Supporting Information

Electronic Band Structure and Ultrafast Carrier Dynamics of Two Dimensional (2D) Semiconductor Nanoplatelets (NPLs) in Presence of Electron Acceptor for Optoelectronic Applications

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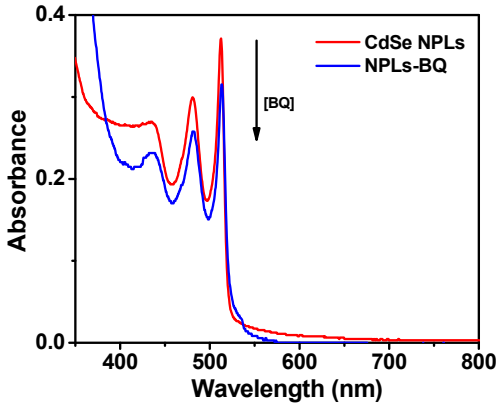


Figure S1. Steady-state absorption spectra of CdSe NPLs with BQ addition (416.25 μM).

Table S1. Time-resolved PL decay parameters of the pure CdSe NPLs and corresponding hybrid.

Systems	$\tau_1^x (a_1)^y$ **(ns)**	$\tau_2^x (a_2)^y$ **(ns)**	$\tau_3^x (a_3)^y$ **(ns)**	$\tau_{average}$ **(ns)**
CdSe NPLs	0.36	2.8	16.2	0.95±0.1
	(0.87)	(0.11)	(0.02)	
CdSe NPLs-BQ	0.2	1.45	7.92	0.34±0.1
	(0.94)	(0.05)	(0.01)	

[x] ± 4% and [y]± 5%

Computational details:

First-principles calculations have been performed within density functional theory (DFT), as implemented in the Vienna *Ab Initio* simulation package (VASP)[1-4] code, have been performed. The projected augmented wave (PAW)[5] pseudo-potentials have been used to describe the ion-electron interactions. The electron exchange and correlation have been treated by the generalized gradient approximation (GGA) as parameterized by Perdew, Burke, and Ernzerhof (PBE)[6] functional. The electronic wave-functions are expanded using the plane-wave basis set

with a kinetic energy cut off of 500 eV which ensures the convergence in total energy to a precision of 10^{-6} eV and maximum force on each atom reached less than 0.02 eV $Å^{-1}$. The Brillouin zone was sampled by 4x4x1 Γ-centered k-point meshes for electronic, dielectric, and optical properties calculations. The absorbance was obtained from the frequency-dependent dielectric function based on the following equation[7]

$$\alpha_{abs} = \sqrt{2}\omega\left(\sqrt{\varepsilon_1^2(\omega) + \varepsilon_2^2(\omega)} - \varepsilon_1(\omega)\right)^{1/2} \quad (1)$$

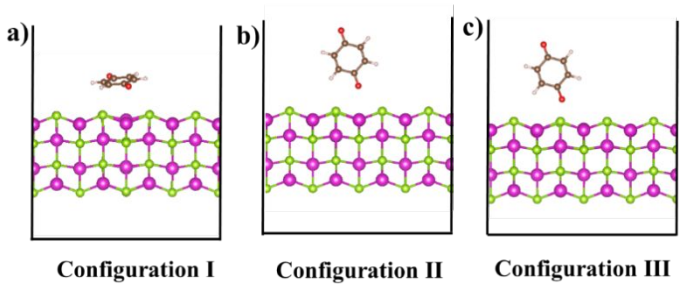
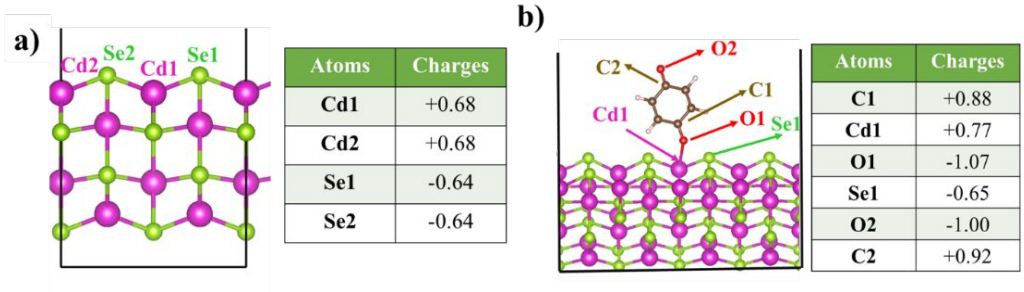


Figure S2. Three different configurations considered for the adsorption of benzoquinone molecule on 4-ML CdSe(110) surface



Atoms	Charges
Cd1	+0.68
Cd2	+0.68
Se1	-0.64
Se2	-0.64

Atoms	Charges
C1	+0.88
Cd1	+0.77
O1	-1.07
Se1	-0.65
O2	-1.00
C2	+0.92

Figure S3. Bader charges on the Carbon(C) ,Cadmium (Cd), Oxygen (O) and Selenium (Se) atoms in a) CdSe(110) surface and b) benzoquinone adsorbed on CdSe(110) surface

Table S2. Calculated excitonic effective mass μ_{ex} (m_0) in units of the rest mass of a free electron (m_0), macroscopic static dielectric constant (ε_r), exciton binding energy E_{exc} (eV) and Bohr exciton radius a^* (Å) for 4-layer CdSe and benzoquinone adsorbed on 4-layer CdSe.

Systems	μ_{ex} (m_0)	ε_r	E_{exc}(eV)	a^* (Å)
4-layer CdSe (110)	0.13	2.80	0.96	2.76
Benzoquinone/4-layer CdSe(110)	0.13	2.98	0.79	3.05

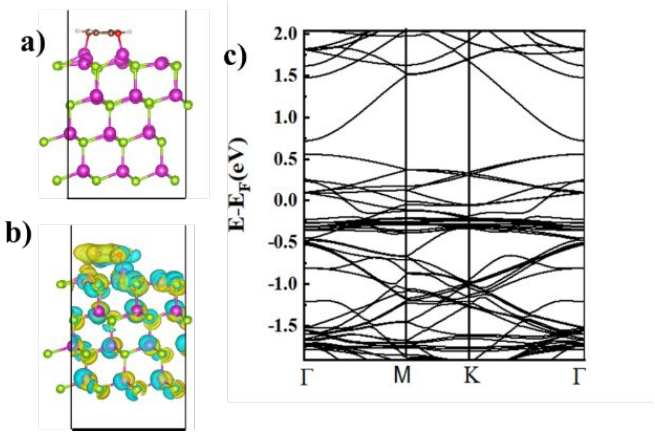


Figure S4. a) Benzoquinone molecule adsorbed on 4-ML CdSe(111) surface, b) 3D iso-surface of charge density difference profile: yellow (cyan) color indicates spatial regions of charge accumulation (depletion) at an iso-surface value of 0.01 e/Å^3 c) Bandstructure of 4-ML CdSe(111).

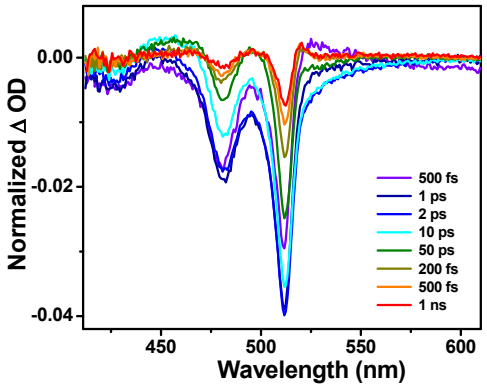


Figure S5. Transient absorption (TA) spectra of pure CdSe NPLs with delay time from 500 fs to 1 ns.

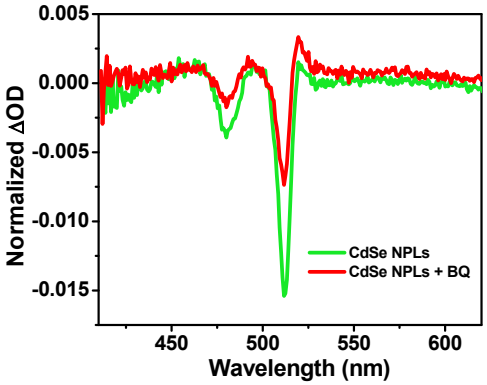


Figure S6. The TA spectrum of pure CdSe NPLs (green) and CdSe NPLs with 416 μM of BQ (red) at 200 ps after excitation at 400 nm.

Table S3. Fitting parameters for pure CdSe NPLs and CdSe NPLs-BQ hybrid

Systems	**τ^g (ps)**	**$\tau_r(a_1\%)$ (ps)**	**$\tau_r(a_2\%)$ (ps)**	**$\tau_r(a_3\%)$**
CdSe NPLs	0.4	-	47.0± 2.3 (55.5%)	>1 ns (44.5%)
CdSe-BQ	0.3	2.1± 0.1 (6.2%)	46.1 ± 2.3 (65.9%)	>1 ns (27.9%)

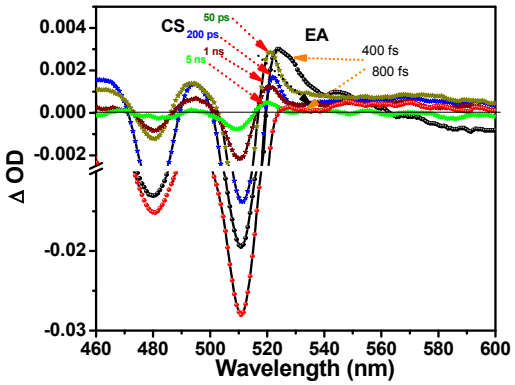


Figure S7. Transient absorption (TA) spectra evolution of the hybrid from 0.4 ps to 5 ns showing EA and CS features.

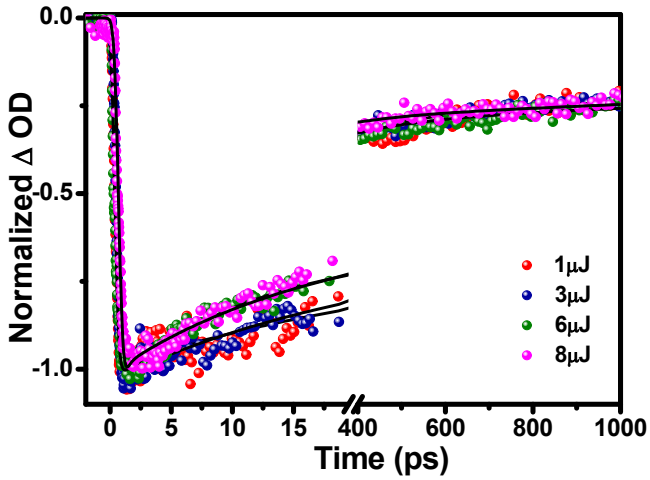


Figure S8. Pump power dependence of the transient bleach decay kinetics in the NPLs-BQ hybrid.

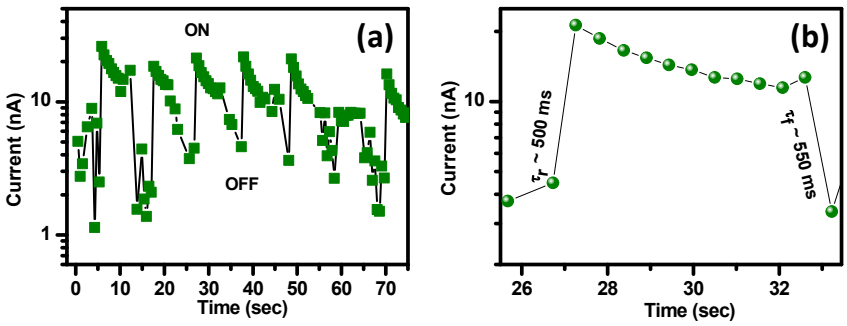


Figure S9. (a) Optical switching characteristics of pure NPLs based device recorded by switching the light source at 2 V bias. (d) Rise and fall time of a single on-off cycle of pure sample.

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