



















**Table 5. TRPL measurement analysis (at 581 nm) of the 552 nm emitting donors varying the A/D concentration ratio.**

TRPL analysis of 552 nm emitting dots at 581 nm with respect to changing A/D								
A/D	A <sub>1</sub>	τ <sub>1</sub> (ns)	A <sub>2</sub>	τ <sub>2</sub> (ns)	A <sub>3</sub>	τ <sub>3</sub> (ns)	τ <sub>int</sub> (ns)	χ <sup>2</sup>
1.8	1566	35.80	3515	15.18	8940	1.41	23.23	1.12
5.3	1042	34.53	2124	14.24	10977	1.44	20.67	1.10
8.9	774	34.15	1518	13.53	12159	1.46	18.64	1.12
14.2	508	34.86	1080	13.33	12809	1.48	16.38	1.16
21.3	486	32.94	913	12.23	13030	1.50	14.85	1.12
32.0	425	31.51	755	10.77	13304	1.49	13.05	1.14
53.3	278	31.41	543	9.88	13606	1.51	10.36	1.14
81.7	237	28.43	568	6.27	13723	1.48	7.96	1.12
110.1	209	25.08	4409	2.48	10629	1.18	6.05	1.05
138.6	181	24.82	5258	2.32	9634	1.13	5.51	1.06
152.8	167	24.72	4266	2.44	10793	1.19	5.22	1.05

**Table 6. TRPL measurement analysis (at 605 nm) of the 552 nm emitting donors varying the A/D concentration ratio**

TRPL analysis of 552 nm emitting dots at 605 nm with respect to changing A/D								
A/D	A <sub>1</sub>	τ <sub>1</sub> (ns)	A <sub>2</sub>	τ <sub>2</sub> (ns)	A <sub>3</sub>	τ <sub>3</sub> (ns)	τ <sub>int</sub> (ns)	χ <sup>2</sup>
1.8	701	42.56	1591	15.37	11970	1.46	23.28	1.13
5.3	548	40.07	1320	14.82	12481	1.51	19.85	1.11
8.9	478	36.82	971	13.21	13060	1.52	16.85	1.15
14.2	342	37.02	763	13.08	13383	1.53	14.63	1.14
21.3	306	33.70	594	10.39	13481	1.50	12.05	1.14
32.0	300	33.09	596	10.16	13407	1.53	11.55	1.17
53.3	255	27.88	897	4.70	13627	1.43	7.99	1.10
81.7	225	25.53	3185	2.73	11804	1.26	6.47	1.09
110.1	161	24.90	5866	2.28	9180	1.11	5.13	1.05
138.6	134	23.86	6681	2.16	8412	1.04	4.42	1.08
152.8	119	25.71	5840	2.25	9138	1.09	4.55	0.98

**Table 7. TRPL measurement analysis (at 581 and 605 nm) of the 581 nm emitting acceptors varying the A/D concentration ratio.**

TRPL analysis of 581 nm emitting Rhodamine B molecules at 581 nm							
A <sub>1</sub>	τ <sub>1</sub> (ns)	A <sub>2</sub>	τ <sub>2</sub> (ns)	A <sub>3</sub>	τ <sub>3</sub> (ns)	τ <sub>int</sub> (ns)	χ <sup>2</sup>
-17	0.001	8277	1.98	7105	1.006	1.68	1.88
TRPL analysis of 581 nm emitting Rhodamine B molecules at 605 nm							
A <sub>1</sub>	τ <sub>1</sub> (ns)	A <sub>2</sub>	τ <sub>2</sub> (ns)	A <sub>3</sub>	τ <sub>3</sub> (ns)	τ <sub>int</sub> (ns)	χ <sup>2</sup>
10225	1.89	25883	0.53	-22115	0.45	1.69	1.23

### 2.3 Light harvesting analyses and remarks

Figs 3 and 4 demonstrate clearly the effect of Förster radius on the lifetime modifications of the donor and acceptor molecules. For further analyses, we also calculate energy transfer efficiency and light harvesting enhancement of the acceptor emission. The energy transfer efficiency is extracted from the amplitude weighted donor lifetime,  $\tau_{amp}$  in the presence and absence of the acceptor molecules using Eq. (4)

$$\eta_{FRET} = 1 - \frac{\tau_{DA}}{\tau_D}. \quad (4)$$

Here  $\tau_{DA}$  is the amplitude weighted lifetime of donors in the presence of acceptors and  $\tau_D$  is that of donors in the absence of acceptors [26].

Figure 5a reveals the comparison of efficiency levels extracted from TRPL measurements. Here we observe that the energy transfer efficiency increases with the increased A/D concentration ratio, as the donors find more acceptors around them to transfer more of their excitation energy. Tuning the A/D concentration ratios and using better optimized 552 nm emitting quantum dot donors, we observe a maximum energy transfer efficiency of 86%, which is obtained at an A/D concentration ratio of 152.8 in our experiments. This comparison shows that the efficiency levels are higher using 552 nm emitting quantum dots than those of 525 nm emitting ones.

To show the effect of nonradiative energy transfer mediated light harvesting on the emission enhancement of the acceptor molecules, we further compute the light-harvesting factor for the acceptor emission (Fig. 5b). These calculations are carried out through fitting SSPL measurements (in Fig. 2) of the donor quantum dots to a Gaussian distribution and comparing the overall emission (donor + acceptor mixture) with the emission of only acceptors (corresponding to the same concentration of acceptor molecules used in each A/D concentration point). In these calculations, the tail overlap of the donor emission on the acceptor emission is also considered, and any possible contribution from the tail (although weak) is also subtracted. Here we observe that the relative enhancement factor of the acceptor emission is decreased with the increased A/D concentration ratio, because the acceptors increasing in number find fewer donors around them to be fed via nonradiative energy transfer, which indicates a tradeoff between the efficiency and light harvesting factor.

Also, we again observe that 552 nm emitting aqueous CdTe quantum dots are better light-harvesting antennas in water for Rhodamine B dye molecules in comparison to those quantum dots emitting at 525 nm. In the light of these experiments and analyses, such light harvesting is possible; however, one needs to consider the tradeoff between efficiency and enhancement factor to choose an operating point. These results also indicate that nonradiative energy transfer assisted light harvesting may enable quantum dot multiplexed dye biodetection systems, on which we are currently working.

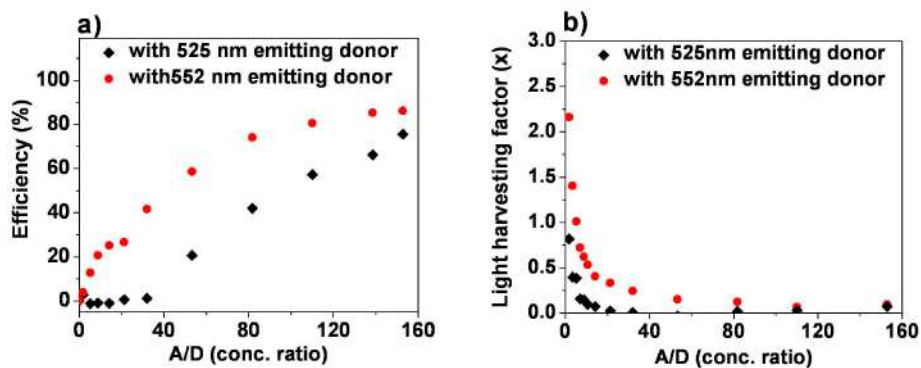


Fig. 5. Comparison of (a) FRET efficiencies and (b) enhancement of the acceptor emission, using 552 nm and 525 nm emitting CdTe quantum dot donors, as a function of the A/D concentration ratio.

### 3. Conclusion

In conclusion, as a proof-of-concept demonstration, we have presented nonradiative energy transfer based light harvesting of aqueous colloidal CdTe quantum dot antennas for dye molecules in water. Our experiments show that these quantum dots used as donors need to be carefully optimized to match Rhodamine B used as acceptors. In our experiments, we have observed strong lifetime modifications of these CdTe quantum dots from 25.3 to 7.2 ns. We have demonstrated the energy transfer efficiency tuning up to 86% as the acceptor-donor

concentration ratio is varied. These experiments indicate that nonradiative energy transfer mediated light harvesting using aqueous quantum dots leads to enhanced emission of dye molecules in water at wavelengths beyond the absorption range of the dyes. One should also note that a good operating point in the A/D concentration ratio for a specific donor-acceptor pair has to be set to provide both reasonably high efficiency and high light harvesting of the acceptor emission. This nonradiative energy transfer assisted light harvesting holds great potential for future quantum dot multiplexed biological and optoelectronic applications.

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