Supplementary information

Improving the photoluminescence quantum yields of quantum dot films through a donor/acceptor system for near-IR LEDs

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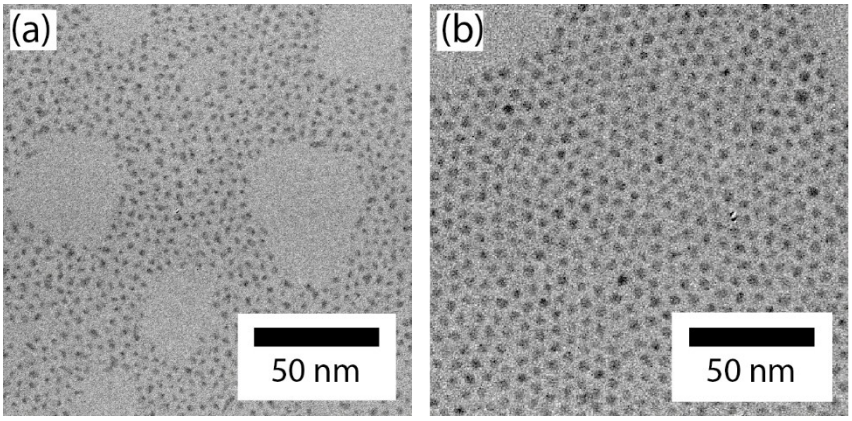


SI Figure 1: (a)TEM image of donor QDs. (b)TEM image of acceptor QDs.

Transient Absorption details:

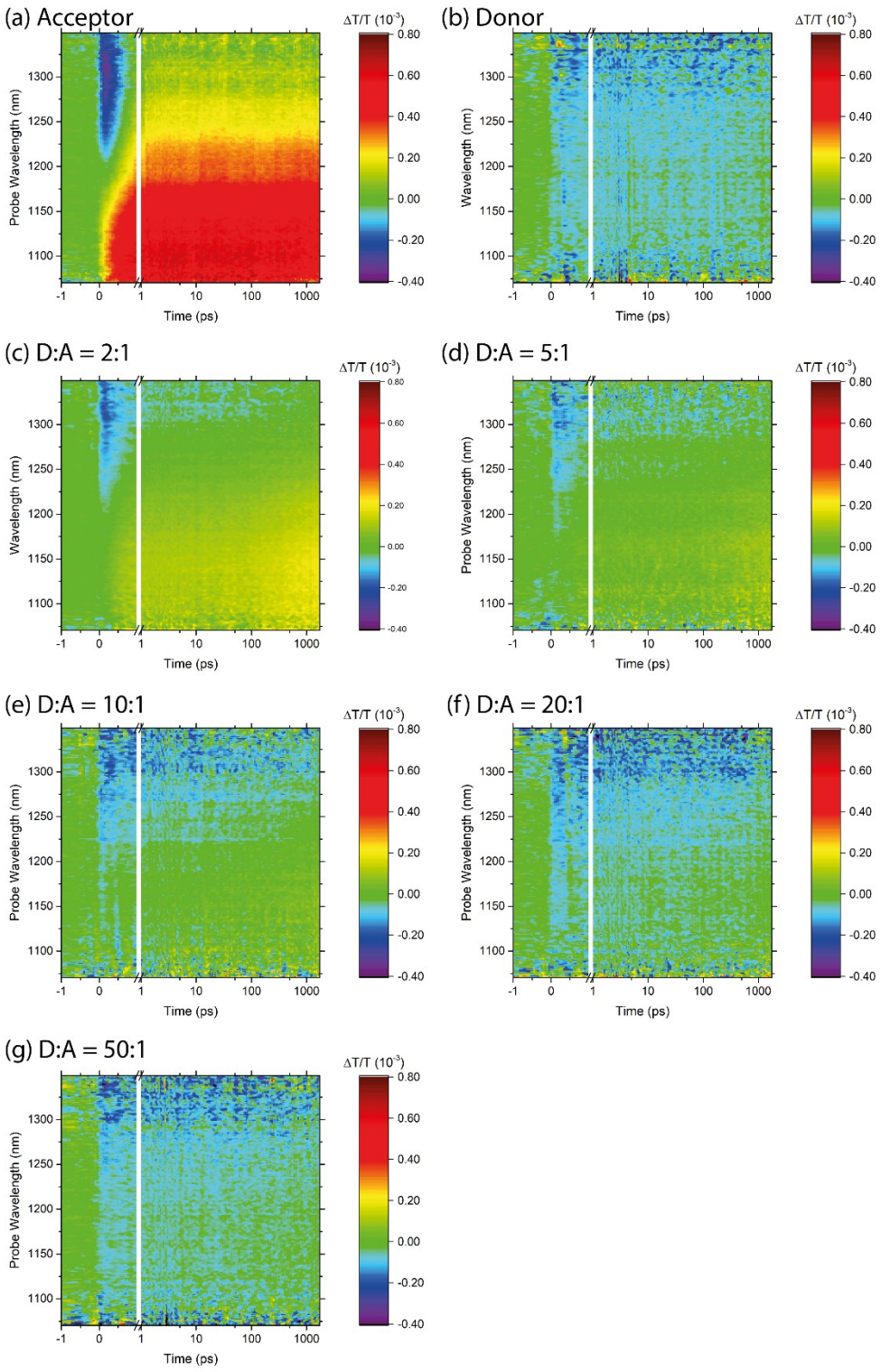
The strength of the acceptor GSB signal is an indicator of the population of excited states on the acceptor, taking into account exciton transfer to the acceptor, radiative and non-radiative decay processes. Whereas, the long lived donor PIA feature is an indication of population of excited states on the donor. The GSB of the donor is not visible as the higher energy quantum dots absorption is situated close to 700 nm, outside the range of the IR probe. Full pico-second TA maps and spectral slices for all the blend ratios is included in the SI (SI Figure 2 and 3). The short lived PIA feature present across the probe range in the acceptor is characteristic of PbS quantum dots and has previously been assigned to the presence of excited biexcitons[1].

The high temporal resolution mechanically delayed pump, ps experiment does not probe long enough after photo-excitation to observe the full dynamics of the quantum dots. By using an electronically delayed pump pulse (lower temporal resolution) we observed the transient transmission signal out to a

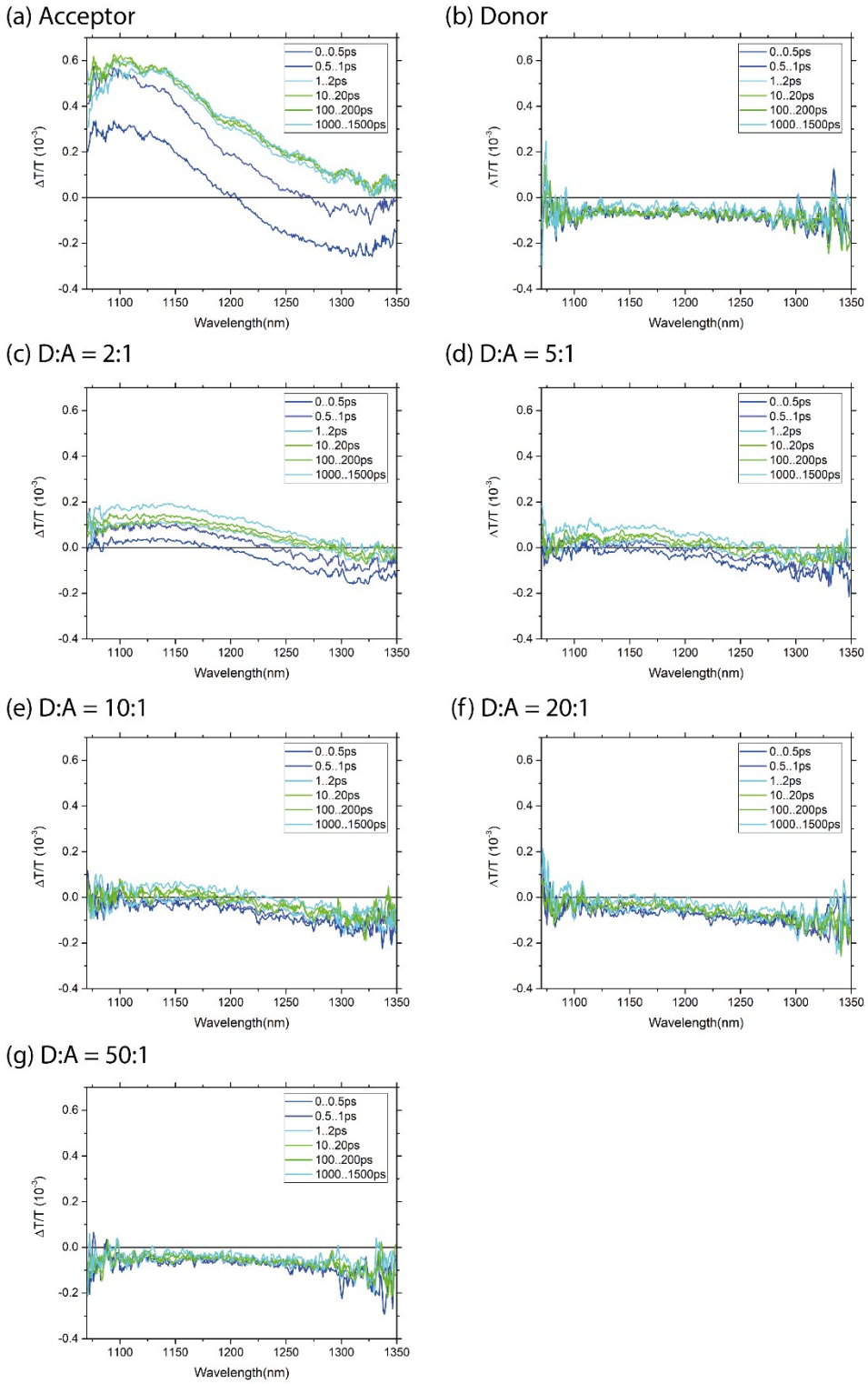
100 μs, resolving the lifetime of the acceptor and donor, and transfer between the two (SI Figure 4 and 5). In the acceptor TA data we observe a red shift of the GSB over the first 4 ns SI figure 4 (a). The acceptor sample is assumed to be mono-disperse, however this red-shifting could indicate the transfer to a subset of slightly larger dots with a lower band gap (which would have red-shifted absorption and GSB) consistent with previous reports[2] however on much faster time scales, 4 ns compared to 100s ns. The acceptor population in isolation and blends follows a bi-exponential decay which could be associated to diffusion leading to bi-exciton annihilation causing a fast (100s of ns) decay component and a longer (10s of μs) intrinsic mono-molecular decay

After photo excitation there does not appear to be any additional spectral features, required to assign all observed TA data for the blends, that might be assigned to additional excited states such as, charge transfer states or free charges. However, due to the broad spectral shape of the acceptor GSB any electro-absorption signal due to the presence of a charge transfer state appearing might be difficult to assign. An electro-absorption signal would result in slight peak shifting in the acceptor GSB, however with the current data we are unable to observe any such shift in the blend samples.

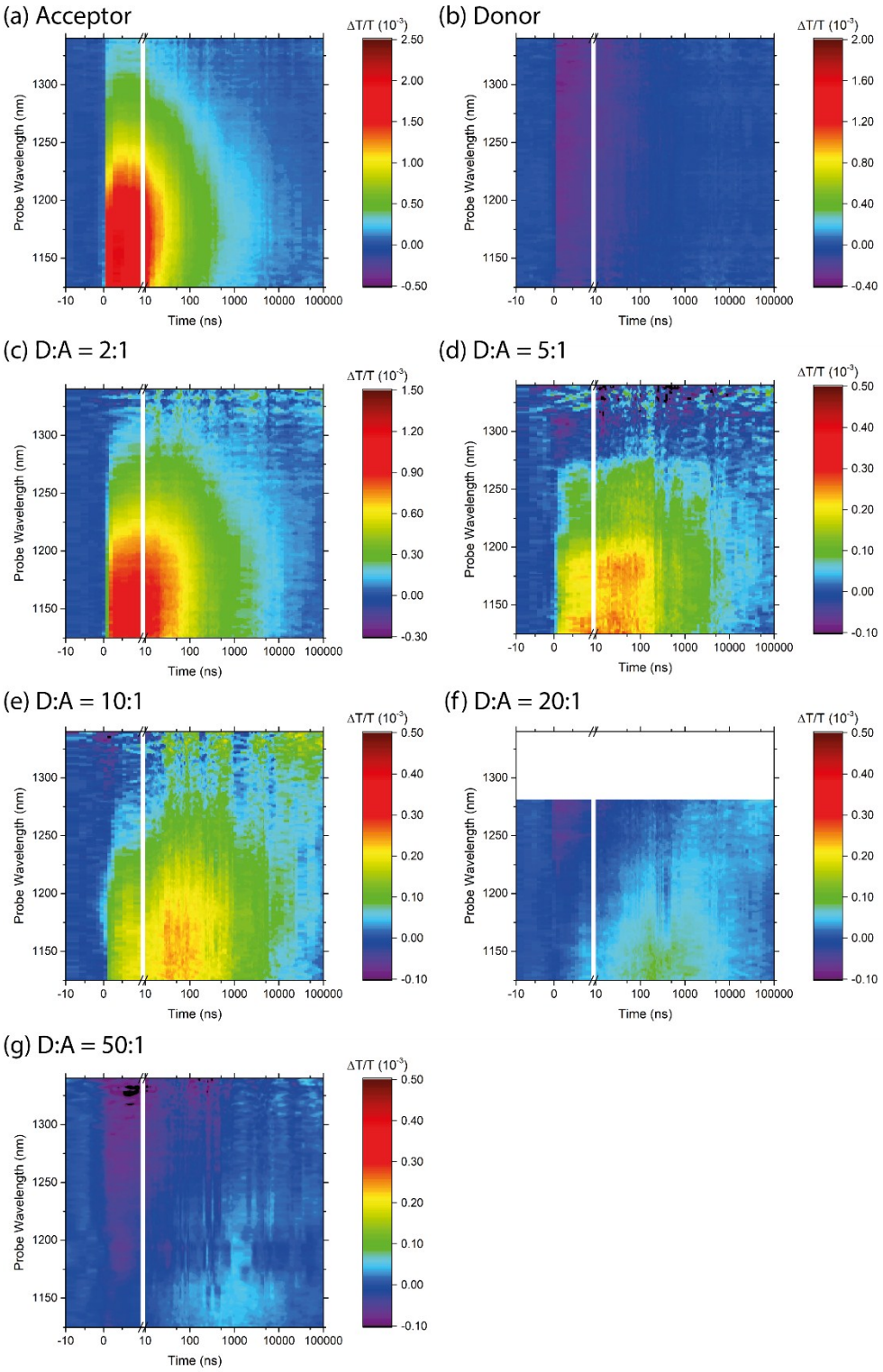
Looking at the ps transfer kinetics we find that the proportionate increase in acceptor signal is less in the higher ratio blends. This transfer rate maybe associated with the initial transfer between adjacent donor and acceptor quantum dots and not the overall system The transfer of excitons from the donor to the acceptor is not accurately modelled by a single exponential function, as the distribution of donor and acceptor sites can be quite complex.



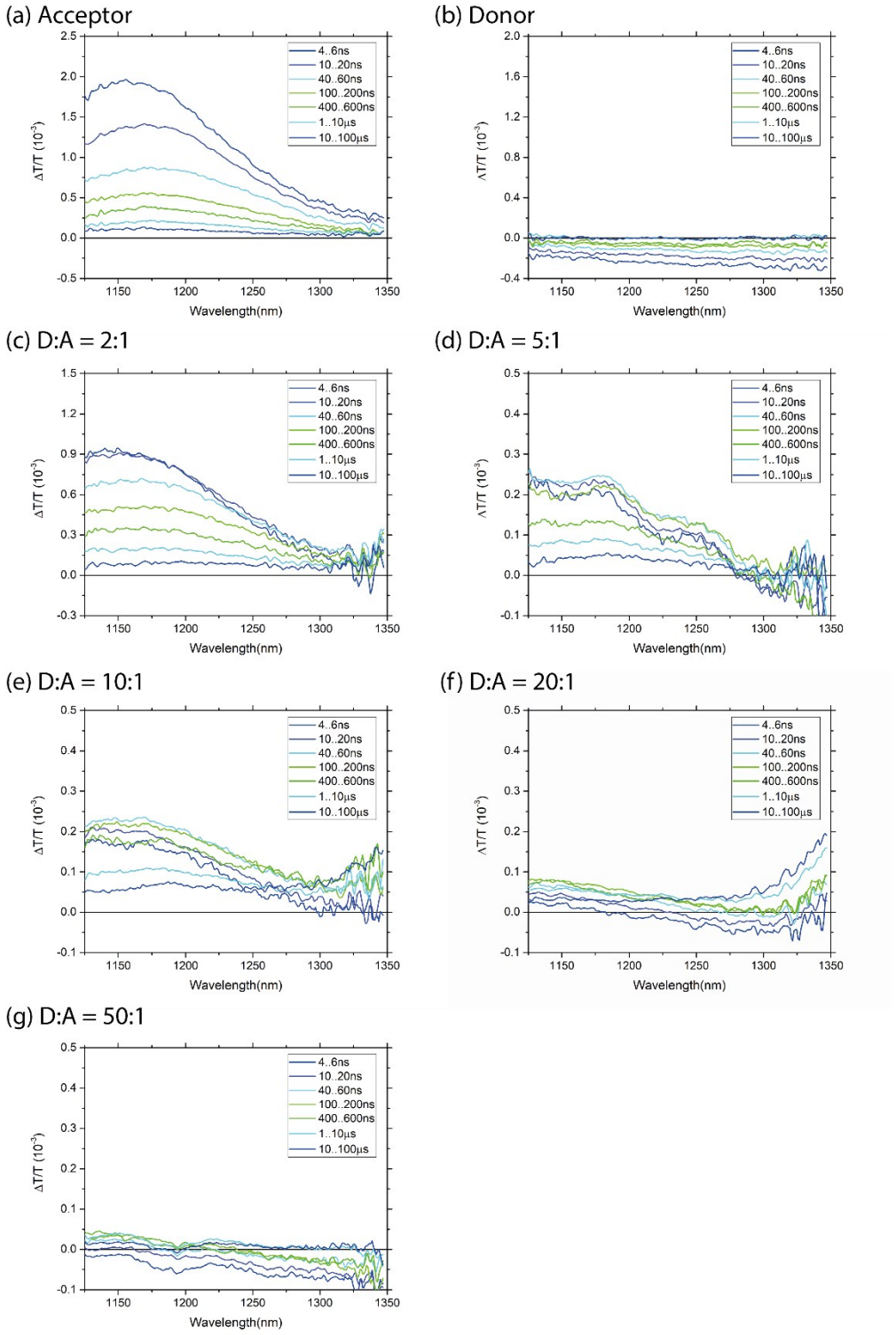
SI Figure 2: IR probe, pico-second transient transmission maps of the GDBD samples, acceptor (a), donor (b), 2:1 blend (c), 5:1 blend (d), 10:1 blend (e), 20:1 blend (f) and the 50:1 blend (g).



SI Figure 3: IR probe, pico-second transient transmission time slices of the GDBD samples, acceptor (a), donor (b), 2:1 blend (c), 5:1 blend (d), 10:1 blend (e), 20:1 blend (f) and the 50:1 blend (g).



SI FIgure 4: IR probe, nano-second transient transmission maps of the GDBD samples, acceptor (a), donor (b), 2:1 blend (c), 5:1 blend (d), 10:1 blend (e), 20:1 blend (f) and the 50:1 blend (g).



SI Figure 5: IR probe, nano-second transient transmission time slices of the GDBD samples, acceptor (a), donor (b), 2:1 blend (c), 5:1 blend (d), 10:1 blend (e), 20:1 blend (f) and the 50:1 blend (g).

Supplementary references:

1. Ellingson, R. J. *et al.* Highly efficient multiple exciton generation in colloidal PbSe and PbS quantum dots. *Nano Lett.* **5,** 865–71 (2005).
2. Clark, S. W., Harbold, J. M. & Wise, F. W. Resonant Energy Transfer in PbS Quantum Dots. *J. Phys. Chem. C* **111,** 7302–7305 (2007).