Supporting Information

Quantum Confined Stark Effect in Ensemble of Phase-Pure CdSe/CdS Quantum Dots

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1. SEM image of QCSE device cross-section

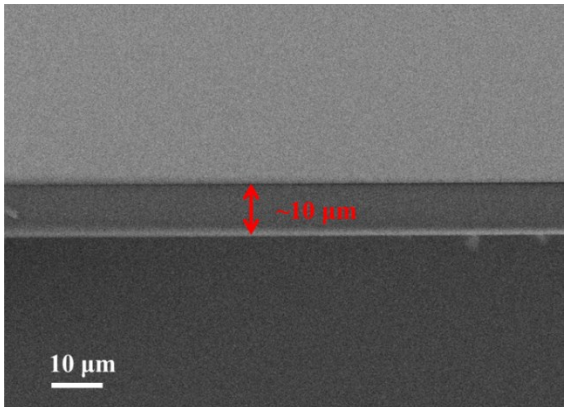

Figure S1. SEM image of QCSE device cross-section. The distance between two ITO electrodes is about ~10 µm.

2. PL spectra of thick-shell CdSe/CdS QDs (11 MLs) under the reverse electric field

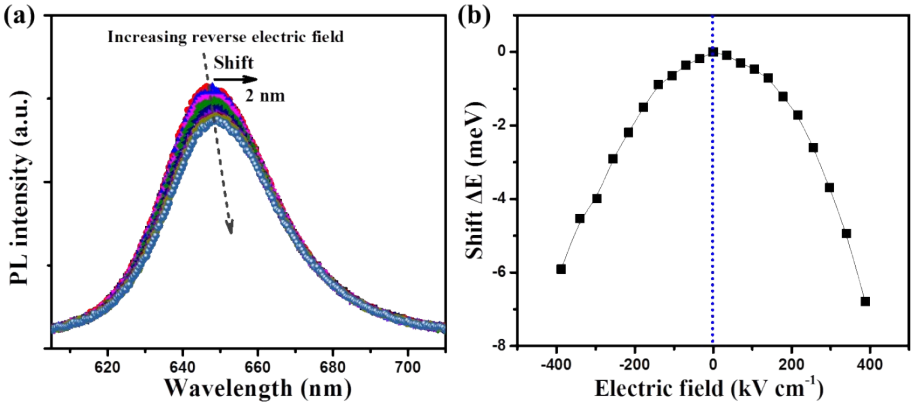

Figure S2. (a) PL spectra of thick-shell CdSe/CdS QDs (11 MLs) with increasing reverse electric field (-390−0 kV cm^{-1}). The PL spectra show a red shift of ~2 nm. (b) The Stark shift versus electric field for the PL spectra of thick-shell CdSe/CdS QDs, under the electric field of -390−390 kV cm^{-1}. The spectra shift at the electric field of 0−390 kV cm^{-1} is symmetric with that under the reverse electric field.

3. Optical properties of thick-shell CdSe/CdS QDs (11 MLs) under electric field

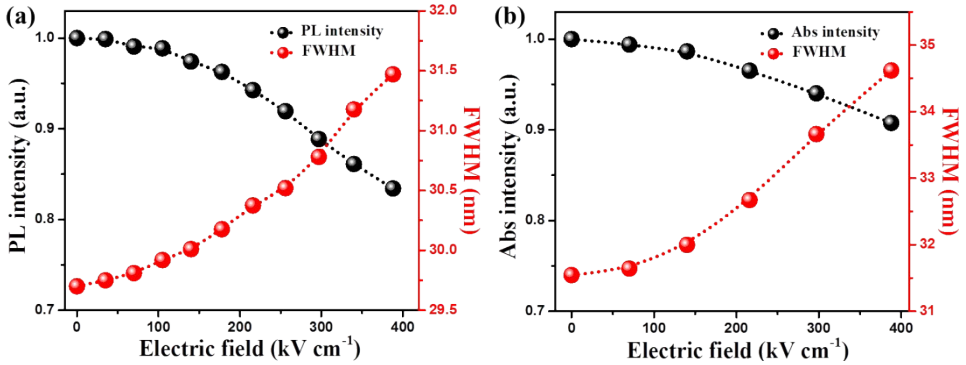

Figure S3. (a) The PL intensity and the full width at half-maximum (FWHM) of PL spectra as functions of the electric field. (b) The absorption intensity and FWHM of the first exciton absorption peak as functions of the electric field.

4. Optical properties of moderate-shell (5 MLs) and ultrathick-shell (15 MLs) CdSe/CdS QDs under electric field

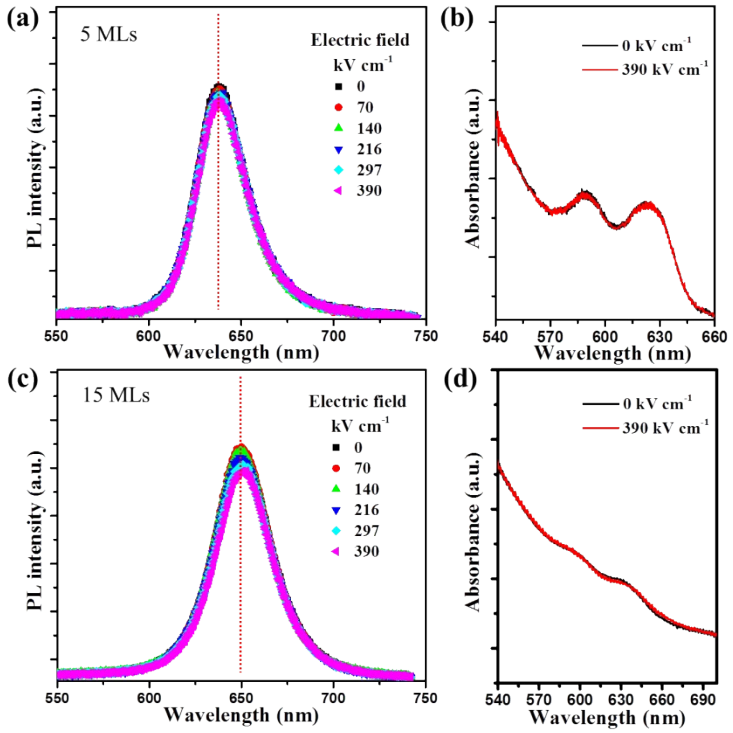

Figure S4. PL (a) and absorption (b) spectra of moderate-shell CdSe/CdS QDs (5 MLs) in QCSE device, measured with increasing electric field. The PL spectra only exhibit a slight decrease of ~6% in emission intensity and an unconspicuous redshift of emission wavelength. The absorption spectra show nearly no obvious changes in the first exciton absorption peak. PL (c) and absorption (d) spectra of ultrathick-shell CdSe/CdS QDs (15 MLs) measured under the electric field. The PL spectra display an emission wavelength redshift of ~1.3 nm and intensity decrease of ~9%. The absorption spectra show a slight change of the first exciton absorption peak.

5. PL blinking traces of single CdSe/CdS QDs with different shell thickness

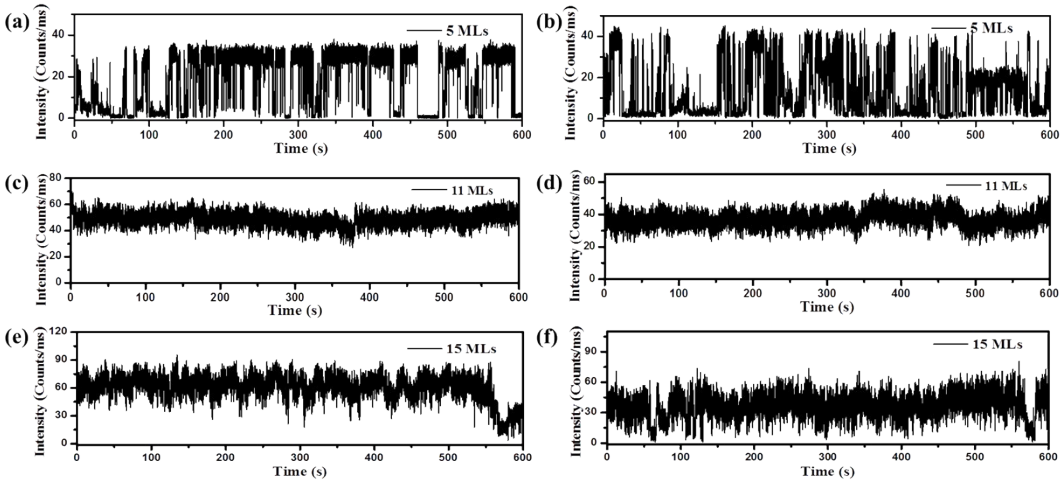

Figure S5. (a-f) The usual PL blinking traces for single CdSe/CdS QDs with 5, 11, and 15 MLs CdS shells, respectively.

6. PL lifetime trace of single ultrathick-shell CdSe/CdS QD (15 MLs)

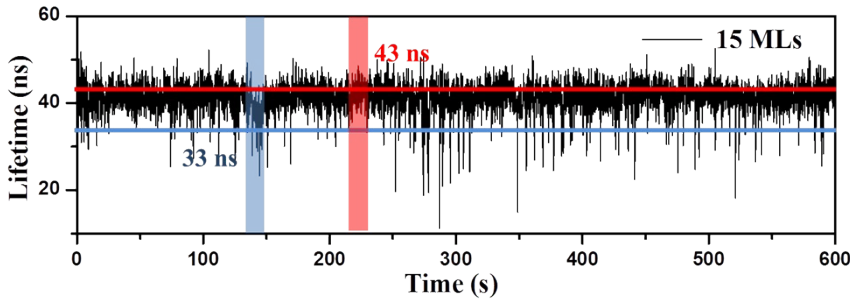

Figure S6. The corresponding time-dependent PL lifetime trace of single ultrathick-shell CdSe/CdS QD (Fig. 2f). The average lifetime of intermediate state (blue region) and "On" state (red region) is 33 and 43 ns, respectively. The experiment result shows that the lifetime of intermediate state is lower than that "On" state. The lower lifetime of intermediate state may be caused by the weakened AR suppression or the increase of strain induced defects with the further increase of CdS shells.[1]

7. Emission spectra of thick-shell CdSe/CdS QDs (11 MLs) in QCSE device

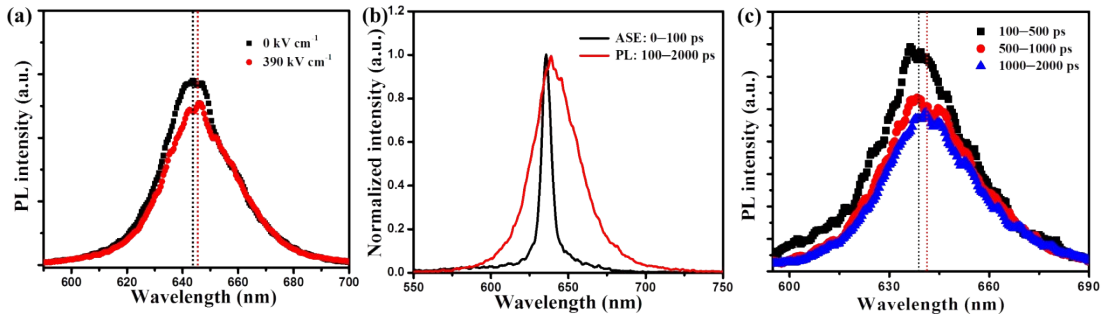

Figure S7. (a) The extracted PL spectra of thick-shell CdSe/CdS QDs (11 MLs) in the PL decay measurements (Fig. 3). The PL spectra show the redshift and broadening of emission peak, as well as the decrease of emission intensity under the electric field, which is consistent with the feature in Fig. 1c. (b) ASE and PL spectra of thick-shell CdSe/CdS QDs for 0 kV cm^{-1}, extracted from Fig. 4a with the integrated time ranges of 0–100 ps and 100–2000 ps, respectively. ASE exhibits an obvious blueshift of ~5 nm relative to PL emission. The reason for the blue-shifted ASE peak is that such QDs have thick CdS shells, which allows the electrons to delocalize freely over the entire QD volume, while the holes are still confined in the core, which results in type-II-like excitons distributions in these thick-shell QDs.[2] According to the reference, because of the repulsive X-X Coulomb interactions in type-II QDs, thus the BX recombination takes place at higher photon energies.[3] (c) Time-dependent PL spectra for thick-shell QDs under the electric field of 390 kV cm^{-1}, extracted from Fig. 4a with the integrated time ranges of 100–500 ps, 500–1000 ps, and 1000–2000 ps, respectively. At the multiexciton state, the excitons tend to occupy a higher energy level near the band edge due to the repulsive Coulomb interactions as mentioned above. When all multiexcitons have already decayed (>100 ps), the remaining single exciton gradually relax to the band-edge emission state following the diminishing repulsive Coulomb interactions of exciton-exciton, thus the single exciton recombination will back to the lowest photon energies. Therefore, the PL spectra display an obvious redshift process (from black dashed line to red dashed line) along with the single exciton decay (100-2000 ps). The continuous redshift process of the emission peak position is shown as the red dashed line in Fig. 4a.

8. ASE emission intensity as a function of pump intensity for thick-shell CdSe/CdS QDs (11 MLs)

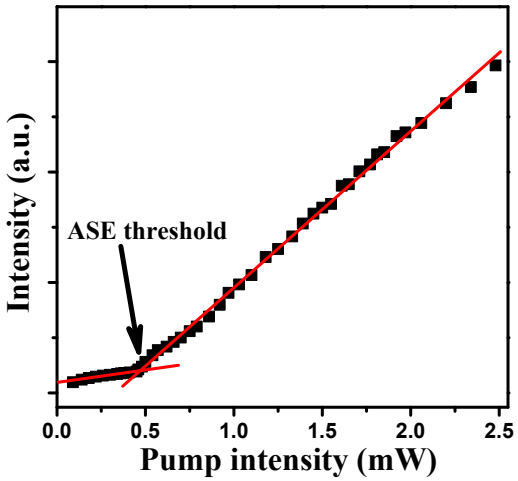

Figure S8. ASE emission intensity as a function of pump intensity, pumped at 400 nm. The emission intensity of ASE shows a steep linear growth, when the pump intensity is increased above threshold.

References

1. P. Spinicelli, S. Buil, X. Quelin, B. Mahler, B. Dubertret and J.-P. Hermier, *Phys.*

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3. S. A. Ivanov, J. Nanda, A. Piryatinski, M. Achermann, L. P. Balet, I. V. Bezel, P. O. Anikeeva, S. Tretiak, V. I. Klimov, *J. Phys. Chem. B*, 2004, **108**, 10625−10630.