Perspectives of Unicolored Phosphor-Sensitized Fluorescence

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Unicolored phosphor-sensitized fluorescence (UPSF) is a dual emitting concept proposed for improving efficiencies and operational lifetimes of blue organic light emitting diodes (OLEDs). To overcome the limitations of the individual emitters, it uses a phosphorescent donor to sensitize a fluorescent acceptor. To quantify the potential of the concept, a multiscale model of a UPSF OLED is developed. It starts from atomistic morphologies, the rates of all processes on the available experimental data are parameterized, and the respective master equation is solved with the help of the kinetic Monte Carlo algorithm. The simulations show that the energy transfer between donor molecules is essential to reproduce the results of the time-resolved photoluminescence experiment. The scope of the experiment is expanded by studying the effect of the acceptor concentration, as well as Förster and (parasitic) Dexter energy transfer from the donor to acceptor, on the characteristics of the UPSF OLED. The study shows that an appropriate material design can further improve efficiency by more than 30% and at the same time achieve radiative decay times below 0.02 µs, thus significantly extending OLED operational lifetime.

1. Introduction

Organic light emitting diodes (OLEDs), utilize small organic molecules or polymers, in order to achieve an emissive electroluminescent layer. Ultra-thin, lightweight, and flexible characteristics offer a highly enticing substitute, in comparison to their inorganic counterpart. It is therefore understandable why there has been a large amount of research, focused on enhancing the efficiency and stability of OLEDs. At present, efficient and long-lasting, red[1] and green[2] OLEDs are achievable, with the weakest link being the blue OLED. Achieving a blue OLED, which is both efficient and stable, has proven to be problematic. The challenge originating with the limitations of the blue emitter: if operational lifetime is prioritized, stable fluorescent emitters can be used. Their efficiency, however, is limited by unfavorable spin statistics. To improve efficiency, phosphorescent emitters can be used. Their large coupling between the exciton spin and the orbital angular momentum allows for radiative decay from the triplet state to the ground state. Additional to this, the spin–orbit coupling allows for intersystem crossing to occur, such that the singlet excited state can also populate the triplet state, helping to achieve almost 100% internal quantum efficiency. The drawback of this highly efficient system is the long lifetime of the triplet state, typically in the order of several microseconds, much longer than the fluorescence lifetime, leading to degradation of the organic material.[3] Since stability is most important to achieve a long-lived consumer product, fluorescent emitters are the chosen source of blue OLEDs. But, with battery life on portable devices being the cost of this inefficiency, it is vital that the blue OLEDs become more efficient.

Thermally activated delayed fluorescence (TADF)[4–9] is one of the existing approaches targeting the OLED efficiency, where a reverse intersystem crossing, from triplet to singlet, is achieved. Combination of TADF and conventional fluorescence emitters, in a sensitizing approach is also a possibility.[10,11] However, the decay times of TADF systems are similar to that of a phosphorescent only system,[12,13] meaning that a short-lived OLED is inevitable.

A phosphor-sensitized fluorescence approach,[14–18] offers an alternative to TADF OLEDs, by utilizing a donor–acceptor concept with a phosphorescent donor and a fluorescent acceptor. In

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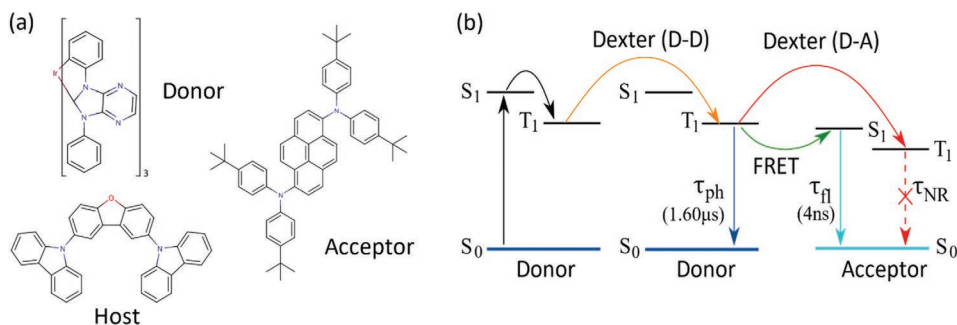+Present address: trinamiX GmbH, Industriestraße 35, 67063 Ludwigshafen am Rhein, Germany
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Figure 1. a) Molecular structure of the donor, acceptor, and host, used to bring the UPSF concept to a realization b) Energy level diagram of the UPSF system. Essential energy transfer, radiative (experimental decay times shown) and non-radiative decay: Initial excitation of the donor, donor-to-donor transfer (D-D), phosphorescent decay from the donor triplet (Ph), FRET from the triplet of the donor to the singlet of the acceptor, followed by fluorescent decay (fl), and Dexter energy transfer from the triplet of the donor to the triplet of the acceptor, followed by non-radiative decay (NR).

addition to phosphorescence and fluorescence, energy transfer can occur between the donor and acceptor molecules, either via short-range Dexter or long-range Förster energy transfer (FRET). FRET takes place from the donor triplet to the acceptor singlet, and allows for fluorescence.[19] This approach leads to shorter radiative decay times, in comparison to phosphorescent only emitters, as recently demonstrated by Kim et al.,[20] where a significant reduction of radiative decay time was shown for a green phosphorescent emitting donor and yellow fluorescent emitting acceptor.

Note that the traditional sensitization of a blue OLED would require a sensitizing donor emitting in the UV spectral range and red-shifted (with respect to the donor) acceptor emission. This approach is not suitable for devices, as high exciton energies would result in very fast device degradation. One can reduce the excitation energy required to pump the acceptor, if the fluorescent acceptor and phosphorescent sensitizer have matching emission spectra. In this case, however, the donor emission and acceptor absorption overlap becomes limited by the Stokes shift of the acceptor, that is, significantly reduced.

These challenges have been resolved in a recent study by Heimel et al., where a unicolored phosphor-sensitized fluorescence (UPSF) approach was used, for an efficient and stable blue OLED.[21,22] The donor, acceptor and host molecules, and a depiction of the processes, with radiative decay times and singlet and triplet energy levels, is shown in **Figure 1**. The sky-blue emission color is preserved, by matching the emission of donor and acceptor. A clear reduction of radiative decay from 1.60 μs, for phosphorescent only emitters, to 0.49 μs with the inclusion of fluorescent acceptors, was shown, leading to a threefold increase in the device lifetime, from 26 to 76 h, as shown in Note 1, Supporting Information (LT$_{70}$ lifetimes measured at initial current density of 25 mA cm^{-2}).[21]

The drawback of the sensitizing approach is the Dexter energy transfer, from the triplet of the donor to the dark triplet of the acceptor, by which a loss in efficiency arises. As the acceptor molecule is not of TADF-type, there is no reverse-intersystem-crossing (RISC) present. Therefore, Dexter triplet-triplet energy transfer results in quenching, since the transferred triplet state cannot decay radiatively, neither directly from the acceptor triplet nor through a RISC-process with subsequent delayed fluorescence. Unfortunately, with increasing acceptor concentration,

the probability of Dexter transfer increases. As a result, the photoluminescence quantum yield (PLQY) is shown to be directly linked to the concentration of the acceptor, with a decrease from 100% (no acceptors) to 63% (1.5 vol% acceptor molecules). Therefore, the UPSF OLED has to be designed in such a manner as to i) optimize the acceptor concentration for increased lifetime and ii) target donor–acceptor combinations with slow Dexter and fast FRET rates, for increased efficiency.

To provide further insight into the potential of the UPSF system, we first adjust the rates of the individual energy transfer processes to match experimentally measured PLQY, phosphorescent-fluorescent emission ratios, time resolved photoluminescence (TRPL) spectra, and radiative decay times. We then expand the scope of experiment by examining further acceptor concentrations, as well as FRET and Dexter energy transfer rates, highlighting what will be the fundamental efficiency and decay time limits of the UPSF system. The simulation workflow is shown in **Figure 2**.

2. Parametrization of Rates

In a PL experiment, all processes of interest within the UPSF OLED begin with an initial excitation of donor molecules, S_0^{donor} to S_1^{donor}, as initial excitation of the acceptor molecules was limited experimentally, as discussed in Note 1, Supporting Information, it was not included in the simulations. This is followed by a practically instantaneous transition from S_1^{donor} to T_1^{donor}, populating the first triplet T_1 state of the donor molecule, as shown in Figure 1. There are then a number of possibilities; first, triplet excitation can decay radiatively, T_1^{donor} to S_0^{donor} (phosphorescence). Second, the donor triplet can be transferred from one donor molecule to another, with a rate k_{DD}, via a Dexter energy transfer. We assume that this rate decays exponentially as the donor-donor separation (R_{DD}) increases, $k_{DD} = k_{DD}^0 e^{-\alpha_{DD}R_{DD}}$. Additionally, the donor triplet can undergo an energy transfer from donor to acceptor, either via the Dexter or Förster mechanism. For short range Dexter energy transfer, T_1^{donor} to $T_1^{acceptor}$, we assume that the Dexter rate exponentially decays with the donor–acceptor separation, R_{DA}, $k_{Dexter} = k_{Dexter}^0 e^{-\alpha_{DA}R_{DA}}$. This results in non-radiative decay from the $T_1^{acceptor}$ state, the rate of which is unknown from experiment. For donor–acceptor pairs at separations larger than 1 nm, FRET is the predominantly chosen pathway for the transition T_1^{donor} to $S_1^{acceptor}$, resulting in the fluorescent radiative decay, $S_1^{acceptor}$ to $S_0^{acceptor}$. Here we assume that the FRET rate depends on the inverse of the phosphorescent decay time (lifetime of the donor excited state),[23] k_{ph} as $k_{FRET} = k_{ph} \left(\frac{R_{FRET}}{R_{DA}} \right)^6$, where R_{FRET} is the Förster radius, at which 50% of the excited donors are transferred via FRET to the acceptor.
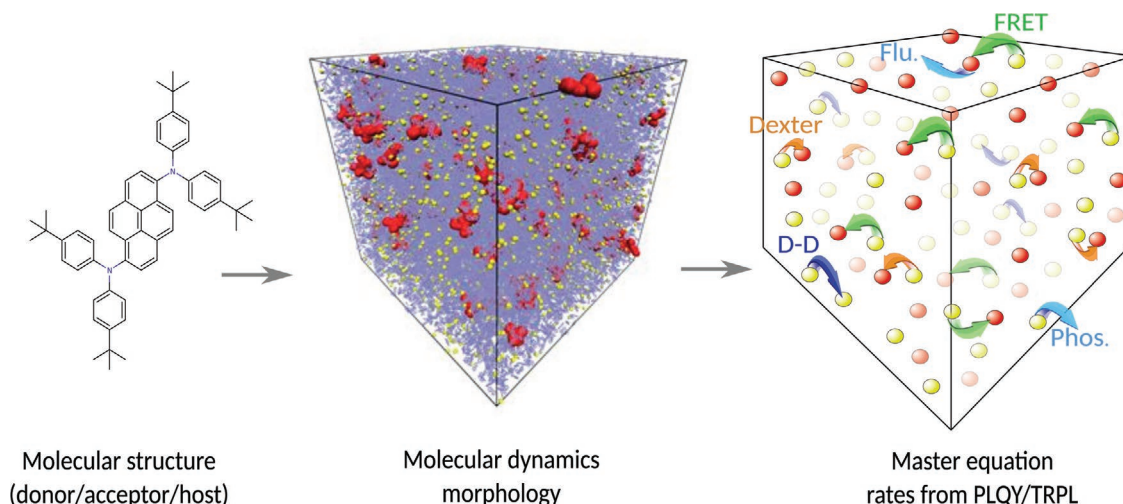
Figure 2. Simulation workflow: from the individual molecular structures, to molecular dynamics, followed by rate parametrization, using experimental data (PLQY and TRPL), then Kinetic Monte Carlo (KMC) to randomly propagate the system through time. Providing OLED properties, such as PLQY and radiative decay times.

Experimentally, three acceptor concentrations were investigated, 0.5, 1.0, and 1.5 vol% acceptor molecules.[21] In addition to, a donor only system and an acceptor only system, providing the radiative decay times of phosphorescence (1.60 µs) or fluorescence (4 ns), respectively. The inverse of the phosphorescence decay time yields the rate $k_{ph} = 6.25 \times 10^5$ s^{-1}. The Förster radius of the given donor–acceptor pair, was also estimated experimentally, with a value of $R_{FRET} = 2.4$ nm. Additionally, for each concentration, a PLQY value, a ratio of phosphorescence:fluorescence emission and a radiative decay time, with TRPL spectra are provided from experiment, included in Note 1, Supporting Information.

Using this experimental data, the unknown rate constants, for donor-to-donor energy transfer, k_{DD}^0, Dexter energy transfer, k_{Dexter}^0, and the non-radiative decay of acceptor, k_{NR}, can be determined. These rate constants represent averaged over assemblies of molecules quantities and hence are directly related to the atomistic-scale morphology. The morphology was generated using molecular dynamics simulations for all studied acceptor concentrations, according to Note 2, Supporting Information, and then converted into a master equation with three types of states (donor triplet state, acceptor triplet, and acceptor singlet states, all of which are presented in Note 3, Supporting Information) and six individual processes. The three unknown rates constants are then determined, as described below. This master equation was solved with the help of the Kinetic Monte Carlo (KMC) algorithm, outlined in Note 4, Supporting Information. The protocols used to obtain simulated photoluminescence (PL) spectra and PLQY values, are described in Note 5, Supporting Information. The unknown rate constants are initially estimated and then varied in accordance, until the experimentally achieved results have been reproduced, as outlined in Note 6, Supporting Information.

2.1. Donor-to-Donor Energy Transfer

It is unknown from experiment the extent to which donor-to-donor energy transfer plays a role in the overall description of

the UPSF system. To demonstrate that this process is essential for a quantitative description of the system, we first fitted the TRPL spectra without taking into account the donor-to-donor energy transfer, as shown in Note 7, Supporting Information, for the 1% acceptor concentration.

It is clear that donor-to-donor energy transfer cannot be omitted: short- and long-range decays cannot be reproduced simultaneously, with the estimated PLQY value higher than the experimentally achieved 66%, at 71%. Aside from this, the radiative decay time was found to be 1.44 µs, opposed to the 0.77 µs from experiment. Signifying, insufficient donor–acceptor Dexter events and a shift to slower radiative decay, as a result of more phosphorescence than expected. Therefore, the intermediate process of donor-to-donor transfer, which would facilitate more Dexter events and lower the number of phosphorescent photons emitted, has to be included. The rate constant, k_{DD}^0 was then varied, where an optimal value of around 5×10^5 s^{-1} was found. This value ensures that donor-to-donor transfer is not the predominant energy transfer in the UPSF system, but allows for adequate movement of energy between the donor molecules, in order to facilitate the required amounts of FRET and Dexter.

2.2. Donor-to-Acceptor Dexter Energy Transfer

In combination with other rate constant variations, the donor–acceptor Dexter rate constant was varied. As this Dexter rate constant increases, the PLQY value decreases. Hence, to match the correct PLQY values with experiment is rather simple. Despite this, the difficulty in accurately reproducing experimental emission is with the ratio of phosphorescence to fluorescence. If the Dexter rate is too fast, there are limited FRET events and so limited fluorescence emission. On the other hand, if the Dexter rate is too slow, there is much more FRET events than expected. It is for this reason that a further quantity, the Dexter cut-off has to be examined. This is defined as the maximum separation distance at which Dexter may occur. It is intuitive that the separation associated with Dexter must

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remain short, due to the nature of Dexter transfer. With this in mind, the Dexter cut-off and the rate constant are varied in accordance with one another, in order to achieve the correct PLQY and emission ratios, for each of the acceptor concentrations. A Dexter rate constant, k_{Dexter}^0, was found to be 2×10^7 s^{-1}, in combination with a (center-of-mass) Dexter cut-off of around 2 nm, compared to a maximum of around 5 nm for FRET.

2.3. Non-Radiative Decay from the Acceptor Triplet State

After a Dexter event has occurred, the first triplet state of the acceptor molecule is populated. The decay from this state is non-radiative, resulting in a loss in efficiency of the UPSF system. Upon this quenching mechanism, the acceptor becomes blocked due to the single occupancy constraint. The role of the non-radiative decay is to clear the occupied state of an acceptor molecule. Coupled with the aforementioned rate constants, the rate at which non-radiative decay is collected, was also varied. If the sites are blocked for a long period of time, phosphorescence is the predominant emission, if the sites are cleared too quickly, fluorescence emission is overestimated. The non-radiative emission was varied and optimized accordingly, resulting in a decay time of 0.02 μs, longer than that of fluorescence at 4 ns, but shorter than the 1.60 μs of phosphorescence.

2.4. Model Validation

The increase in acceptor concentration causes an increase in the amount of FRET events (consequently fluorescence), but also Dexter events. Ultimately, this shifts the emission from largely slower phosphorescence, toward faster fluorescence, giving a reduced radiative decay time. As the number of Dexter events also increases, it is expected and shown from experiment that the PLQY should decrease with increasing acceptor concentration. The PLQY values and all emission data, are from the contribution of both phosphorescent and fluorescent photons. With the rate constants optimized, averaging over the three acceptor concentrations, the final values of PLQY, emission ratios, and radiative decay times, in comparison to those achieved from experiment, are listed in **Table 1**, for each of the acceptor concentrations.

The radiative decay times are achieved from the plots of emission, in a simulated TRPL spectrum, making use of the multiexponential fitting method, described in Note 5, Supporting Information. An example of such spectrum and fit, for an acceptor concentration of 1%, are shown in **Figure 3**a,

together with the equivalent experimental fit. Additionally, for all three acceptor concentrations, the simulated multiexponential fits are plotted with the corresponding experimental TRPL multiexponential fits, in Figure 3b.

The correlation between the experimental and simulated TRPL multiexponential fits is clear and is coupled with the agreement of PLQY values and calculated radiative decay times, listed in Table 1, signifying an accurate theoretical representation of the UPSF system. The trends observed from both simulations and experiment, show that with increasing acceptor concentration, the radiative decay time is lowered due to a shift from slower to faster emission, or the increase in the number of fluorescent photons, from more FRET events. Additionally, the PLQY values decrease with more acceptor molecules, reinforcing the expectation of more Dexter events.
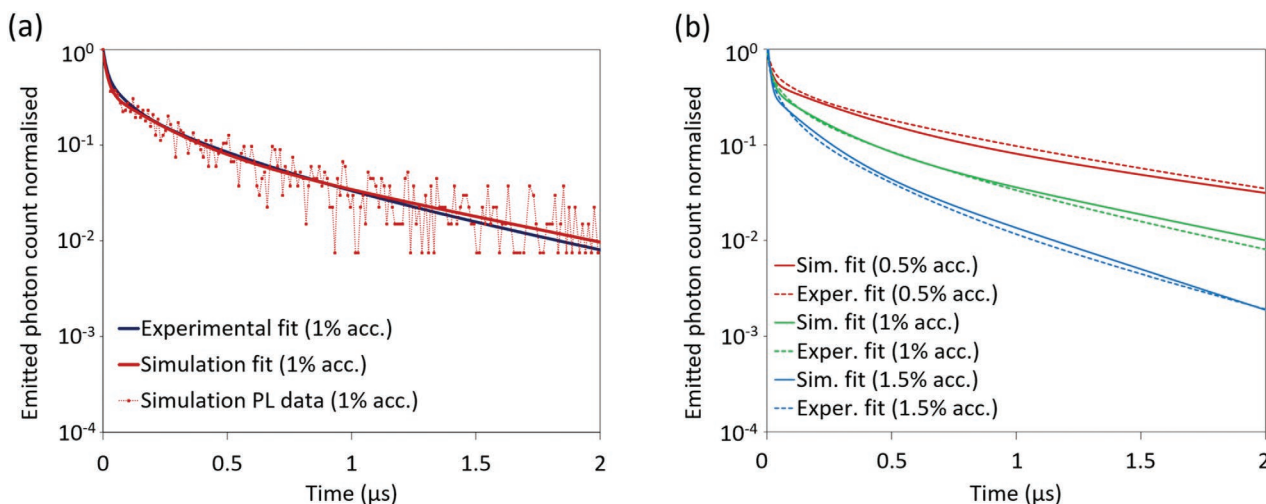
3. Beyond Experiment

3.1. Higher Acceptor Concentrations

With the simulation results closely matching that of experiment, it is possible to study the system in more depth and expand on the experimental results. To do this, three additional concentrations were chosen, with 2, 2.5, and 3 vol% acceptors. In the same manner as the previous concentrations, the PLQY values and radiative decay times were evaluated. The PLQY values were found to remain almost constant at around 62%, with values of 62.5% (2% acc.), 62.6% (2.5% acc.), and 61.8% (3% acc.). All simulated and experimental PLQY values are shown in **Figure 4**a. This indicates that the system reached a degree of saturation, in terms of the number of Dexter events, as a consequence of the slow, non-radiative, acceptor triplet decay. The radiative decay times also appeared to show a lower limit approaching, with 0.33, 0.20, and 0.19 μs, for the 2%, 2.5%, and 3% concentrations, respectively. The simulated multiexponential fits for all concentrations, are shown in Note 8, Supporting Information, indicating a saturation of FRET events and resulting fluorescence, where the addition of further acceptors has little impact on the radiative decay. The radiative decay rate, (the inverse of the radiative decay time), is plotted for each of the concentrations, with comparison to experiment, in Figure 4b. As a result of the radiative decay time limit, the OLED lifetime will also ultimately reach a limit, due to a direct correlation between radiative decay rates and the lifetime of the OLED, demonstrated experimentally.[21] Additionally, the long-lived acceptor triplet state can contribute to degradation of the UPSF OLED, leading to a further limitation of the device lifetime.

3.2. Relative Contributions of Phosphorescence and Fluorescence

A further possibility of understanding the UPSF system more clearly, is with the breakdown of the emission into the constituent phosphorescent or fluorescent photons. The experimental quantity of transfer efficiency, FRET efficiency, is defined as

Table 1. PLQY and radiative decay times from experiment and simulations, listed with increasing acceptor concentration.

Acceptor concentration [%]	PLQY (Experiment) [%]	PLQY (Simulations) [%]	τ_{rad} (Experiment) [μs]	τ_{rad} (Simulations) [μs]
0.5	82	76	1.06	1.35
1.0	66	67	0.77	0.83
1.5	63	62	0.49	0.41

Figure 3. a) Simulated photoluminescence (PL) plot for the 1% acceptor system, emitted photon counts normalized, plotted over a time of 2 µs, emission data (red dots and dashed line), and multiexponential fit (solid red line), with the equivalent experimental TRPL fit (blue). b) Simulated photoluminescence (PL) multi-exponential fits (solid lines) and equivalent experimental TRPL fits (dashed lines), for 0.5% (red), 1.0% (green), and 1.5% (blue) acceptor concentrations.

the proportion of excitons decaying radiatively due to FRET, between the donor and acceptor molecules, that is, the efficiency of the FRET process.[21] This quantity can be directly plotted with the donor and acceptor emission values, achieved with simulations, for each of the concentrations.

It is evident from **Figure 5** that there is a direct link between the amount of fluorescence predicted from simulations and efficiency of FRET found from experiment. Thus, it can be determined that the fluorescence observed is a direct consequence of FRET from the donor to the acceptor. This is an obvious conclusion from the simulated acceptor emission, as there are no direct (initial) acceptor excitations possible, therefore any fluorescence collected can only be due to FRET. The correlation of experimental FRET efficiency and the fluorescence emission of simulation, signify a further validation of the code and reinforce the accuracy of the representation

of the UPSF system. In addition to this, the trend for higher acceptor concentrations is also shown. It is clear that as the number of acceptors continues to increase, further than the 1.5% concentration, the fluorescence emission also continues to increase. That being said, it is clear that this increase begins to level out after 2.5% acceptor concentration, which is not obvious from experimental results, as a simple linear relationship was shown from the FRET efficiency values. The leveling of the fluorescence emission emphasises the idea of a saturation of acceptor molecules, as found from the lower limits of both PLQY and radiative decay values. This would signify that simply adding acceptor molecules to the UPSF system cannot reduce the radiative decay time to that of fluorescence alone. Since there exists a limit to the amount of fluorescence that is achievable, there is also a limit to the lifetime of the device.
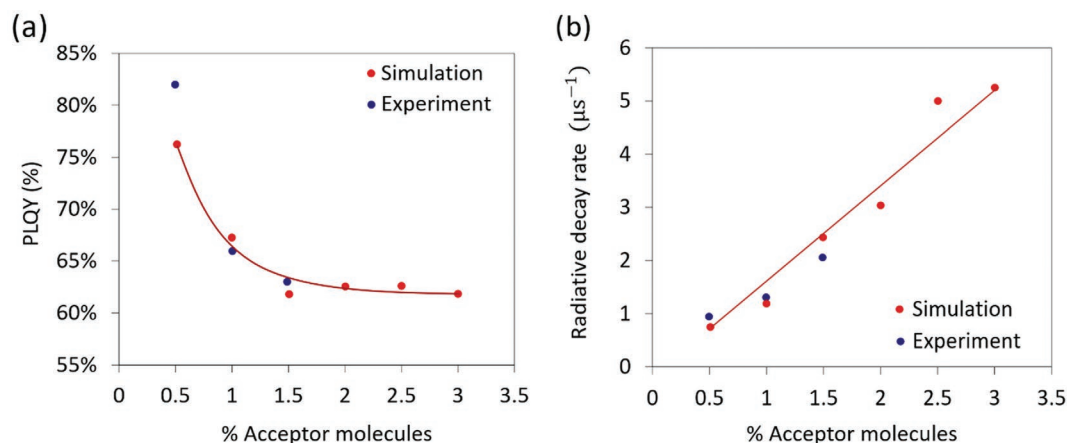
Figure 4. Comparison of simulated (red) and experimental (blue) a) PLQY values and b) radiative decay rates, for all acceptor concentrations. The red lines serve only as a visual aid.
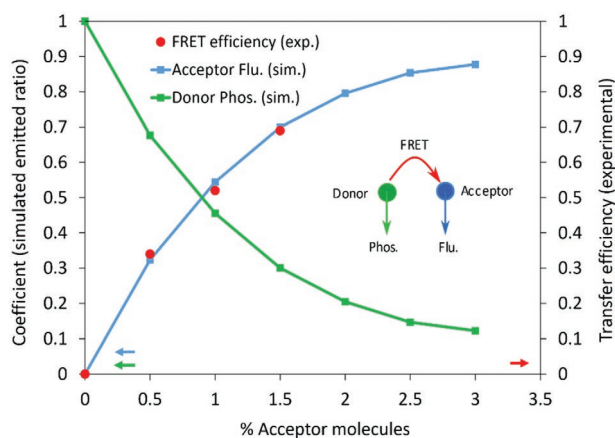
Figure 5. Left axis: Simulated emission coefficients, referring to the fraction of emission type, over total emission. Phosphorescence from the donor (green) and fluorescence from the acceptor (blue). Right axis: transfer efficiency calculated from experimentally achieved results, FRET efficiency (red).
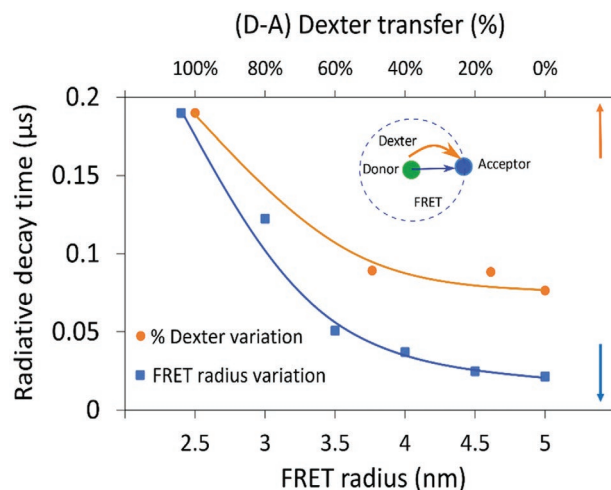
3.3. Supressing Dexter Transfer and Increasing the FRET Radius: An Ideal UPSF OLED

The undesired, donor to acceptor, Dexter events cost the UPSF system a loss of efficiency. However, the extent to which this Dexter transfer inhibits FRET, is unknown. Therefore, in the ideal case, which would be the complete suppression of donor–acceptor Dexter energy transfer, any change to the FRET events can be examined.

The extent to which Dexter energy transfer impacts FRET is clearly shown in **Figure 6**, the percentage of FRET events is calculated with respect to phosphorescence or phosphorescence and Dexter, in the case of no Dexter or with the inclusion of Dexter, respectively. In the normal UPSF system, where Dexter energy transfer would be present, the percentage of FRET events reaches a maximum of 54%, for the 3% acceptor concentration. On the other hand, in the ideal scenario, where no Dexter transfer occurs between donor and acceptor, this
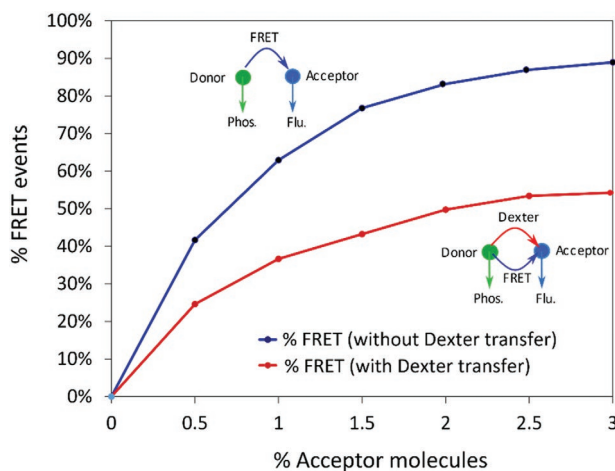
Figure 6. Simulation results of the percentage of FRET events, with respect to all events from the donor molecules, with (blue) and without (red) Dexter energy transfer, as a function of acceptor concentration.

Figure 7. Radiative decay time (µs) reduction, toward an ideal UPSF OLED. Top axis: The effects of supressing Dexter transfer (orange) or bottom axis: increasing the FRET radius (blue). The blue and orange lines serve only as a visual aid.

increases to 89%, an increase of 35%. This shows that limiting Dexter energy transfer would have a large impact on the amount of FRET and therefore fluorescence. The result of this would be that the UPSF OLED would have a significantly increased operational lifetime, as almost 90% of the events would result in fluorescence, moving away from the degradation of the long-lived phosphorescence state.

Evidently, the suppression of donor–acceptor Dexter transfer can lead to a potentially substantial decrease in the radiative decay time. This is shown in **Figure 7** for the 3% acceptor concentration, by reducing the Dexter transfer, from 100% in the original system, to 0% in the ideal case. A reduction by a factor of 0.4, leading to a radiative decay time of 0.08 µs, highlighting the significant impact of Dexter transfer. A second modification to the system, to achieve an ideal UPSF OLED, would be with an increase of the FRET radius, R_{FRET}. Also, independently demonstrated in Figure 7 for the 3% acceptor concentration. By increasing the FRET radius from 2.4 to 5 nm, a reduction by a factor of 0.1 was found, resulting in a radiative decay time of 0.02 µs, exceeding the effect of Dexter elimination. Therefore, when used in combination, the complete suppression of Dexter transfer and the increase of the FRET radius to 5 nm, would significantly improve the UPSF system, with simulations predicting a radiative decay time of 0.02 µs and a PLQY of 100%.

We should, however, note that tuning the FRET radius is particularly challenging in the UPSF system. Broadening of the donor emission will most likely result in higher-energy photons already in the initial phosphorescent OLED, leading to faster degradation. Broadening of the acceptor absorption will also broaden its emission, leading to a shift of the OLED color coordinate toward green. Another alternative is to align transition dipole moments of the donor and acceptor and increase the FRET rate by increasing the dipole–dipole interaction term. However, the dipole–dipole interaction depends not only on the relative molecular orientations, but also on the orientation of the vector connecting the molecules, which still varies in space. A simple estimate for a cubic lattice shows that the orientation-dependent

prefactor increases from $2/3 = 0.67$ for molecules oriented isotropically, to only ≈ 0.7 if donor and acceptor transition dipoles would be oriented perfectly in the plane of the substrate (while their connection vector remains isotropic), to 0.8 if donor and acceptor transition dipoles are perfectly aligned perpendicular to the substrate (while their connection vector remains isotropic). Only if donor–acceptor pairs are chemically bound so that their transition dipole moments are parallel to each other and to the vector connecting them, one can achieve a sizable prefactor of four. Hence, the chemical design of a dual donor–acceptor system is a clear scientific challenge.

4. Conclusions

We have estimated the potential of a UPSF system, which facilitates both singlet and triplet emission, keeping in mind its application in a blue OLED. We examined the individual energy transfer processes and optimized their rate constants to match experimental findings, hence building a multiscale model of a UPSF OLED. By expanding the scope of experiment, we have shown that the current set of materials is limited to radiative decay times of around 0.20 µs, which can be achieved by increasing the acceptor concentration (≈ 3 vol%). At these concentrations, the undesired Dexter energy transfer, which results in a loss of efficiency, also saturates, resulting in PLQY values around 60%. Both PLQY and radiative decay time cannot be improved by addition of further acceptors. In fact, being detrimental to this goal, as the continued population of the long-lived acceptor triplet, contributes to degradation of the OLED.

We also examine an ideal UPSF system, where there would be no Dexter energy transfer, the extent to which it limits the number of FRET events in the system was apparent. By stopping donor–acceptor Dexter transfer it is possible for the UPSF system to increase FRET (and hence fluorescence) by 35%. Additionally, if the FRET radius can be doubled, in combination with Dexter suppression, the UPSF system can achieve radiative decay times of around 0.02 µs, which is a remarkable decrease, illustrating the possibilities of the UPSF concept. The chemical design of such donor–acceptor combinations is, however, a challenge, as it easily leads to a trade-off between the FRET efficiency and the OLED color coordinate.

Supporting Information

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

Conflict of Interest

The authors declare no conflict of interest.

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