Triazatruxene: A Rigid Central Donor Unit for a D–A$_3$
Thermally Activated Delayed Fluorescence Material Exhibiting
Sub-Microsecond Reverse Intersystem Crossing and Unity
Quantum Yield via Multiple Singlet–Triplet State Pairs

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Jessica E. Stacey, Thomas J. Penfold, Andrew P. Monkman,* and Martin R. Bryce*

By inverting the common structural motif of thermally activated delayed
fluorescence materials to a rigid donor core and multiple peripheral accep-
tors, reverse intersystem crossing (rISC) rates are demonstrated in an organic
material that enables utilization of triplet excited states at faster rates than
Ir-based phosphorescent materials. A combination of the inverted structure
and multiple donor–acceptor interactions yields up to 30 vibronically coupled
singlet and triplet states within 0.2 eV that are involved in rISC. This gives
a significant enhancement to the rISC rate, leading to delayed fluorescence
decay times as low as 103.9 ns. This new material also has an emission
quantum yield ≈1 and a very small singlet–triplet gap. This work shows that
it is possible to achieve both high photoluminescence quantum yield and fast
rISC in the same molecule. Green organic light-emitting diode devices with
external quantum efficiency >30% are demonstrated at 76 cd m$^{-2}$.

1. Introduction

Organic light-emitting diodes (OLEDs) have become a central
part of materials research, with the ever-growing requirement
for more efficient, higher quality display devices. There is sig-
nificant interest in OLED materials which emit light via a ther-
maslly activated delayed fluorescence (TADF) mechanism$^{[1]}$ that
converts dark, triplet excited states to emissive singlet states by
reverse intersystem crossing (rISC). This can be achieved using
aromatic donor–acceptor (D–A) molecules, which typically
are conjugationally separated with the D and A units orthog-
onal. These systems emit from a singlet charge transfer state
($^1$CT), which is energetically very close to its $^3$CT state through
minimized electron exchange. A further excited state such as a local excited triplet
state ($^3$LE) situated very close in energy to this $^1$CT is also required.$^{[2]}$ Therefore,
having a small singlet–triplet gap ($\Delta E_{ST}$) is crucial, but is not the only requirement
for efficient rISC. rISC can harvest up to 100% of triplet states into singlet states.$^{[3]}$
Currently, the main challenges facing the TADF community are the long overall
residence times of emitter molecules in triplet excited states, and the low oscillator
strengths of the $^1$CT radiative transitions. Here, we report a new TADF molecular
design, incorporating a rigid, planar, central donor unit with multiple acceptor
units bound via C–N bridges. This new design gives a key step forward in TADF
efficiency through multiple coupled singlet–triplet states. The
resulting fast rISC rates lead to delayed fluorescence (DF) emission
lifetimes shorter than the phosphorescence lifetimes of
most Ir complexes currently used in OLEDs.$^{[4]}$ Critically, a unity
photoluminescence quantum yield (PLQY) is also maintained.

Recent research has shown that the underlying spin–flip
mechanism in rISC is a second order spin–vibronic process.$^{[2,5]}$
Here, specific molecular vibrations promote mixing between a
manifold of singlet and triplet states driving efficient rISC. Other
vibrational modes can contribute more to nonradiative decay,
requiring careful molecular design.$^{[6]}$ Gibson and Penfold have
recently published further work detailing this process.$^{[7]}$ Pre-
nviously synthesized, 1-substituted phenothiazine (D) D–A–D

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TADF candidates show molecular restriction with several con-
formers in solution on the 1H NMR timescale.[8] As well as the
phenothiazine donor being tilted, it is clear that there is some
rotational restriction around the C–N bond in these systems,
which switches off TADF due to a lack of vibronic coupling.
The conformation of the phenothiazine with respect to the acceptor
is also important in these molecules.[9] All of the above factors
must be taken into account when considering new molecular
designs. Another key challenge in the design of TADF molecules
is to balance the rates of rISC (and intersystem crossing, ISC)[7]
with the fluorescence quantum yield (ΦF). Ideally, the desired
molecule should have a ΦF close to 1 with a short emissive state
lifetime. This requires strong coupling of the 1CT to the ground
state. However, to ensure near degenerate 1CT, 3CT, and 3LE
states (which is a requirement for efficient rISC), D–A orthogo-
nality is required. This effectively decouples the 1CT states from
the ground state. Therefore, either a compromise is required,
or nonradiative quenching to the ground state, i.e., internal
conversion, must be dramatically curtailed. The new TADF mol-
ecule based on a triazatruxene central donor
functionalized with three peripheral accep-
tors, TAT-3DBTO2 (Scheme 1) is now shown
to overcome many of the issues faced when
designing an efficient TADF emitter.

2. Results and Discussion

2.1. Synthesis and Chemical Characterization

The design of TAT-3DBTO2 (Scheme 1) is based upon the reversal of the donor and
acceptor motif typically found in current TADF emitters.[10] Tri-PXZ-TRZ was prepared
by Tanaka et al. and has a central triazine acceptor unit and three peripheral phenoxa-
zine donor units; this work shows the benefits of symmetry as the PXZ-TRZ and bis-PXZ-
TRZ analogs showed lower device efficiency compared to the C3 symmetric Tri-PXZ-TRZ.
In TAT-3DBTO2 the threefold C3 symmetry is maintained, as in Tri-PXZ-TRZ, but the donor
is a central triazatruxene core (TAT). On this, TAT-3DBTO2 was synthesized in 40% yield and has good
solubility in various organic solvents, allowing for efficient
synthesis and purification of the molecule. This is an efficient
synthesis considering that three Buchwald–Hartwig couplings
were performed within one overnight reaction.

The 1H NMR spectrum of TAT-3DBTO2 at room temperature
(298 K) shows a mixture of broad and sharp peaks, suggesting
that parts of the molecule are rotating slowly on the NMR
timescale, giving rise to multiple environments for the same
protons. This has been shown to be the case by using variable
temperature (VT) 1H NMR, see Figure 1, and further 2D NMR
experiments in the Figures S5–S9 (Supporting Information).

The VT NMR data in combination with pure shift 1H NMR
studies (Figure S6, Supporting Information) show that there
are different conformers of TAT-3DBTO2 in solution; and
1H rotating frame nuclear Overhauser spectroscopy (ROESY)
NMR experiments (Figure S9, Supporting Information) con-
firm that these conformers all interconvert between each other.
The VT 1H NMR data indicate that there is an energy barrier to

Figure 1. Temperature dependent solution state 1H NMR spectra of TAT-3DBTO2 in dimethyl-
sulfoxide-d6 (DMSO-d6). The VT NMR data in combination with pure shift 1H NMR studies
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The VT 1H NMR data indicate that there is an energy barrier to
rotation around the D–A bridging bond and that increasing the temperature to 353 K overcomes this barrier. It is suggested that these conformers relate to the orientation of the three acceptor units with respect to each other and the triazatruxene core (see Section 2.2).

The highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) energies for TAT-3DBTO₂ were estimated by cyclic voltammetry and differential pulse voltammetry (see Figure S10 in the Supporting Information) at −5.60 and −3.00 eV, respectively. Within the respective solvent windows, as well as a reversible single-electron reduction, TAT-3DBTO₂ displays three reversible well-resolved single-electron oxidations (ΔE_{1/2}^{Eox(1)/Eox(2)} = 487 mV, ΔE_{1/2}^{Eox(2)/Eox(3)} = 603 mV). These correspond to sequential oxidations of the TAT core unit.

2.2. Quantum Chemistry Studies

Figure 3 shows the ten possible conformers of TAT-3DBTO₂, all of which are within 0.03 eV of each other, reflecting the results from the VT ¹H NMR studies (Figure 1). The different conformations consist of different combinations of acceptor orientations with respect to the donor moiety.

2.2.1. Electronic Structure—Absorption

The absorption spectrum of TAT-3DBTO₂ has been computed in the gas phase including the twenty lowest singlet and triplet states. The absorption spectrum exhibits three dominant peaks

![Figure 3](image-url)
2.2.2. Electronic Structure—Emission/Relaxation

The emission energy for each conformer is simulated by optimizing the lowest singlet and triplet states. In each case, a rotation of one acceptor moiety to become nearly orthogonal with respect to the donor core is observed. This motion allows minimization of the overlap between the orbitals involved in the excitation and therefore decreases the singlet–triplet energy gap ($\Delta E^{(T_1)/S_1} = 0.01 \text{ eV}$). The minimum energy geometry of both $S_1$ and $T_1$ is very similar (0.001) as seen in Figure 3. Consequently, one can expect three energy minima in $S_1$ corresponding to a charge transfer from the donor core to each of the different acceptor moieties. We thus find that within 0.2 eV of the $T_1$ state, there are 12 excited states, all of which are likely to be vibrationally coupled. If one considers all of the angular momentum components (i.e., all $M_i$ levels of the triplets), then TAT-3DBTO$_2$ has potentially 30 coupled states involved in rISC. This will give significant enhancement to the rISC rate.

The oscillator strength of TAT-3DBTO$_2$ is very similar (0.001) to a literature TADF material DPTZ-DBTO$_2$ (0.0007), as expected by the similar lifetime of the prompt emission in both molecules. Consequently, from these quantum chemistry simulations, it would appear that the enhanced performance of TAT-3DBTO$_2$ is primarily associated with the enhanced rISC rate derived from the higher density of states.

2.3. Photophysical Properties

2.3.1. Solution Properties

Figure 4a shows the extinction coefficient spectra of TAT-3DBTO$_2$ and the individual D and A units in dichloromethane ($\text{CH}_2\text{Cl}_2$). By comparison to the individual D and A units, the extinction coefficient at all wavelengths is greatly enhanced in TAT-3DBTO$_2$. This increase in absorption intensity strongly reflects the higher density of states predicted from the quantum chemical calculations described above. Particularly, the absorption band at lower energy (350–425 nm), which is not observed in the D or A units, and is ascribed to a direct absorption from the CT states, is very strong in this new material. Figure S15 (Supporting Information) shows a slight redshift on the right edge of the spectra.
by increasing the polarity of the solvent, which is associated with a strongly mixed $n \rightarrow \pi^*$ / $\pi \rightarrow \pi^*$ character transition,[13] also confirmed by its relatively strong transition. Excitation into this band directly populates $^1$CT excited states, as we have shown in the analogous D–A–D system DPTZ-DDBTO$_2$.[14]

Figure 4b shows the photoluminescence (PL) spectra of TAT-3DBTO$_2$ in different solvents together with the separated D and A units in toluene solution. The spectra show clear and strong CT emission, displaying a Gaussian band shape and strong redshift compared to the individual D and A emission spectra. The PL spectra shift to longer wavelengths upon increasing the solvent polarity. This indicates strong positive solvatochromism, as observed in other D–A–D-type molecules.[15,16]

2.3.2. Solid State Properties

BCPO, (bis-4-((N-carbazolyl)phenyl)phenylphosphine oxide)[15] was used as a host for TAT-3DBTO$_2$ to maintain the low energy splitting between $^1$CT and $^3$LE with correct host polarity. The polarity of the P–O bond in BCPO redshifts the $^1$CT energy compared to 1,3-bis(N-carbazolyl)benzene (mCP) matrix. The $^1$CT is tuned so that it is very close in energy to the $^3$LE state. Tuning the $\Delta E_{ST}$ in such a fashion has previously been demonstrated in the literature. The correct host polarity in this context is defined as a host with a polarity that will minimize the $\Delta E_{ST}$.[15,16] Furthermore, a PLQY of approximately unity was obtained from evaporated films of TAT-3DBTO$_2$-BCPO (see Section S16 in the Supporting Information).

Three different emission decay regimes are observed. Figure 5a: region I is fast decay, associated with prompt CT emission (PF); region II is early time DF; and region III is long-lived DF. The PF decay curves show no temperature dependence, indicating negligible migration of the singlet $^1$CT excited state. The decay curve at 320 K was fitted using a biexponential function: $\tau_1 = 10 \text{ ns}$ ($I_1 = 4.4$) and $\tau_2 = 35 \text{ ns}$ ($I_2 = 1.2$). Consequently, $\tau_{\text{average}} = 22.4 \text{ ns}$ for region I (see Section S17 in the Supporting Information). Region II shows strong TADF, the DF emission increasing in intensity with increasing temperature. The decay times related to region II are $\tau_1 = 103.9 \text{ ns}$ ($I_1 = 42 777$), $\tau_2 = 3.2 \mu s$ ($I_2 = 112 006$), and $\tau_3 = 15.1 \mu s$ ($I_3 = 61 040$). Consequently, $\tau_{\text{average}} = 11.7 \mu s$ for region II (see Section S17 in the Supporting Information). Usually, DF lifetimes of TADF emitters are in the microsecond timeframe, whereas TAT-3DBTO$_2$ has a DF component with a lifetime on the order of 100 ns, which is a result of fast rISC. Hofbeck and Yersin showed that $\text{fac-Ir}(ppy)_3$ has multiple component emission lifetimes ($\tau_1 = 200 \text{ ns}$, $\tau_2 = 6.4 \mu s$, $\tau_3 = 116 \mu s$).[14] The fastest delayed component of TAT-3DBTO$_2$ emission (103.9 ns) is faster than any emission component from $\text{fac-Ir}(ppy)_3$. This demonstrates that fast emission can be achieved without the need of a heavy metal.

As the PLQY for TAT-3DBTO$_2$ in BCPO is ~1, this is not a quenched component, and from its fit weighting it represents ~20% of all the delayed emissions. This complex multicomponent DF decay is ascribed directly to the multiple conformations possible in TAT-3DBTO$_2$. Region III has an inverse temperature dependence: the intensity of the emission increases as the system temperature drops. This has been observed before in highly efficient TADF molecules, and is associated with longer-lived DF components and phosphorescence (PH) at low temperatures.[14c]

The reverse intersystem crossing rates ($k_{\text{ISC}}$) of TAT-3DBTO$_2$:BCPO film (320 K) were calculated using two different approaches (see Section S18 in the Supporting Information). Three different values of $k_{\text{ISC}}$ were calculated (see Table 1), each value is associated with a distinct lifetime of DF in a TAT-3DBTO$_2$:BCPO film. The fastest lifetime of the DF emission ($\tau_1$ in region II) gives very high $k_{\text{ISC}} > 10^7 \text{ s}^{-1}$. Both the $k_{\text{ISC}}$ calculation methods show good agreement. Calculating the ISC rate, $k_{\text{ISC}}$, using $\Phi_{\text{ISC}} = \tau_{\text{prompt}} k_{\text{ISC}}$ (using the data from Section S18 in the Supporting Information) yields $k_{\text{ISC}} = 3.5 \times 10^7 \text{ s}^{-1}$. Taking the fastest $k_{\text{ISC}} = 1.5 \times 10^7 \text{ s}^{-1}$ (see Section S18 in the Supporting Information), we see that $K_{\text{ISC}} = K_{\text{ISC}}$ and so it is not surprising that the TADF becomes so efficient. The recycling rate of singlet to triplet and back approaches 1 in this case, indicative of very fast and efficient rISC.[14d] Figure 5b shows the area-normalized emission spectra in region I at 320 K. The PF emission shows a continuous dynamic redshift. This redshift is associated with the energetic relaxation of the $^1$CT state, primarily due to rotation about the D–A bond. Calculations and experiments suggest the D and A units twist toward a more orthogonal geometry, and stabilize in around 70 ns. Region II (Figure 5c) shows stabilized $^3$CT emission at 320 K: the onset of each spectrum collected in this region is at 2.78 ± 0.02 eV. The intensity dependence of the DF emission in this region as a function of the laser excitation dose was found to be linear with a gradient of 1, indicative of TADF (see Section S19 in the Supporting Information). Figure 5d shows late time decay (weak emission). Between 70 and 400 µs (still exponential decay), emission as in region II is observed. From 400 µs to 10 ms (power law decay), very weak emission is detected. Likely, this region includes DF emission from additional conformers and weak PH emission.

Spectral analysis at 80 K was performed to identify the PH emission. The harvesting of triplet states to singlet states in BCPO host is so rapid that obtaining a clear PH spectrum is problematic. This is due to residual $^1$CT emission masking the very weak PH emission. Therefore, the PH spectrum was measured in mCP. In this host the $\Delta E_{ST}$ is larger (0.21 ± 0.03 eV), allowing the PH spectrum to be clearly identified at low temperature (80 K) (see Section S20 in the Supporting Information). The TAT-3DBTO$_2$:mCP PH spectrum was also compared to the PH spectrum collected in polyethylene oxide matrix, and both the spectra show the same onset energy (see Section S20 in the Supporting Information).

Figure 5e shows the PH spectra of TAT-3DBTO$_2$:mCP film and the A and D units. The PH spectrum of TAT-3DBTO$_2$ shows mostly $^3$LE character from the acceptor units, while a peak around 550 nm is strongly enhanced. Comparison of the phosphorescence vibronic intensities may indicate a perturbed geometry for the LE triplet state in TAT-3DBTO$_2$ compared to the isolated acceptor unit. Considering that the $^3$LE states are almost unaffected by the polarity of the host environment, the triplet levels of TAT-3DBTO$_2$ doped into BCPO will have onsets very close to those observed in mCP and polyethylene oxide.

Figure 5f shows the PH spectrum together with the PL spectrum of TAT-3DBTO$_2$:BCPO film, for better comparison. The $^1$CT and $^3$LE states have onset energies of 2.78 ± 0.02 and
2.75 ± 0.02 eV, respectively, leading to $\Delta E_{ST} = 0.03 \pm 0.03$ eV. Therefore, it is clear that for TAT-3DBTO$_2$ in BCPO host, the $^1$CT state energy lies very close to the triplet states, as required for fast rISC and highly efficient TADF. However, we also note that there are twelve states very close in energy (described above), which would also couple to mediate rISC.

### 2.4. OLED Performance

BCPO is an ideal ambipolar host for TAT-3DBTO$_2$, yielding the minimal $\Delta E_{ST}$ and a PLQY of ≈100%. Optimization studies concerned finding the best guest-host ($x$:$y$) ratios. Therefore, two different device architectures were used: one designed for optimization of maximum external quantum efficiency.

**Table 1.** rISC rates determined from the three exponential decays from TAT-3DBTO$_2$:BCPO film using approach a ($k_{ISC}^a$) and approach b ($k_{ISC}^b$) for comparison.[14a,b]

<table>
<thead>
<tr>
<th>Decay component</th>
<th>$k_{ISC}^a$</th>
<th>$k_{ISC}^b$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$r_1$ = 10.3 ns</td>
<td>$1.5 \times 10^7$ s$^{-1}$</td>
<td>$1.3 \times 10^7$ s$^{-1}$</td>
</tr>
<tr>
<td>$r_2$ = 3.2 $\mu$s</td>
<td>$4.9 \times 10^3$ s$^{-1}$</td>
<td>$4.3 \times 10^3$ s$^{-1}$</td>
</tr>
<tr>
<td>$r_3$ = 15.1 $\mu$s</td>
<td>$1.0 \times 10^4$ s$^{-1}$</td>
<td>$9.2 \times 10^4$ s$^{-1}$</td>
</tr>
</tbody>
</table>
(EQE) values (OLED 1) and the another aiming for low roll-off (OLED 2). For optimization of maximum EQE, a lower amount of TAT-3DBTO2 was coevaporated with BCPO host, 1:9 v/v, and for optimization of roll-off the ratio of TAT-3DBTO2 to host was higher (1.7:8.3). The architecture of the optimized devices was: indium-tin-oxide (ITO)/NPB(40 nm)/TCTA(10 nm)/TAT-3DBTO2:BCPO(x:y,30 nm)/TPBi(40 nm)/LiF(1 nm)/Al 100 nm). NPB (N,N′-bis(naphthalen-1-yl)-N,N′-bis(phenyl)-benzidine) and TCTA (tris(4-carbazol-9-ylphenyl)amine) were used as commercial hole transport layers, TPBi (1,3,5-tris(N-phenylbenzimidazol-2-yl)benzene) as electron transport layer, LiF (lithium fluoride) as electron injection layer, and Al (aluminum) was used as cathode.

Figure 6a shows the green electroluminescence (EL) spectra of both the devices collected at 10 V. The Commission Internationale de L’Eclairage xy chromaticity coordinates for these EL spectra are (0.26, 0.46) and (0.29, 0.50) for OLED 1 and OLED 2, respectively. The emission from OLED 2 is slightly redshifted, which is likely associated with the increase in the overall polarity of the emissive layer induced by the increased TAT-3DBTO2 concentration.

Figure 6b shows representative EQE versus brightness curves. OLED 1 shows a maximum EQE value of 30.9% (76 cd m⁻²). Given this high EQE value, and the fact that the PLQY was found to be ≈100%, it can be concluded that the device has a charge balance (γ) close to unity. This implies that all the triplet excitons are harvested to singlet states, (ηST = 1), assuming an outcoupling efficiency (ηout) of ≈0.3 (see Section S21 in the Supporting Information). At 1000 cd m⁻², OLED 1 shows an EQE above 15%, exhibiting good resistance to roll-off with maximum brightness values up to 10 000 cd m⁻² (EQE = 4.4%). By increasing the concentration of TAT-3DBTO2 molecules in the emissive layer (OLED 2), the maximum EQE value drops to 20.2% (74 cd m⁻²), but significantly lower efficiency roll-off is observed. At 10 000 cd m⁻², OLED 2 shows an EQE of 8.8%, with brightness levels reaching 18 410 cd m⁻² (EQE = 3.9%).

Figure 6c shows the current density versus voltage. Both OLEDs show very low turn-on voltages of ≈2.5 V. However, in OLED 1, this value is slightly lower, which may be associated with the fact that TAT-3DBTO2 molecules are not as ambipolar as BCPO, so by decreasing its concentration, a better J–V curve (lower turn-on voltage) is observed. The same explanation holds for the current efficiency (ηc) versus voltage curves (Figure 6d) up to 8 V: both the devices show similar current efficiency (ηc1,max = 50.8 cd A⁻¹, ηc2,max = 52.9 cd A⁻¹), although at higher voltages OLED 1 exhibits much better resistance to high current efficiency levels. Table 2 highlights all the electrical properties of these devices and the values of each efficiency at 100 and 1000 cd m⁻², showing their electrical stability.

The reproducibility of these devices with such high EQEs and low roll-offs was studied in more detail (see Section S22 in the Supporting Information). Several other sets of devices with slightly distinct device structures also show EQE values around 30%. Therefore, the data presented in the main paper are the most representative among the OLEDs tested.
Table 2. Electrical properties of OLED 1 and OLED 2. $V_{on}$ = Turn on voltage; $\eta_{ext}$ = External quantum efficiency; $\eta_{brt}$ = Brightness; $\eta_{P}$ = Current efficiency; and $\eta_{P}$ = Power efficiency. Subscript 100 and 1000 refers to values taken at 100 and 1000 cd m$^{-2}$, respectively.

<table>
<thead>
<tr>
<th>Device</th>
<th>$V_{on}$</th>
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<th>$\eta_{brt,\text{max}}$ [cd m$^{-2}$]</th>
<th>$\eta_{c,\text{max}}$ [cd A$^{-1}$]</th>
<th>$\eta_{P,\text{max}}$ [lm W$^{-1}$]</th>
<th>$\eta_{ext,100}$ [%]</th>
<th>$\eta_{c,100}$ [cd A$^{-1}$]</th>
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<td>2.3</td>
<td>30.9$^{a}$</td>
<td>10 420</td>
<td>50.8</td>
<td>38.7</td>
<td>29</td>
<td>49.8</td>
<td>38.0</td>
<td>16.5</td>
<td>42.9</td>
<td>21.8</td>
</tr>
<tr>
<td>OLED 2</td>
<td>2.5</td>
<td>20.2$^{b}$</td>
<td>18 410</td>
<td>52.9</td>
<td>44.1</td>
<td>19</td>
<td>49.5</td>
<td>43.8</td>
<td>16.6</td>
<td>49.1</td>
<td>32.4</td>
</tr>
</tbody>
</table>

$^{a}$At 76 cd m$^{-2}$; $^{b}$At 74 cd m$^{-2}$.

3. Conclusion

In conclusion, TAT-3DBTO$_2$ introduces a new design for TADF emitters. The multi-acceptor single-donor motif imparts a large number of energy states which gives a short prompt 1CT lifetime and unitary PLQY. Moreover, we find 12 singlet–triplet state (pairs) within 0.2 eV of each other, which we believe gives rise to a DF component with a very fast rISC rate, on the order of $1 \times 10^{-7}$ s$^{-1}$. This shows that it is possible to achieve both a unitary PLQY and a sub-microsecond TADF lifetime in the same molecule. The conformational complexity of the molecule, however, gives rise to different rISC rates, as observed in the emission decays. Nevertheless, in devices, these optimal photophysical properties translate into an EQE which exceeds 30% at a useful brightness of 76 cd m$^{-2}$. Thus, this new TADF molecular design opens up a new dimension for achieving truly high performance TADF OLEDs and provides a solution to overcome the main concerns of current TADF molecular designs.

4. Experimental Section

Three types of samples were studied in this work: i) TAT-3DBTO$_2$ solutions ($10^{-3}$–$10^{-2}$ M) in methylcyclohexane, toluene, and dichloromethane (CH$_2$Cl$_2$) solvents; ii) drop-casted blend film of TAT-3DBTO$_2$:mCP 1:9 molar ratio; and iii) evaporated doped films of TAT-3DBTO$_2$:BCPO, 1:9 v/v. All the solutions were stirred for several hours to ensure complete dissolution. The films were dispersed onto quartz substrates.

Steady state absorption and emission spectra were acquired using a UV-3600 Shimadzu spectrophotometer and a Jobin Yvon Horiba Fluoromax 3, respectively. Time-resolved spectra were obtained by exciting the sample with a Nd:YAG laser (EKSPLA), 10 Hz, 355 nm or by using a nitrogen laser, 10 Hz, 337 nm. Exciting the sample with a UV-3600 Shimadzu spectrophotometer and a Jobin Yvon Horiba Fluoromax 3, respectively. Time-resolved spectra were obtained by exciting the sample with a Nd:YAG laser (EKSPLA), 10 Hz, 355 nm or by using a nitrogen laser, 10 Hz, 337 nm. Sample emission was directed onto a spectrograph and gated intensified charged couple device (ICCD) camera (Stanford Computer Optics).

OLED devices were fabricated using precleaned ITO–coated glass substrates purchased from Ossila with a sheet resistance of 20 $\Omega \cdot \text{cm}$. The small molecule and cathode substrates were previously purified by vacuum sublimation. In the emission decays. Nevertheless, in devices, these optimal photophysical properties translate into an EQE which exceeds 30% at a useful brightness of 76 cd m$^{-2}$. Thus, this new TADF molecular design opens up a new dimension for achieving truly high performance TADF OLEDs and provides a solution to overcome the main concerns of current TADF molecular designs.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

fast reverse intersystem crossing rates, thermally activated delayed fluorescence (TADF), triazatruxene

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