Supporting Information

Direct Three-Dimensional Observation of Core/Shell Structured Quantum Dots with a Composition-Competitive Gradient

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Figure S1. SEM image of agglomerated CdSSe/ZnS QDs on Si wafer.



Figure S2. Determination of mean diameter of the STEM-observed QDs.



Figure S3. (a-c) APT reconstruction of CdSSe/ZnS QD data set that is different from the one in figure 4. Also, Cd 13.6 #/nm³ (green), Se 5.3 #/nm³ (red), and Zn 8.8 #/nm³ (purple) isodensity surfaces were chosen in the reconstruction as each threshold value to distinctly display Cd-, Se-, and Zn-enriched regions. (d) 3D atom map of Cd (green), Se (red), Zn (S) (purple) including the additionally detected molecular species, Se₂ (orange), ZnS (blue), CdS (violet) and Cd₂S (pink).



Add Edit				
Ion Name	Charge States	Include in Calculation		
Cd	+1, +2			
Se	+			
Se2	+1, +2			
Zn	+1, +2			
ZnS	+1, +2			
CdS	+1			
Cd2S	+2			
S	+1			
IS2	+1			

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Decompose Reset Ranges

Ions Mass Spectrum Decomposition Bulk Composition Decomposition Details Table					
lon	Decomposed Count	Atomic %	Atomic Error%		
Se	2,039	8,694%	±0,222%		
S	9,696	41,346%	±0,276%		
Cd	6,543	27,895%	±0,365%		
Zn	5,176	22,066%	±0,693%		

Figure S4. APT mass spectrum of CdSSe/ZnS QDs: (a) peak identification of all detected ions including molecular ions. Substantial peak overlaps between Zn and S or between Se and Se₂ were found in the spectrum, as marked with the blue boxes. (b) All of the peaks with ion overlaps discovered in the mass spectrum are presented by green lines. (c) Ions included in peak decomposition details table. (d) Bulk composition measured after such peak decomposition method of APT software.



Figure S5. APT mass spectrum, extracting from only core region of QDs: (a) 32-34 Da, (b) 64-68 Da. Ranges of mass-to-charge-state ratios detected from other species are omitted. By quantifying peak contributions from isotopic natural abundances, we determined that for the QD core the peaks at 32-34 Da were derived predominantly from S_2^{2+} and also S^{2+} at 64-68 Da.