure S2.** Excitation power-dependent PL intensity of a solid film of m-MTDATA:3TPYMB blend in log-log representation. The PL intensity increases with a slope of 1.8 ± 0.1 (fit in blue), indicating a second-order process.

Experimental Details of this experiment: PL spectra of a m-MTDATA:3TPYMB blend (20 mg/ml in chlorobenzene spin-coated on a glass substrate) were recorded with FLS980 spectrometer from Edinburgh Instruments. The excitation source was a Xenon Lamp with different transmission filters (OD 0.1, OD 0.2 and OD 0.4) and a selected emission wavelength of 365 nm. Possible transmissions of the xenon lamp or fluorescence of the glass substrate were subtracted by separately measured spectra of the pure glass substrate.
**Figure S3**: Photoexcitation (black) and photoluminescence (green) spectra of solid films of m-MTDATA:3TPYMB blend. The CT/exciplex state shows no absorption.

Experimental details of this experiment: Photoexcitation (PE) and PL spectra of a solid film of m-MTDATA:3TPYMB blend (spin-coated from 20 mg/ml chlorobenzene solution) were recorded with a FLS980 spectrometer from Edinburgh Instruments. Photoexcitation was measured at an emission wavelength of 540 nm. Photoluminescence was measured with an excitation wavelength of 365 nm.

**Figure S4**: Temperature dependence of the determined TTA rate $k_{TT}$. The rate is exponentially temperature-activated following $k_{TT} \sim \exp (\text{const} \cdot T)$ with $\text{const} = (52.1 \pm 1.3) \times 10^{-3} \text{K}^{-1}$ (fit in blue).
**Figure S5.** Fit results for transient EL measurements from 200 K to 300 K. The fit is always starting at 10 µs to avoid the influence of electrical artifacts.

### Transformation of TTA rate (equation 1):

The TTA process is split up into three parts, either forming a quintet, triplet or the singlet state (equation 1). Hence, the individual loss-rates with respect to the original triplet population are given by the probability of the transition times the number of lost triplets in the process. This results in rates of $\frac{5}{9} k'_{TT}$ for the depopulation of the triplet state and $\frac{1}{9} k'_{TT}$ for the population of the singlet state through TTA. Defining $k_{TT} = \frac{9}{4} k'_{TT}$, these rates can be transformed to the following:

$$\frac{dT_1}{dt} = \ldots - \left(0 \cdot \frac{5}{9} + 1 \cdot \frac{3}{9} + 2 \cdot \frac{1}{9}\right) k'_{TT} = \ldots - \frac{5}{9} k'_{TT} = \ldots - \frac{5}{4} k_{TT}$$

$$\frac{dS_1}{dt} = \ldots + \left(1 \cdot \frac{1}{9}\right) k'_{TT} = \ldots + \frac{1}{9} k'_{TT} = \ldots + \frac{1}{4} k_{TT}$$

*) $k_{TT} = \frac{9}{4} k'_{TT}$

In context of TTA, the formation of a quintet state is sometimes not considered at all. In this way the given rates are formed directly:

$$\frac{dT_1}{dt} = \ldots - \left(1 \cdot \frac{3}{4} + 2 \cdot \frac{1}{4}\right) k_{TT} = \ldots - \frac{5}{4} k_{TT}$$

$$\frac{dS_1}{dt} = \ldots + \left(1 \cdot \frac{1}{4}\right) k_{TT} = \ldots + \frac{1}{4} k_{TT}$$
REFERENCES

- Deotare2015: Deotare, P. B., Chang, W., Hontz, E., Congreve, D. N., Shi, L., Reusswig, P. D., Modtland, B., Bahlke, M. E., Lee, C. K., Wilard, A. P., Bulović, V., Van Voorhis, T. & Baldo, M.A. (2015). Nanoscale transport of charge-transfer states in organic donor–acceptor blends. Nature materials, 14(11), 1130-1134. DOI: 10.1038/nmat4424
- Goushi2012: Goushi, K., Yoshida, K., Sato, K., & Adachi, C. (2012). Organic light-emitting diodes employing efficient reverse intersystem crossing for triplet-to-singlet state conversion. Nature Photonics, 6(4), 253-258. DOI: 10.1038/nphoton.2012.31
- Kasemann2011: Kasemann, D., Brückner, R., Fröb, H., & Leo, K. (2011). Organic light-emitting diodes under high currents explored by transient electroluminescence on the nanosecond scale. Physical Review B, 84(11), 115208. DOI: 10.1103/PhysRevB.84.115208
- Tanaka2007: Tanaka, D., Takeda, T., Chiba, T., Watanabe, S., & Kido, J. (2007). Novel electron-transport material containing boron atom with a high triplet excited energy level. Chemistry Letters, 36(2), 262-263. DOI: 10.1246/cl.2007.262
- Yuan2018: Yuan, P., Qiao, X., Yan, D., & Ma, D. (2018). Magnetic field effects on the quenching of triplet excitons in exciplex-based organic light emitting diodes. Journal of Materials Chemistry C, 6(21), 5721-5726. DOI: 10.1039/C8TC01260A
- Zhang2010: Zhang, Y., Whited, M., Thompson, M. E., & Forrest, S. R. (2010). Singlet–triplet quenching in high intensity fluorescent organic light emitting diodes. Chemical Physics Letters, 495(4-6), 161-165. DOI: 10.1016/j.cplett.2010.06.079