Table S1 Percentage contribution of atoms to the energy levels of passivated QDs in vaccum.

model QDs	molecular orbitals	composition in terms of most important atomic orbitals
QD1	HOMO-5	+ 12.2% 6PX(S10)3 + 8.6% 6PY(S7)3 - 7.2% 6PZ(S11)3 + 5.5% 7PX(S10)3 - 5.2% 6PX(S7)3 - 4.6% 6PY(S6)3
	HOMO-4	+ 8.8% 6PX(S9)3 - 6.2% 6PY(S3)3 - 6.1% 6PZ(S6)3 + 5.9% 6PX(S4)3 + 4.6% 6PZ(S3)3 + 4.6% 6PY(S6)3
	HOMO-3	+ 15.0% 6PZ(S8)3 + 9.3% 6PY(S8)3 + 6.4% 7PZ(S8)3 + 5.7% 6PZ(S3)3 - 5.1% 6PZ(S2)3 + 4.6% 6PZ(S1)4
	HOMO-2	+ 10.0% 6PY(S1)4 - 6.8% 6PX(S1)4 - 5.0% 6PX(S10)3 + 4.5% 6PY(S3)3 + 4.2% 7PY(S1)4 - 3.9% 6PZ(S1)4
	HOMO-1	+ 12.8% 6PX(S13)3 - 7.6% 6PZ(S2)3 + 7.2% 6PX(S1)4 + 6.0% 7PX(S13)3 - 4.8% 6PZ(S1)4 + 4.8% 6PY(S13)3
	HOMO	+ 13.4% 6PZ(S2)3 + 8.2% 6PY(S2)3 + 7.7% 6PX(S13)3 + 6.1% 7PZ(S2)3 + 6.0% 6PY(S1)4 + 4.2% 6PZ(S1)4
	LUMO	+ 6.5% 6S(Cd24)3 + 6.4% 6S(Cd29)3 + 6.3% 6S(Cd27)3 + 6.2% 6S(Cd15)3 + 4.1% 6S(Cd21)3 + 4.0% 6S(Cd16)3
	LUMO+1	+ 13.5% 6S(Cd27)3 - 13.2% 6S(Cd15)3 - 11.1% 6S(Cd29)3 + 9.3% 6S(Cd24)3 + 4.0% 6S(Cd17)3 + 3.2% 5S(Cd27)3
	LUMO+2	+ 12.3% 6S(Cd16)3 - 8.4% 6S(Cd24)3 + 8.1% 6S(Cd21)3 - 5.4% 6S(Cd29)3 + 4.0% 6S(Cd28)3 - 2.6% 6S(Cd20)3
	LUMO+3	+ 17.1% 6S(Cd21)3 - 12.1% 6S(Cd16)3 - 5.7% 6S(Cd25)3 + 5.0% 6S(Cd23)3 - 4.8% 6S(Cd19)3 + 4.0% 5S(Cd21)3
	LUMO+4	+ 11.6% 6S(Cd29)3 - 8.2% 6S(Cd24)3 + 7.1% 6S(Cd27)3 - 6.5% 6S(Cd15)3 - 3.5% 6S(Cd16)3 + 3.3% 5S(Cd29)3
	LUMO+5	+ 9.9% 6S(Cd15)3 - 9.5% 6S(Cd14)3 + 8.8% 6S(Cd27)3 + 4.3% 11PZ(Cd27)3 - 3.8% 5S(Cd14)3 - 3.7% 6S(Cd24)3
QD2	HOMO-5	+ 5.9% 6PZ(S50)3 + 5.6% 6PX(**S38**)4 + 3.8% 6PZ(S47)3 - 3.6% 6PX(S32)3 - 3.2% 6PX(**S31**)4 + 3.1% 6PZ(S45)3
	HOMO-4	+ 5.7% 6PX(**S52**)4 - 5.3% 6PY(S45)3 - 4.7% 6PY(S47)3 + 3.6% 6PZ(S54)3 + 3.0% 6PX(S54)3 - 2.9% 6PX(**S31**)4
	HOMO-3	+ 5.2% 6PZ(S43)3 + 5.0% 6PY(S32)3 - 3.8% 6PX(**S38**)4 - 3.8% 6PX(**S52**)4 - 3.7% 6PX(**S31**)4 - 2.9% 6PY(S54)3
	HOMO-2	+ 5.1% 6PX(**S38**)4 + 5.1% 6PX(**S52**)4 + 5.1% 6PX(**S31**)4 + 3.0% 6PX(S30)4 + 3.0% 6PX(S55)4 + 2.9% 6PX(S40)4
	HOMO-1	+ 14.9% 6PY(S32)3 - 7.6% 6PZ(S43)3 + 6.9% 7PY(S32)3 - 5.8% 6PZ(**S57**)4 + 4.6% 6PY(S49)4 - 3.5% 7PZ(S43)3
	HOMO	+ 9.9% 6PZ(S54)3 + 7.6% 6PZ(S43)3 + 6.3% 6PY(S54)3 + 4.7% 6PY(**S39**)4 + 4.7% 6PZ(**S49**)4 + 4.6% 7PZ(S54)3
	LUMO	+ 3.0% 6S(Cd17)3 + 3.0% 6S(Cd29)3 + 3.0% 6S(Cd12)3 + 2.8% 6S(Cd25)3 + 2.8% 6S(Cd58)3 + 2.8% 6S(Cd15)3
	LUMO+1	+ 12.1% 6S(Cd25)3 + 7.5% 6S(Cd17)3 + 5.0% 6S(Cd13)3 + 4.9% 5S(Cd25)3 - 3.4% 6S(Cd58)3 - 3.1% 6S(Cd29)3
	LUMO+2	+ 9.5% 6S(Cd15)3 - 8.7% 6S(Cd58)3 + 6.7% 6S(Cd12)3 - 4.7% 6S(Cd19)3 - 4.6% 6S(Cd29)3 + 4.0% 5S(Cd15)3
	LUMO+3	+ 8.0% 6S(Cd59)3 + 7.9% 6S(Cd28)3 + 7.9% 6S(Cd21)3 - 2.3% 6S(Cd14)3 - 2.3% 6S(Cd23)3 - 2.3% 6S(Cd9)
	LUMO+4	+ 7.7% 6S(Cd59)3 + 7.7% 6S(Cd28)3 + 7.7% 6S(Cd21)3 - 3.6% 6S(Cd58)3 - 3.6% 6S(Cd25)4 - 3.6% 6S(Cd15)3
	LUMO+5	- 6.5% 6S(Cd13)3 + 5.9% 6S(Cd17)3 + 5.1% 5S(Cd9) + 4.9% 6S(Cd9) - 3.8% 6S(Cd29)3 + 3.6% 6S(Cd19)3
QD3	HOMO-5	+ 7.5% 6PY(S7)3 - 6.1% 6PY(S27)3 - 5.3% 6PX(S17)3 + 4.0% 6PY(S2)3 + 3.5% 7PY(S7)3 + 3.5% 6PZ(S7)3
	HOMO-4	+ 7.2% 6PZ(S25)3 - 6.5% 6PY(S25)3 + 5.5% 6PZ(S6)3 - 4.8% 6PX(S6)3 - 4.5% 6PX(S16)3 + 3.0% 7PZ(S25)3
	HOMO-3	+ 9.9% 6PY(S2)3 + 6.4% 6PX(S24)3 + 6.1% 6PZ(S24)3 + 5.7% 6PY(S24)3 + 4.2% 7PY(S2)3 + 3.8% 6PZ(S2)3
	HOMO-2	+ 14.3% 6PZ(S32)3 - 12.1% 6PX(S30)3 + 9.0% 6PZ(S30) + 7.0% 7PZ(S32)3 - 5.8% 7PX(S30)3 + 5.6% 6PZ(S12)4
	HOMO-1	+ 12.1% 6PY(S273) - 8.4% 6PX(S33)3 - 6.5% 6PZ(S33)3 - 6.3% 6PZ(S32)3 + 6.2% 7PY(S27)3 - 4.1% 7PX(S33)3
	HOMO	+ 17.3% 6PY(S27)3 + 9.6% 6PZ(S33)3 + 8.8% 6PX(S33)3 + 8.6% 7PY(S27)3 + 5.1% 6PY(S33)3 + 4.7% 7PZ(S33)3
	LUMO	+ 5.5% 6S(Cd53)3 + 3.3% 6S(Cd59)3 + 2.6% 6S(Cd69)3 + 2.6% 6S(Cd43)3 + 2.1% 6S(Cd66)4 + 2.0% 6S(Cd49)3
	LUMO+1	+ 11.5% 6S(Cd53)3 - 6.6% 6S(Cd693) + 5.0% 6S(Cd59)3 + 4.0% 6S(Cd78)3 + 3.8% 5S(Cd53)3 - 3.1% 6S(Cd48)3
	LUMO+2	+ 9.2% 6S(Cd63)3 + 8.6% 6S(Cd49)3 + 5.6% 6S(Cd72)3 + 5.3% 6S(Cd77)3 + 3.1% 5S(Cd63)3 + 2.8% 5S(Cd49)3
	LUMO+3	- 7.5% 6S(Cd65)3 + 7.4% 6S(Cd69)3 - 6.9% 6S(Cd52)3 - 4.8% 6S(Cd40)3 + 4.2% 6S(Cd62)3 + 3.7% 6S(Cd36)3
	LUMO+4	+ 10.9% 6S(Cd53)3 - 7.7% 6S(Cd59)3 + 5.3% 6S(Cd69)3 + 5.2% 6S(Cd70)3 - 5.2% 6S(Cd67)3 + 2.8% 11PZ(Cd53)3
	LUMO+5	+ 18.7% 6S(Cd78)3 + 7.2% 5S(Cd78)3 - 5.6% 6S(Cd43)3 - 4.9% 6S(Cd70)3 + 3.4% 6S(Cd69)3 + 3.1% 6S(Cd65)3

Bold letters are used for the surface 4-coordinated S-atoms near the 6 acetates in the case of QD2.

Table S2 Optical data for theoretical absorption spectra with diffuse basis 6-311++G(d,p) for dye-QD1.

system	excited states (@S_0)	excitation energy E(eV)	wavelength (nm)	oscillator strength	eigenvalue (NTO)
vacuum	$S_0 \rightarrow S_3$	3.21	386.66	0.2387	0.9032
	$S_0 \rightarrow S_4$	3.23	384.02	0.2362	0.8755
	$S_0 \rightarrow S_5$	3.27	379.01	0.1012	0.9444
	$S_0 \rightarrow S_6$	3.30	376.26	0.3926	0.8360
chloroform	$S_0 \rightarrow S_1$	3.06	405.13	0.3914	0.9898
	$S_0 \rightarrow S_2$	3.11	398.76	0.1827	0.9596
	$S_0 \rightarrow S_3$	3.16	392.79	0.3297	0.9465
	$S_0 \rightarrow S_4$	3.17	391.37	0.3994	0.7675
	$S_0 \rightarrow S_5$	3.18	389.54	0.1697	0.9826
toluene	$S_0 \rightarrow S_2$	3.12	397.30	0.2301	0.9035
	$S_0 \rightarrow S_3$	3.17	394.04	0.6532	0.7386
	$S_0 \rightarrow S_4$	3.16	391.79	0.2728	0.6195
	$S_0 \rightarrow S_5$	3.20	388.80	0.2892	0.9612

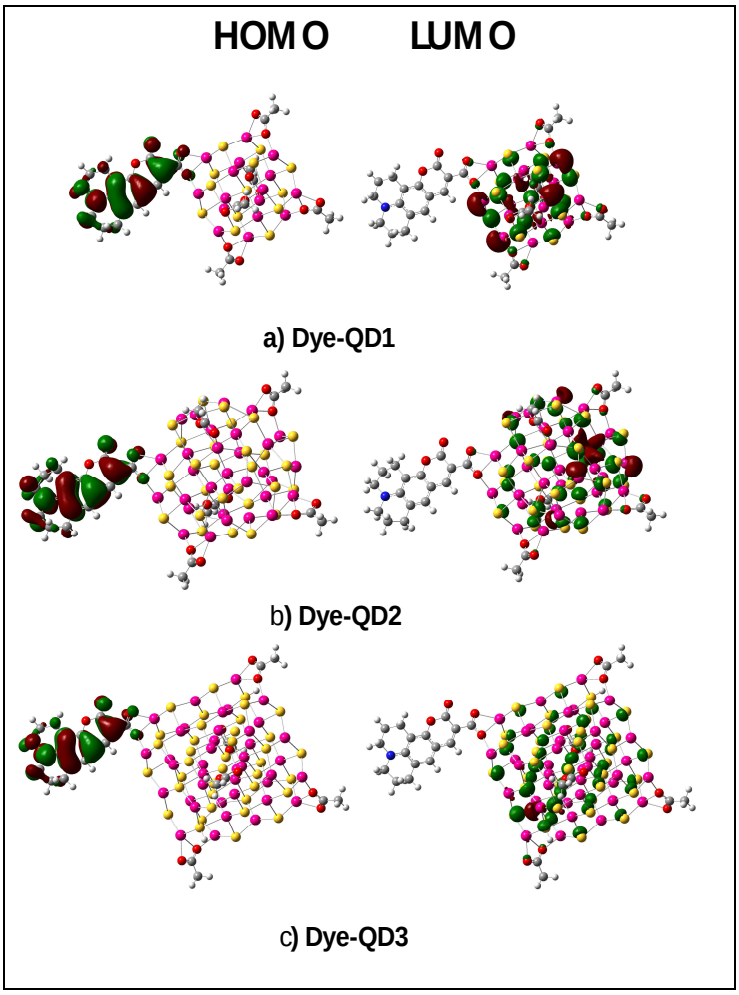

Fig. S1 Molecular orbitals in dye-sensitized QDs in vacuum.

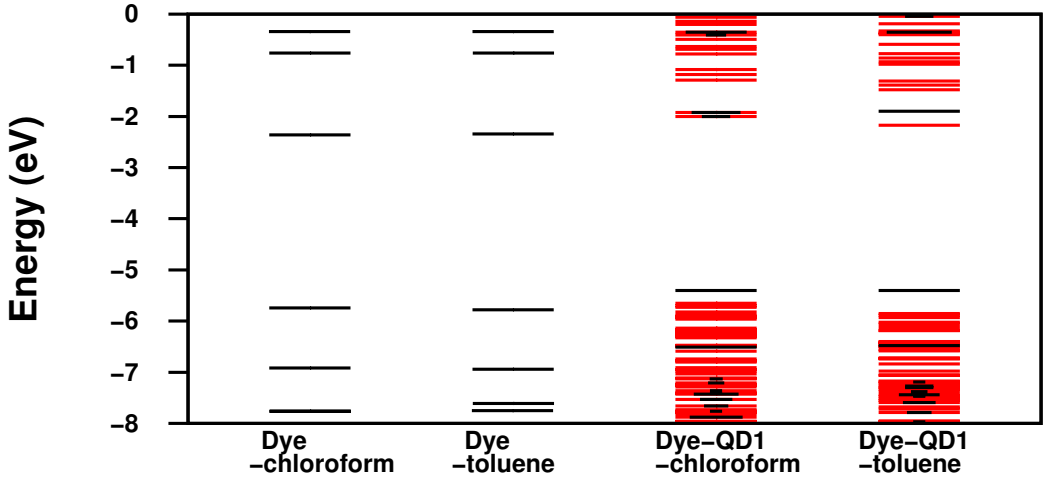

Fig. S2 Energy levels for the model dye sensitized QDs in solvents.The black lines in the dye-QD system refer to the levels arising from the dye and their width refers to the percentage contribution of the dye.

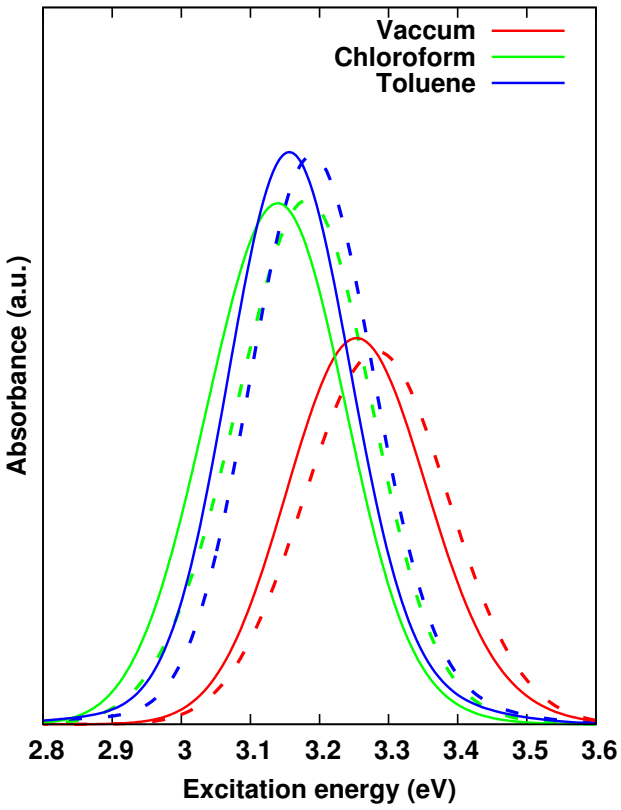

Fig. S3 Optical absorption spectra showing the comparative results for diffuse basis set(6-311++G(d,p)) for dye-QD1. Dotted line refers to the spectra without diffuse basis set.

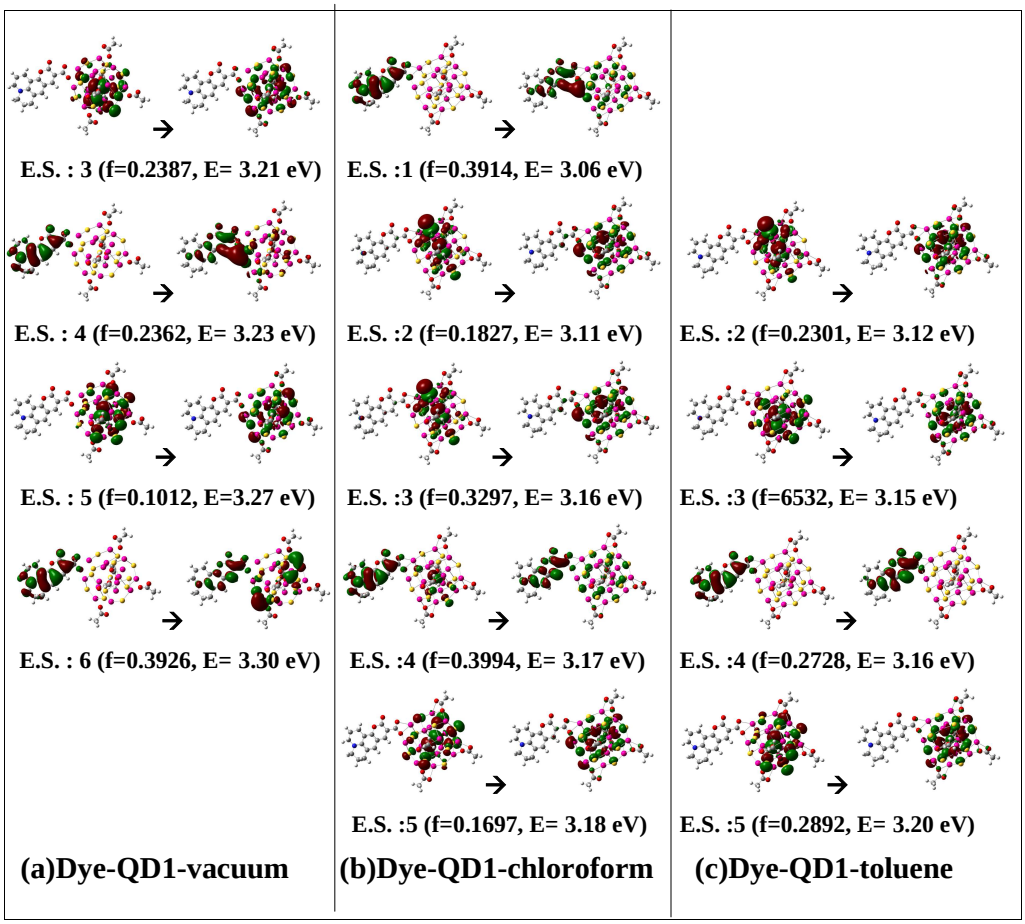

Fig. S4 The dominant natural transition orbital pairs for the excited states of absorption with diffuse basis set(6-311++G(d,p)).