

Supplementary information for

**From AI Discovery to Macroscopic Formula: A Quasi-Stability
Criterion for Quantum Dots Inspired by Elon Musk's Vision**

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Supplementary Method 1.

Key prompt for grok4 based AI-supervisor

##Objective

Develop a unified theoretical framework for quantum dot (QD) stability (internal stability and aggregate stability), generate an equation or inequation clearly to judge the stability, using measurable macroscale parameter. Prioritizing the nucleation stage during initial growth. Use only macroscopically measurable physical quantities. The framework must integrate:

##Core Theories:

LSW (Ostwald ripening), DLVO (colloidal stability), depletion theory (polymer/solvent interactions), CNT (nucleation), and Lamer mechanism (burst nucleation), and others related

Size Regime Focus: Explicit treatment for QDs <3 nm (quantum confinement effects)

Morphologies:

Spherical (isotropic growth)

Rod-like (anisotropic growth with aspect ratio dependence)

##Derivation Requirements:

Use time-dependent Hamiltonian to model dynamic evolution during nucleation/growth

Justify separation of timescales (e.g., fast electrostatic vs. slow diffusive processes)

Incorporate potential energy terms from integrated theories with rigorous coupling analysis (e.g., depletion-modified DLVO barriers, LSW-quantum size coupling)

##Key Couplings:

Quantum confinement → cohesive energy modulation

Solvent-mediated phase transitions

Ligand-QD bonding energy (Pb-S vs. Pb/ligand)

Depletion forces → DLVO barrier suppression

DLVO and depletion forces affect nucleation and QD shape (symmetrical sphere or asymmetrical, like rod)

##Critical Parameters:

Material: Cohesive energy, surface energy, Pb-S bond energy

Solvent: Dielectric constant (ϵ), solvent molecule size (σ), concentration (c)

Ligand: Chain length (L),

Environmental: Temperature (T), pressure (P)

Electrostatic: Bjerrum length (λ_B), Debye length (λ_D)

and so on.

##Stability Definition

Internal Structural Stability:

Threshold for lattice integrity (bond breakage prevention from ligand or solvent molecule)

Quantum size-dependent cohesive energy ($E_{\text{coh}}(\text{size})$)

presents a clear equation or inequation for the stability criteria condition, in the equation or inequation, if possible, using more macroscale property of all the materials, including QDs (like dielectric constant, mass density, band gap and others), solvent (mass density, viscosity, and others) molecules,

##Dispersion Stability:

Aggregation prevention

lattice collapse

##Structural Guidelines:

Explicitly state assumptions for theory integration

Use tensor notation for anisotropic rod-like QD stability

Include phase diagrams showing stability regimes in (size, solvent ϵ , ligand L) parameter space

##Key Improvements:

The mathematic form present in latex printed form, make sure detailed derivation

##Tool Usage Rules

You should call tools whose names start with AI Supervisor to perform search, data collection, and information extraction

Before producing any final output for the user, you must call the regulator tool Regulator.review, providing the draft you intend to output and your supporting evidence/citations.

If Regulator.review indicates that the draft does not meet requirements or includes required actions, you must continue reasoning and tool use until the requirements are satisfied or the iteration limit is reached.

##Factuality and Citations

All factual conclusions must be supported by evidence obtained through AI Supervisor tools; do not fabricate details.

##Answer Style

Always give a direct, user-ready answer first, then briefly provide “why / basis / citations”.

Avoid exposing long reasoning traces (keep those internal).

If beyond your capability or lacking evidence, state this explicitly and suggest a feasible next step.

##Termination and Safety

At most <MAX_ITERATIONS> cycles of “continue reasoning → self-check”.

If the limit is reached without approval, gracefully degrade: provide the most reliable partial answer available + highlight missing parts + suggest next steps.

Key response from grok4 based AI-supervisor

AI Supervisor: Step-by-Step Deduction of a Quantum Dot Stability Criterion

1) Data Ingestion and Standardization

- Corpus scope: Group meeting discussion record including many literatures analysis and information.
- Entity extraction: Material system, ligand type, solvent, process/environmental parameters, outcome label (stable/unstable).
- Variable alignment: QD radius r , ligand effective thickness L , solvent viscosity η , absolute temperature T , Boltzmann constant k_B , solvent dielectric constant ϵ , and electronegativity differences $\Delta\chi_{QD}$ and $\Delta\chi_{ads}$.

2) Physical Priors → Observable Mapping

The AI system adopts the principle of “macroscopically measurable observables only” and formulates four stabilization mechanisms: (I) Surface bond energy prevents lattice decomposition; (II) Ligand adsorption prevents other molecules from approaching; (III) Ligand steric hindrance creates a physical barrier; (IV) Solvent diffusion dynamics govern encounter rates. A DLVO-type electrostatic destabilization effect is added via the dielectric constant.

3) Mechanism-by-Mechanism Derivation

3.1 Surface Bonding and Size Effect

- Pauling relation for heteronuclear bonds:

$$\Delta \equiv D_{XY}^{(meas)} - 1/2(D_{XX} + D_{YY}) \quad (S1.1)$$

Empirically $\Delta \propto (\Delta\chi)^2$, hence:

$$E_{bond} \propto (\Delta\chi_{QD})^2 \#(S1.2)$$

- Gibbs–Thomson: $\Delta\mu(r) = 2\gamma\Omega/r$ shows that smaller QDs have higher chemical potential and decompose more easily. Stronger bonds counteract this.

3.2 Ligand Adsorption Thermodynamics

- Langmuir isotherm:

$$\theta = \frac{Kc}{1 + Kc} \quad K \propto e^{-\frac{\Delta G_{ads}}{k_B T}} \#(S1.3)$$

- Bond analogy: adsorption strength scales as

$$E_{ads} \propto (\Delta\chi_{ads})^2 \#(S1.4)$$

3.3 Steric Hindrance

Due to missing information on ligand coverage and flexibility in the literature, we consider reasonable approximations.

With $x = L_{eff}/r \geq 0$, geometry ensures $(1+x)^3 - 1 \geq 3x$ and $(1+x)^2 - 1 \geq 2x$. Thus a conservative bound:

$$steric \geq C_0 \theta \frac{L_{eff}}{r} \#(S1.5)$$

This reflects geometric exclusion and is independent of the diffusion time τ_L .

3.4 Diffusion-Controlled Dynamics

- Stokes–Einstein:

$$D = \frac{k_B T}{6\pi\eta r} \#(S1.6)$$

- First-passage time:

$$\tau_L = \frac{(r+L)^3 - r^3}{3Dr} \approx \frac{rL}{D} \#(S1.7)$$

Substitution gives:

$$\tau_L \propto \frac{\eta r^2 L}{k_B T} \#(S1.8)$$

3.5 Dielectric Destabilization

DLVO models predict the repulsive barrier decreases as ϵ rises. A factor $1/\epsilon$ captures this.

4) Stability Criterion

Combining contributions yields:

$$(\Delta\chi_{QD})^2 (\Delta\chi_{ads})^2 \cdot \frac{L}{r} \cdot \frac{\eta r^2 L}{k_B T \epsilon} > \tau_0 \#(S1.9)$$

τ_0 is a critical threshold (~ 6 ps) from empirical calibration.

5) Dimensional Consistency

- Left-hand side has units of time, directly comparable to τ_0 .

6) Failure Modes

- Extremely small QDs: surface energy dominates, requiring explicit γ .
- Reactive/photochemical conditions: reaction terms must be added.
- Mixed ligands/solvents: weighted averages are needed.

Supplementary Method 3.

Quantum Dot Synthesis and Sources

The preparation methods and sources of the three types of quantum dots used in this study are described as follows.

Synthesis of CdSe Quantum Dots

CdSe quantum dots were synthesized using the classic hot-injection method, which was based on previously reported methods with optimization improvements.¹ The specific synthesis process is as follows:

0.255 g of cadmium oxide (CdO, 99.99%, Aldrich), 3.1 mL of oleic acid (OA, technical grade), and 30 mL of 1-octadecene (ODE, technical grade) were loaded into a three-neck flask and degassed at 100°C for 1 hour. After degassing, the temperature was increased to 230°C under nitrogen flow and maintained for 15 minutes to ensure complete conversion of cadmium oleate. Subsequently, a TOPSe mixture (0.3 mL of 2M TOPSe dissolved in 2.5 mL ODE) was rapidly injected. Eight minutes after the first injection, 0.3 mL of 2M TOPSe was added dropwise. To promote further growth of quantum dots, the dropwise injection step was repeated twice (0.3 mL of 2M TOPSe for each injection), with a reaction time of 5 minutes for each injection. Finally, the heating mantle was removed to cool the three-neck flask.

Synthesis of PbS Quantum Dots

PbS quantum dots were synthesized using the hot-injection method as well, based on previously reported methods with corresponding optimizations.² The synthesis steps are as follows:

0.45 g of lead oxide (PbO, 99.999%, Alfa Aesar), 1.5 mL of oleic acid (OA, 99%), and 10 mL of 1-octadecene (ODE) were placed in a three-neck flask. The mixture was degassed at 100°C for 1 hour. Then, the temperature was increased to 110°C under nitrogen flow, and 210 μ L of

bis(trimethylsilyl)sulfide ((TMS)₂S) dissolved in 4 mL ODE was rapidly injected. The heating mantle was removed immediately after injection to cool the flask.

Source of InP/ZnS Quantum Dots

InP/ZnS quantum dots were directly purchased from commercial suppliers with the following information:

- Supplier: Sigma-Aldrich (Product No.: 776785, St. Louis, MO, USA)

Quantum Dot Purification Methods

All synthesized quantum dots were purified using standard solvent precipitation methods:

CdSe Quantum Dot Purification: 15 mL of ethanol and 15 mL of acetone were added to 5 mL of CdSe quantum dot crude product and centrifuged at 6000 rpm for 10 minutes. The supernatant was discarded, and the precipitate was redispersed in 2 mL of toluene. After adding 10 mL of ethanol and 10 mL of acetone, the solution was centrifuged for 10 minutes at 6000 rpm, and finally the quantum dots were redispersed in toluene, tetrachloroethylene, or deuterated benzene for subsequent characterization.

PbS Quantum Dot Purification: A similar purification process was used, where 15 mL of ethanol and 15 mL of acetone were added to 5 mL of PbS quantum dot crude product, centrifuged, and then redispersed in 5 mL of toluene. After further purification, they were dispersed in corresponding solvents.

Quantum Dot Property Characterization

The properties of quantum dots were characterized by the following methods: measured using a fluorescence spectrometer at room temperature with a xenon lamp as the excitation source.

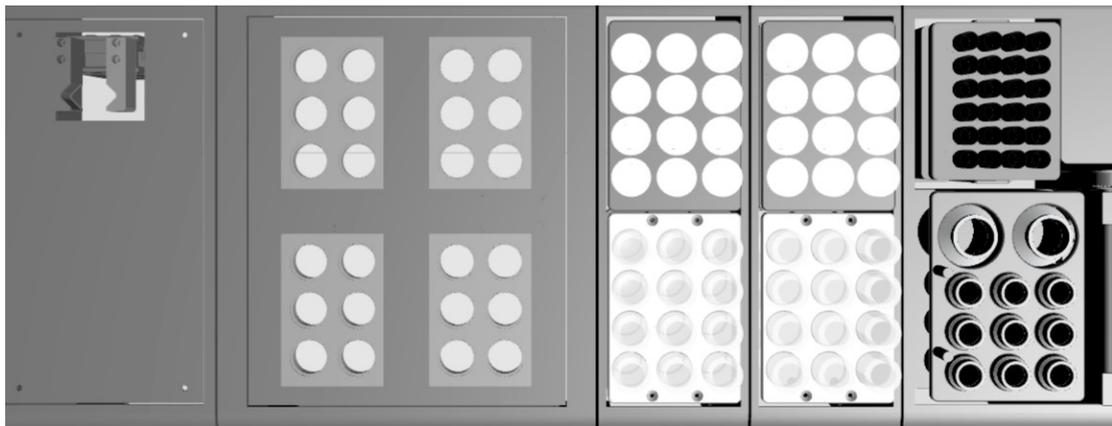
Supplementary Method 4.

All procedures described in this study were performed using the HT03 high-throughput workstation. The platform enables fully automated operations, including reagent dispensing, reaction under inert atmosphere, liquid-liquid extraction, solid-phase extraction, and preparation of samples for HPLC analysis

Instrumentation

HT03 System (codename “Yimùmián”)

1. Module overview (from left to right)



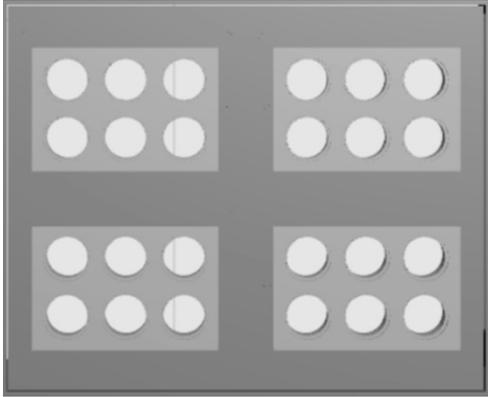
2. Capping/decapping zone

Automates opening/closing of bottle caps for sealing management and state switching.



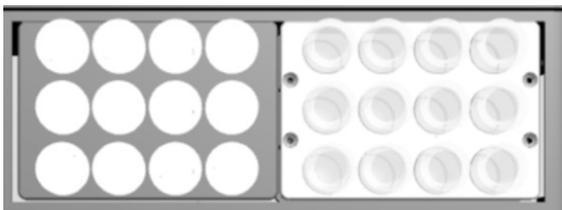
3. High-temperature module

Four independent temperature zones, each with 6 positions; settable range 25–300 °C. Optional magnetic stirring, 0–1000 rpm. This module does not include bottles by itself—the bottles are moved in from the reaction module before setting reaction conditions.



4. Reaction module

Upper “cap area” and lower “bottle area,” 12 positions each; caps and bottles are paired one-to-one. Bottles in the reaction module can be dosed from the reagent module for liquid mixing and sample storage.



5. Needle/tip zone

Storage and changeover area for pipetting components, providing 24 disposable tips for liquid-

handling operations.



6.Reagent module

Holds reagents required for the current experiment; replenished manually. Includes 2×100 mL containers and 9×40 mL containers.



Reference

- (1) Peng, Z. A.; Peng, X. Formation of High-Quality CdTe, CdSe, and CdS Nanocrystals Using CdO as Precursor. *Journal of the American Chemical Society* **2001**, *123* (1), 183-184. DOI: 10.1021/ja003633m.
- (2) Hines, M. A.; Scholes, G. D. Colloidal PbS nanocrystals with size-tunable near-infrared emission: observation of post-synthesis self-narrowing of the particle size distribution. *Advanced materials* **2003**, *15* (21), 1844-1849.