LAM Seminar Series lecture

October 21, 2024, 10:00am, Large conference room,
Comenius University Science Park,

Open to students, faculty and public

Internal and External Control of Excitons in Colloidal Quantum Dots

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Colloidal quantum dots (cQDs) synthesized in simple laboratory flasks are finding real-world applications in demanding technologies from displays and lighting to photovoltaics and photodetectors. In the future, cQDs may be the basis for single-photon devices in quantum networks. Beyond quantum-size control, we pursue an expanded "structural toolbox" to synthetically engineer new quantum emitters with targeted photophysical properties, including non-blinking behavior, biexciton enhancement, dual-color emission.[1-9] Taking advantage of their solution-phase processibility, we prepare cavity- or antenna-coupled cQD hybrid materials using, e.g., deterministic, direct-write nanointegration.[10,11] The former—precision synthesis—affords *internal control* over excitonic properties,[1-9] while the latter—hybrid materials fabrication—affords *external control* via local environmental effects.[10-14]

References. (1) Singh, A. et al. *Small Sci.* **3**, 2300092 (2023).; (2) Krishnamurthy, S. et al. *ACS Nano* **15**, 575 (2021); (3) Dennis, A. M. et al. *Adv. Funct. Mater.* **29**, 1809111-1-10 (2019); (4) Hanson, C. J. et al. *J. Am. Chem. Soc.* **139**, 11081 (2017); (5) Mishra, N. et al. *Nature Commun.* **8**, 15083-1-9 (2017); (6) Mangum, B. D., et al. *Nanoscale* 6, 3712 (2014); (7) Dennis, A. M. et al. *Nano Lett.* **12**, 5545 (2012); (8) Ghosh, Y. et al. *J. Am. Chem. Soc.* **134**, 9634 (2012); (9) Chen, Y. et al. *J. Am. Chem. Soc.* **130**, 5026, (2008); (10) Lubotzky, B. et al. *Nano Lett.* **24**, 640 (2024); (11) Abudayyeh, H. et al. *APL Photonics* **6**, 036109-1-7 (2021); (12) Mishra, S. et al. *ACS Nano* **18**, 8663 (2024); (13) Dolgopolova et al. *Nanoscale Horiz.* **7**, 267 (2022); (14) Gao, Y. et al. *Adv. Optical Mater.* **3**, 39 (2015).

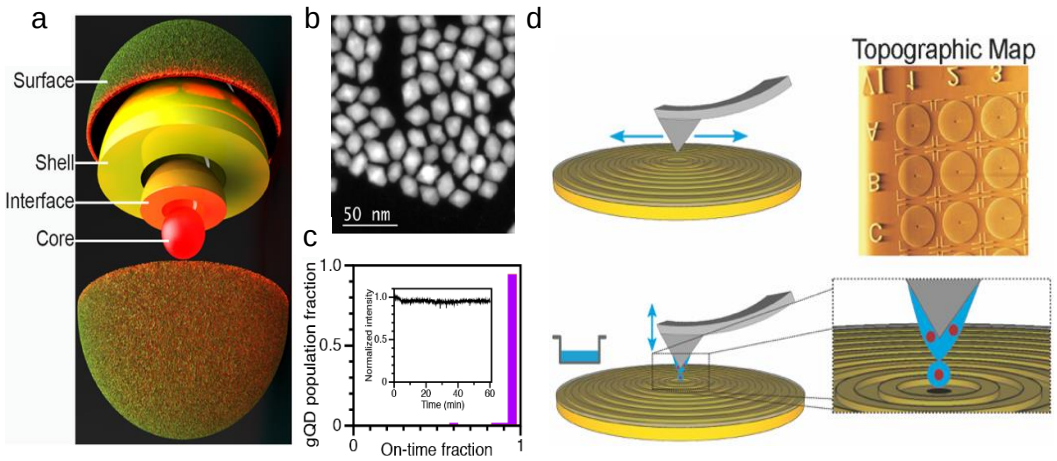

Figure 1. (a) Schematic illustration of the components of a core/shell QD accessible for precision synthetic manipulation and internal exciton control. (b),(c) CdSe/CdS core/thick-shell "giant" QDs (gQDs): electron microscopy images (b) and plot of population fraction versus on-time fraction revealing extreme non-blinking/non-photobleaching photoluminescence (c);1 such on-demand photon emission now demonstrated for gQDs from blue-green to the full telecommunications window.1-9 (d) "Direct-write" method for coupling quantum emitters to nanoantennas and photonic structures for external exciton control:10-11 (top) AFM scan of area of interest; right panel shows image of scanned hybrid metal–dielectric bullseye antenna array; (bottom) AFM writing tip is wetted, and a droplet of dilute QD suspension is placed at the center of each bullseye structure (guided by the image created in the first step), leaving behind a single QD.

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Dr. Jennifer A. Hollingsworth is a Laboratory Fellow at Los Alamos National Laboratory (LANL). She is a Fellow of the American Chemical Society (ACS), the American Physical Society (Materials Physics) and the American Association for the Advancement of Science (Chemistry), and serves as an elected Councilor for the ACS Division of Colloid & Surface Chemistry. She holds a BA in Chemistry from Grinnell College and a PhD degree in Inorganic Chemistry from Washington University in St. Louis. She joined LANL as a Director's Postdoctoral Fellow in 1999, becoming a staff scientist in 2001. In 2013, she was awarded the LANL Fellows' Prize for Research for her discovery of non-blinking "giant" quantum dots. She serves as Thrust Leader for Nanophotonics & Optical Nanomaterials in the Center for Integrated Nanotechnologies (CINT), a US-DOE Nanoscale Science Research Center and User Facility. Her research interests include discovery, rational design, and development of novel colloidal quantum emitters, unique synthesis and assembly techniques (e.g., automated synthesis, microfluidics-enabled nanowire growth, and scanning-probed-enabled direct-write nanointegration), and nanomaterials applications, such as bioimaging, solid-state lighting and single-photon quantum light sources. In these areas, she has ~125 publications, >17,000 citations, and an h-index of 54.

Selected Representative Publications:

1. Y. Chen, J. Vela, H. Htoon, J. L. Casson, D. J. Werder, D. A. Bussian, V. I. Klimov, J. A. Hollingsworth. "Giant" Multishell CdSe Nanocrystal Quantum Dots with Suppressed Blinking: *J. Am. Chem. Soc.* **2008,** *130*, *15*, 5026–5027
2. J.M. Pietryga, R. D. Schaller, D. Werder, M. H. Stewart, V. I. Klimov, J. A. Hollingsworth. Pushing the band gap envelope: mid-infrared emitting colloidal PbSe quantum dots, *J. Am. Chem. Soc.* **2004**, *126*, *38*, 11752–11753.
3. J. M. Pietryga, D. J. Werder, D. J. Williams, J. L. Casson, R. D. Schaller, V. I. Klimov, J. A. Hollingsworth. Utilizing the lability of lead selenide to produce heterostructured nanocrystals with bright, stable infrared emission. *J. Am. Chem. Soc.* **2008**, *130*, *14*, 4879–4885.
4. S. Jeong, M. Achermann, J. Nanda, S. Ivanov, V. I. Klimov, J. A. Hollingsworth. Effect of the thiol−thiolate equilibrium on the photophysical properties of aqueous CdSe/ZnS nanocrystal quantum dots. *J. Am. Chem. Soc.* **2005**, *127*, *29*, 10126–10127
5. Y. Ghosh, B. D. Mangum, J. L. Casson, D. J. Williams, H. Htoon, J. A. Hollingsworth. New insights into the complexities of shell growth and the strong influence of particle volume in nonblinking "giant" core/shell nanocrystal quantum dots. *J. Am. Chem. Soc.* **2012**, *134*, *23*, 9634–9643