Quantum Dot to Quantum Dot Förster Resonance Energy Transfer: Engineering Materials for Visual Color Change Sensing

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Supporting Information.

Quantum Dot Characterization:

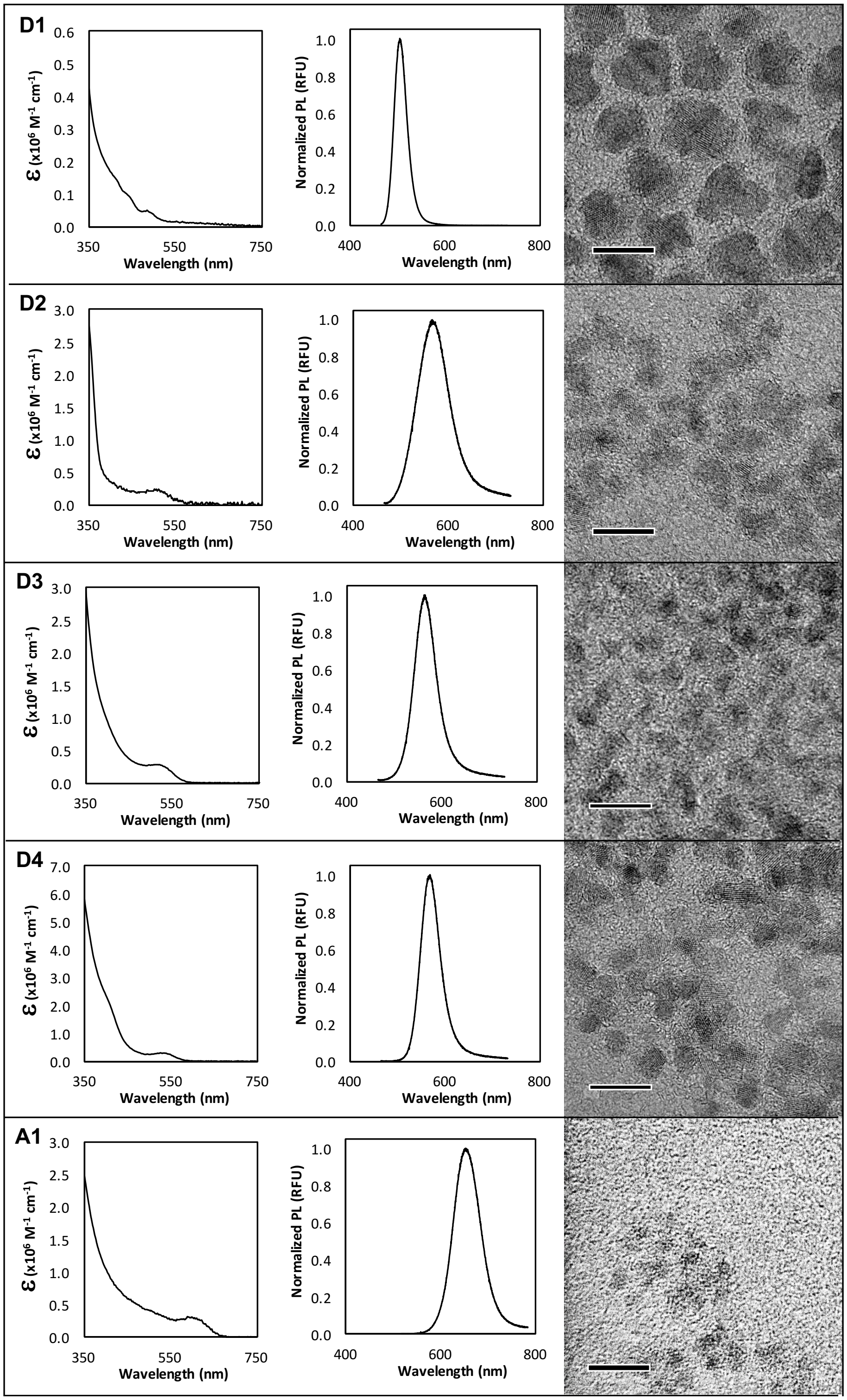


Figure S1. Optical and physical properties of the QDs used in this work. From left to right: molar extinction coefficient, photoluminescence spectra, and TEM images (scale bar = 10 nm) are shown. Each quantum dot is labeled as either a donor (D#) or acceptor (A#) in the top left corner, corresponding to row labels used in Table 1 in the main text.

PL decays for select donor-acceptor pairs:

To confirm that color change is energy transfer related, PL decay curves (lifetimes) were taken in addition to steady-state PL. 200 µL of selected FRET assays were taken directly from the plate assays described in the main text and loaded into low volume quartz cuvettes and measured in the donor fluorescence channel using a fluorescence lifetime spectrometer (LifeSpec II, Edinburg Instruments) employing a time-correlated single photon counting (TCSPC). A 405 nm pulse diode laser (EPL-205, Edinburg Instruments) was used to excite the samples at a 2 µs pulse period. Photons were counted over a 0.5 – 2 µs time range with channel widths of 1.02 ns. The collected lifetimes were fit to a tri-exponential decay (F980 Software, Edinburg Instruments):

$$I(t) = A_1\tau_1 e^{-\frac{t}{\tau_1}} + A_2\tau_2 e^{-\frac{t}{\tau_2}} + A_3\tau_3 e^{-\frac{t}{\tau_3}} \quad \text{(S1)}$$

where t represents time and A_i are coefficients that indicate the weight associated with each decay time. Average amplitude weighted lifetimes were calculated using:[1]

$$\tau_{ave} = \frac{A_1\tau_1 + A_2\tau_2 + A_3\tau_3}{A_1 + A_2 + A_3} \quad \text{(S2)}$$

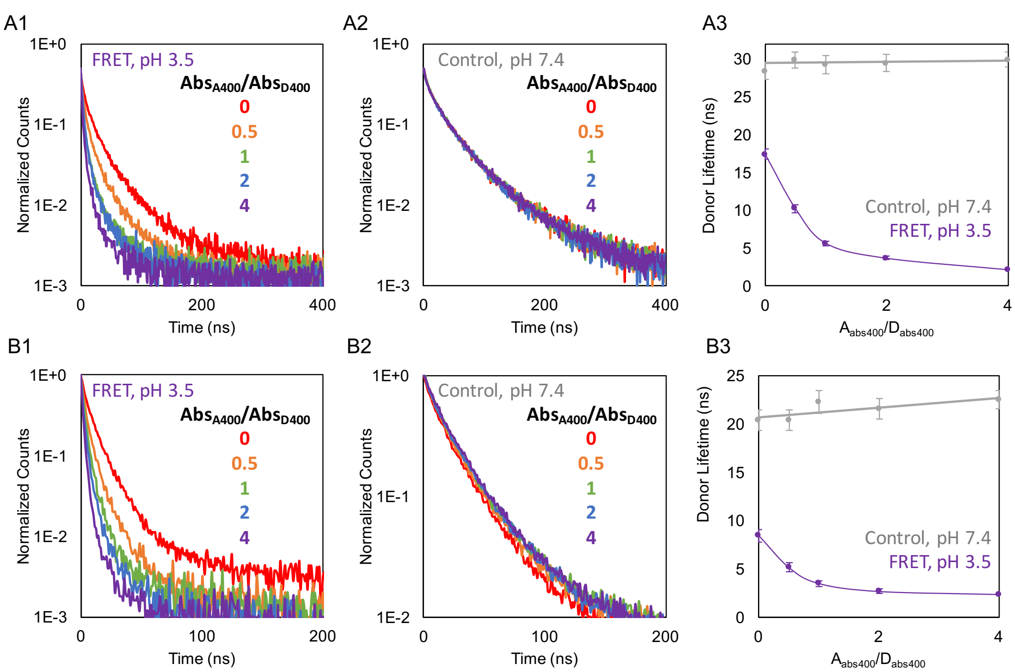
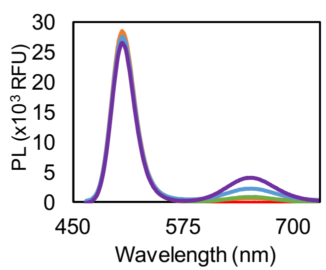
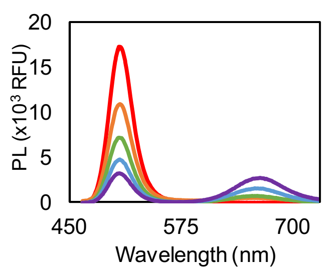
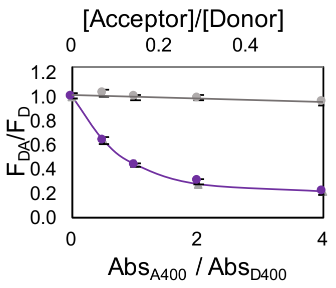
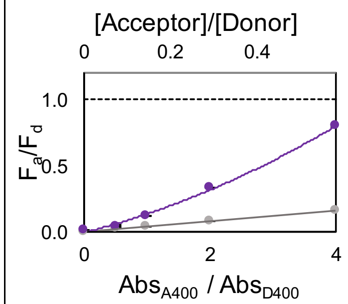
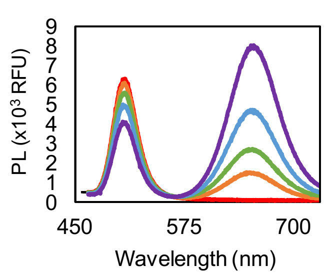
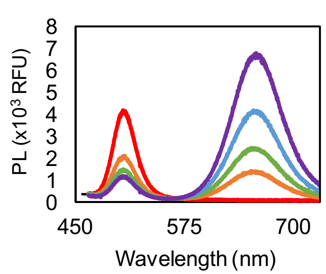
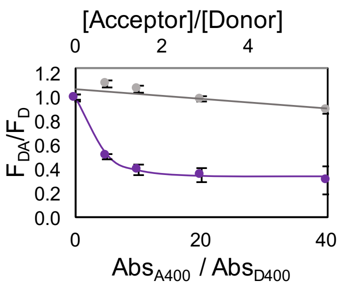
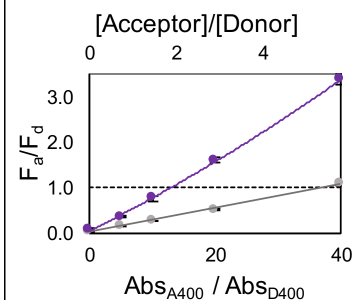
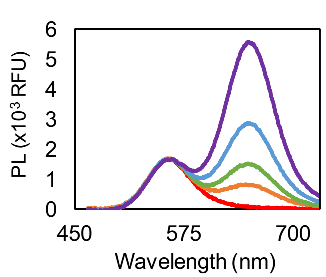
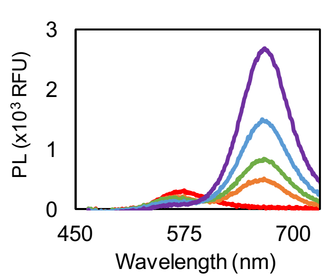
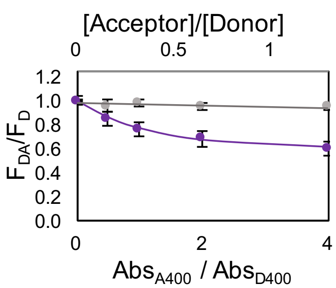
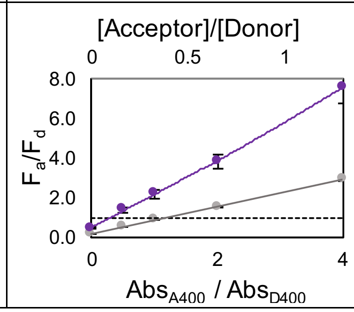
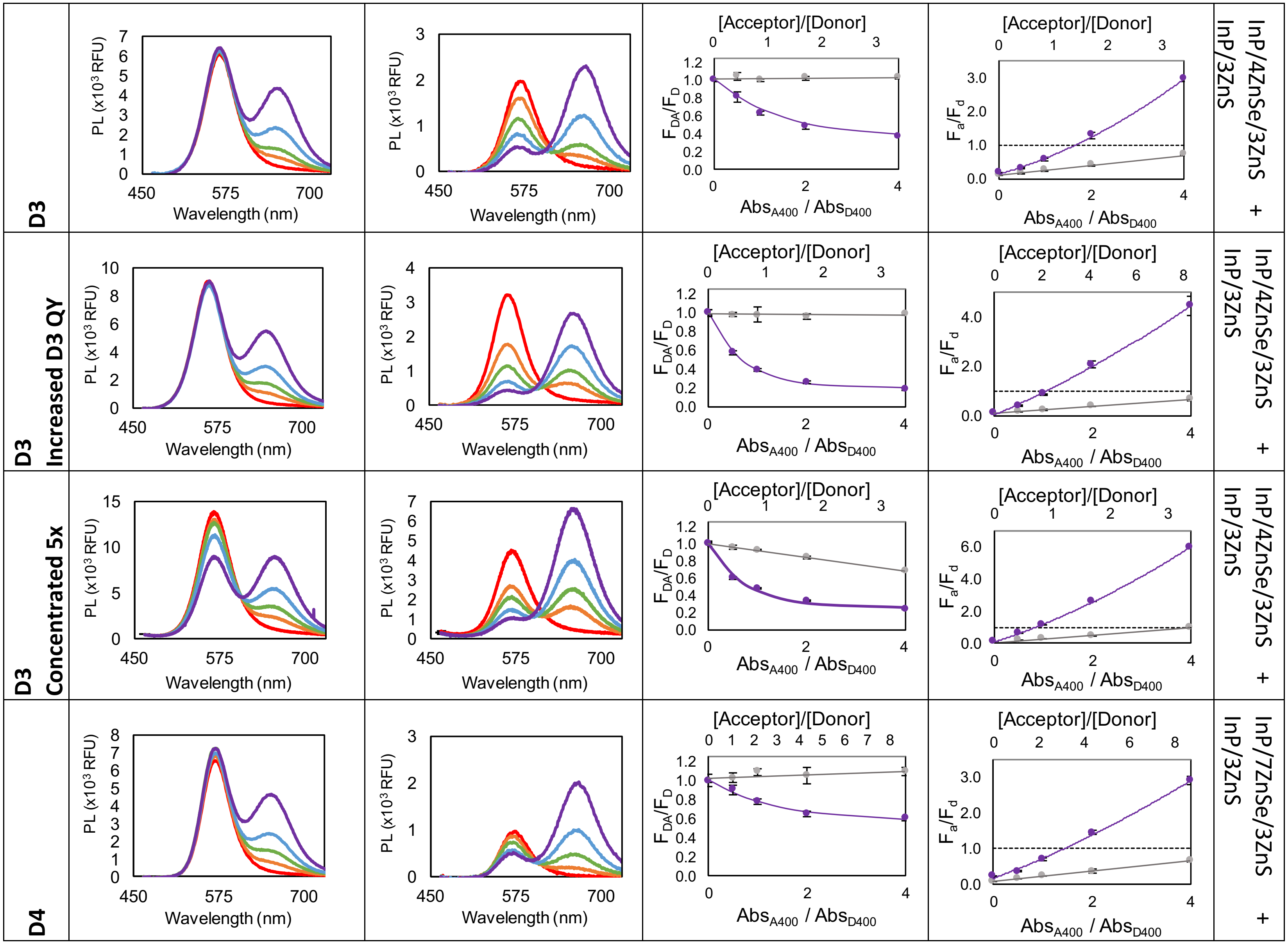


Figure S2. PL Decay curves for an InP/4ZnSe/3ZnS QD (D3) and a $CdxZn_{1-x}SeyS_{1-y}$ QD (D1) acting as donors to an InP/3ZnS acceptor (A1). Data were collected for each acceptor/donor ratio at pH 3.5 (column 1) and pH 7.4 (column 2) for the FRET and control systems respectively. The decay curves were fit to Eqn 1 and average lifetimes were calculated using Eqn 2. Donor lifetimes were plotted in column 3 and decreases as acceptor to donor ratio increases at pH 3.5 indicating energy transfer. Donor lifetime significantly decreases after aggregation, even with no acceptors present, indicating that donor-donor homoFRET is occurring.

Spectral Data of Aggregation Assays:

Table S1. PL Spectra of all assays described in the main paper. Column 1 indicates the donor paired with the A1 acceptor, column 2 shows PL at pH 7.4, column 3 shows PL at pH 3.5, and column 4 and 5 depict the donor quenching and F_a/F_d, respectively, at each acceptor/donor ratio in the unaggregated (grey) and aggregated (purple) states.

	pH 7.4	**pH 3.5**	**Donor Quenching**	**F_a/F_d**	
D1					$Cd_xZn_{1-x}Se_yS_{1-y}$ + InP/3ZnS
D1 Donor diluted 10x					$Cd_xZn_{1-x}Se_yS_{1-y}$ + InP/3Zn
D2					InP/7ZnS + InP/3ZnS



D3
D3 Increased D3 QY
D3 Concentrated 5x
D4
PL (x10³ RFU)
Wavelength (nm)
450
575
700
[Acceptor]/[Donor]
F_DA/F_D
Abs_A400 / Abs_D400
F_a/F_d
InP/4ZnSe/3ZnS + InP/3ZnS
InP/4ZnSe/3ZnS + InP/3ZnS
InP/4ZnSe/3ZnS + InP/3ZnS
InP/7ZnSe/3ZnS + InP/3ZnS

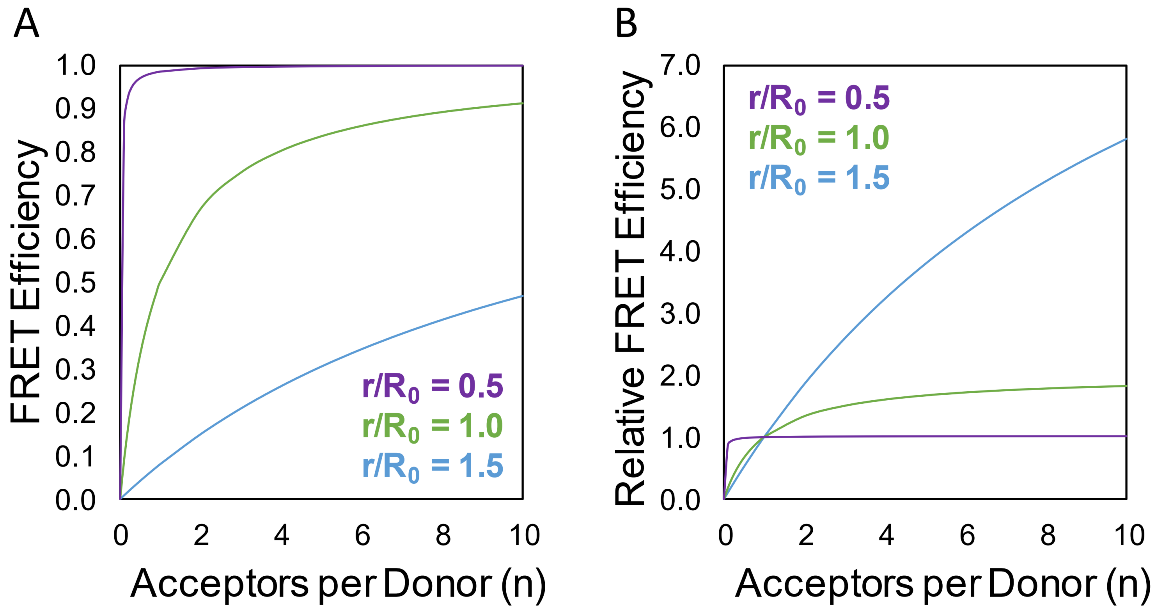


Figure S3. (A) FRET efficiency as a function of the number of acceptors per donor for different donor-acceptor distances. If using the same number of acceptors per donor, the more compact FRET pair always has the greatest FRET efficiency. In systems with larger donor-acceptor distances, however, low FRET efficiency can be alleviated by increasing acceptor to donor ratio. In panel **(B)** each of the lines are normalized to their FRET efficiencies where n = 1 in order to more easily visualize the effect of increased acceptor to donor ratio on each FRET system. Lines are calculated by plugging into Eqn. 1 from the main text.