Supporting Information

Ultrafast separation of multiexcitons within core/shell quantum dot hybrid systems

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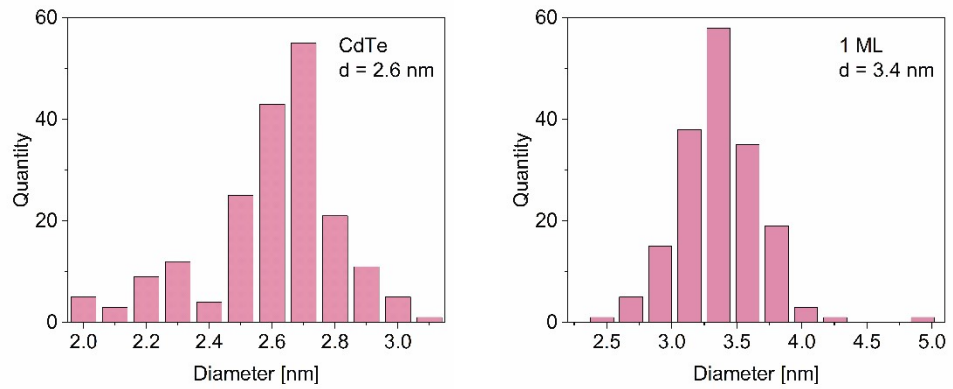

Fig. S1: Size determination of CdTe and 1 ML.

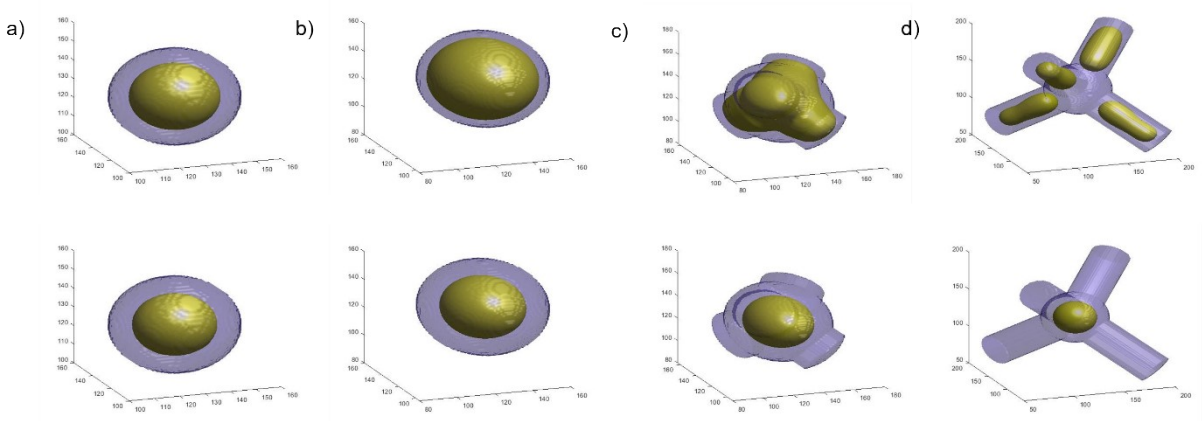

Fig. S2: Calculated electron (top) and hole (bottom) ground state wavefunction amplitudes of a) CdTe, b) 1 ML, c) 2 ML and d) 3 ML. The QD surface is blue and the wavefunctions yellow. The wavefunctions were calculated with the split operator method with following stats for the QDs (r = radius, L = length) – CdTe: r_{core} = 1.3 nm; 1 ML: r_{core} = 1.3 nm, r_{shell} = 0.4 nm; 2 ML: r_{core}=1.3 nm, r_{shell} = 0.4 nm, r_{arms} = 1.0 nm, L_{arms} = 0.4 nm; 3 ML: r_{core}=1.3 nm, r_{shell} = 0.4 nm, r_{arms} = 1.0 nm, L_{arms} = 3.7 nm.

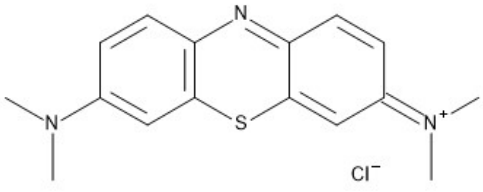

Fig. S3: Molecular structure of methylene blue.

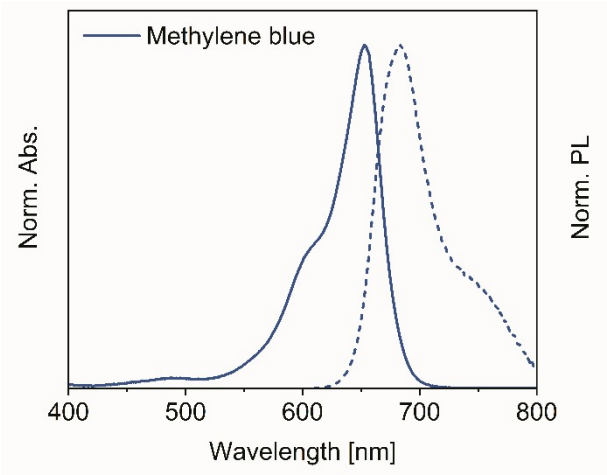

Fig. S4: Absorbance (solid line) and photoluminescence spectra (dotted line) of methylene blue.

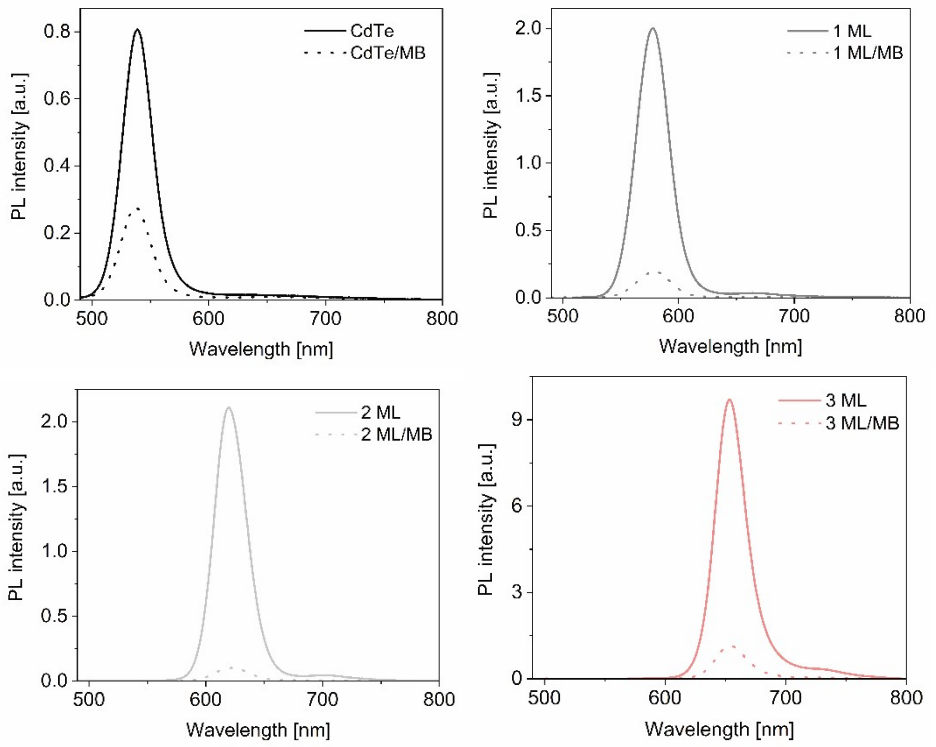

Fig. S5: Photoluminescence spectra of CdTe, 1 ML, 2 ML, 3 ML and the corresponding QD/MB complexes.

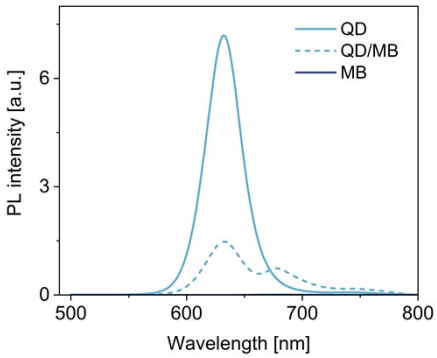

Fig. S6: Photoluminescence spectra of CdSe/ZnS, CdSe/ZnS/MB and pure MB. The absence of MB PL in Fig. S5 could also result from the attachment to the QD surface. Therefore, a control experiment has been conducted with another QD (CdSe/ZnS). The complex shows a quenched donor emission, as well as an enhanced MB PL. The direct excitation of MB shows no PL at all, indicating that the MB PL is due to FRET. Consequently, we can assume that binding to the QD surface doesn't affect the MB PL. Therefore, for CdTe and CdTe/CdSe the QD PL quenching can be assigned to an ET.

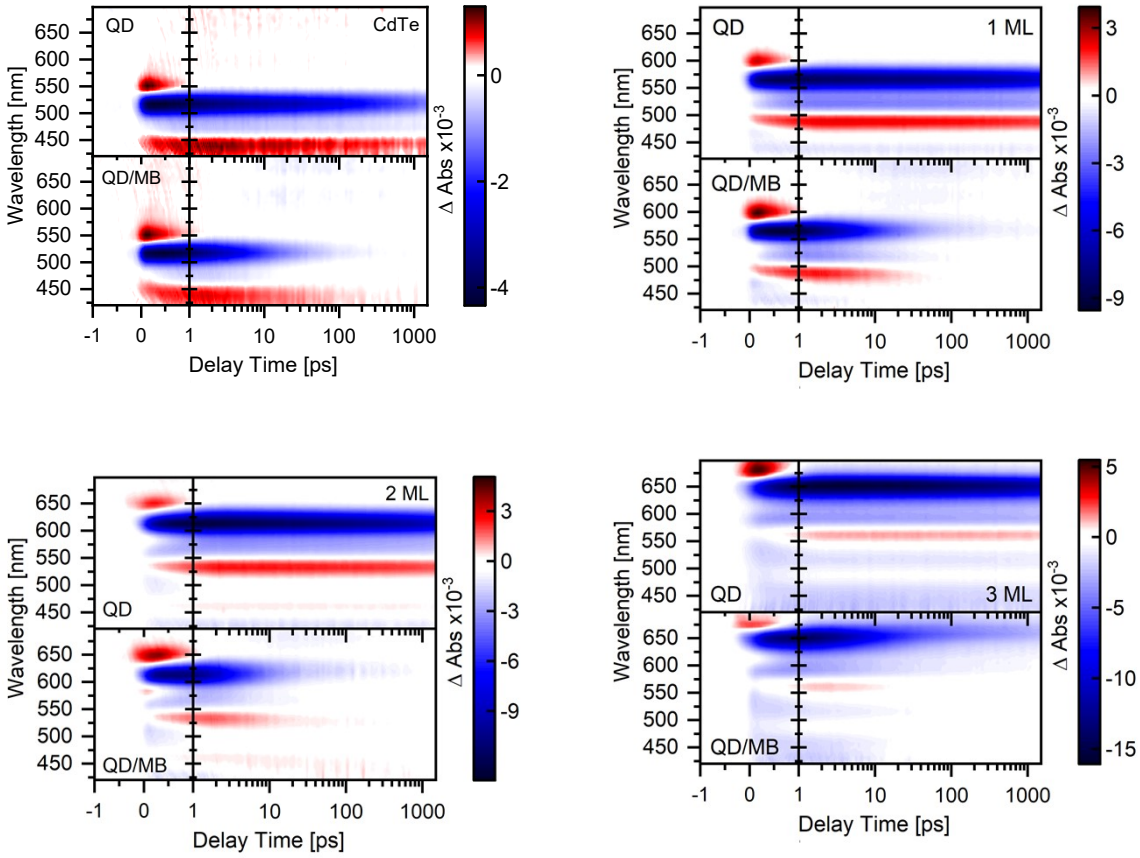

Fig. S7: TA spectra of QD (upper part of figures) and QD/MB complexes (lower part of figures): 1 ML left), 2 ML (top right), 3 ML (bottom).

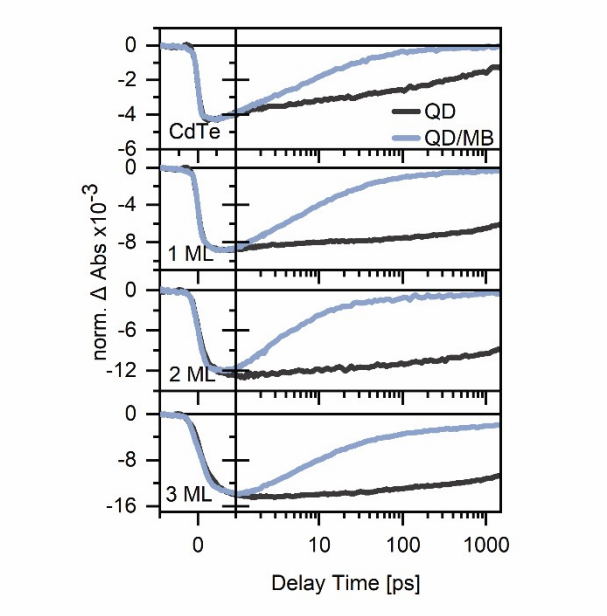

Fig. S8: Single transients in the QD-bleach region for QDs and the respective complexes. Selected probe wavelengths: CdTe: 515 nm, 1 ML: 560 nm, 2 ML: 607 nm, 3 ML: 640 nm.

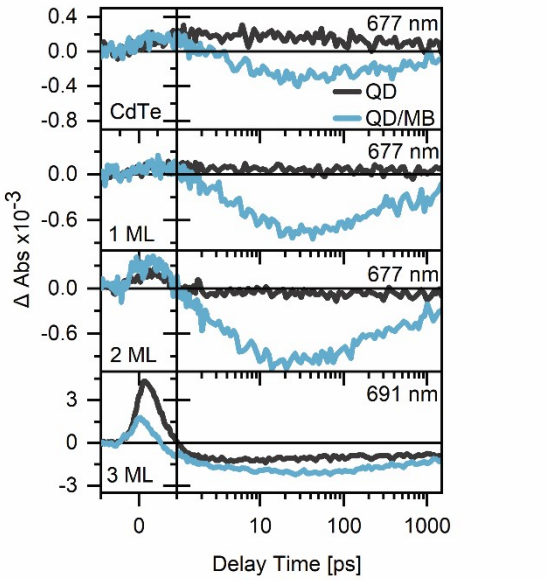

Fig. S9: Transient absorption traces within the MB-bleach region for the QDs and the respective complexes. For the 3 ML complex, the overlap with the QD bleach is very strong at 677 nm, and thus the transient at 691 nm is depicted (cf. Fig. S8).

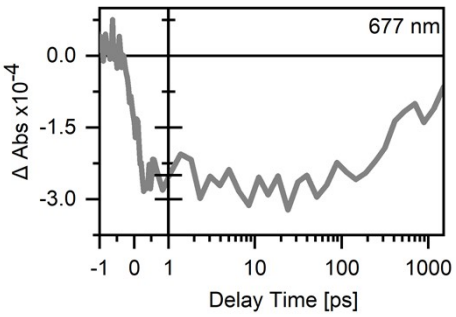

Fig. S10: Transient absorption traces at 677 nm of pure methylene blue.

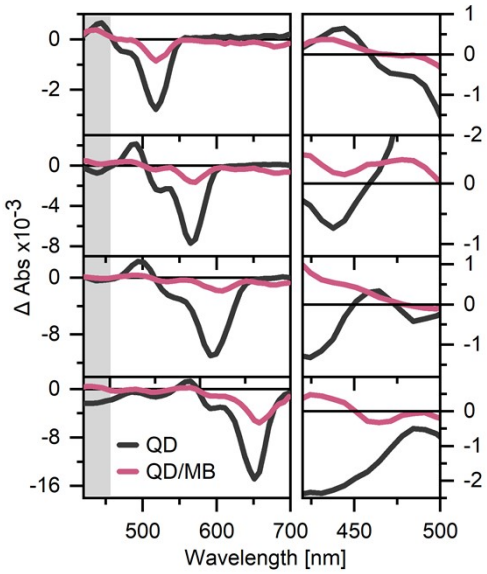

Fig. S11: Spectra at a fixed delay time of 50 ps for CdTe, 1 ML, 2 ML, 3 ML and the respective complexes (from top to bottom).

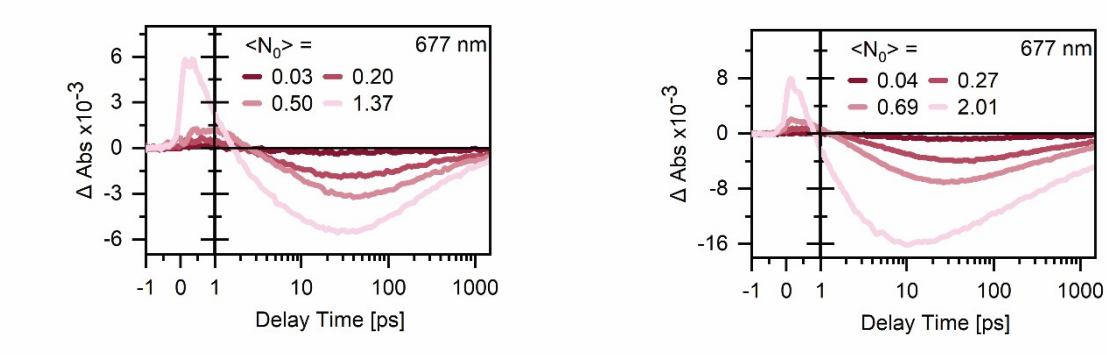

Fig. S12: Excitation power dependent measurements of CdTe/MB (right) and 1 ML/MB (left).

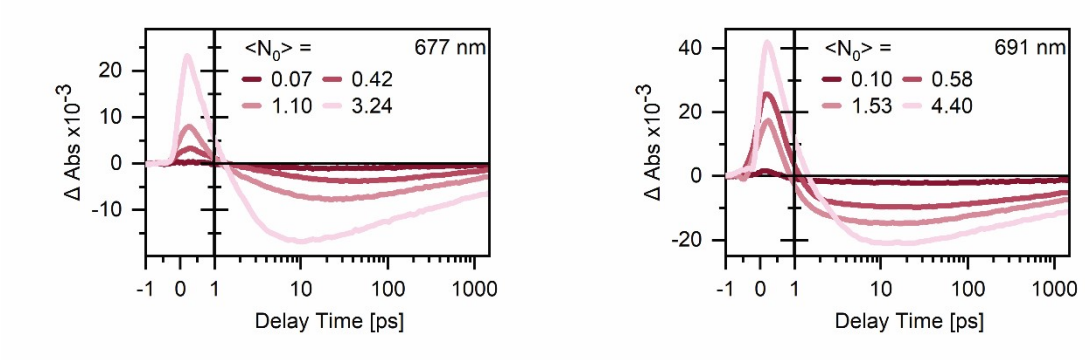

Fig. S13: Excitation power dependent measurements of 2 ML/MB (right) and 3 ML/MB (left).