

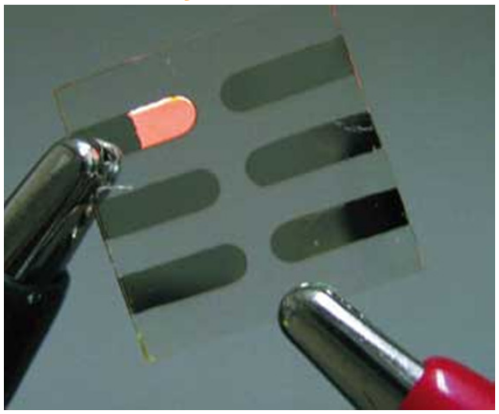
Continuous-wave pumped lasing using colloidal CdSe quantum wells

Joel Q. Grim, Sotirios Christodoulou, Francesco Di Stasio, Roman Krahne, Roberto Cingolani, Liberato Manna, Iwan Moreels

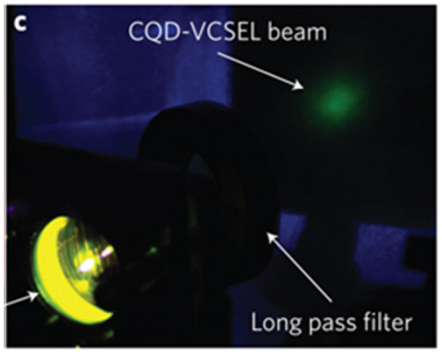
Istituto Italiano di Tecnologia,
Genova, Italy

Nanocrystals in photonic applications

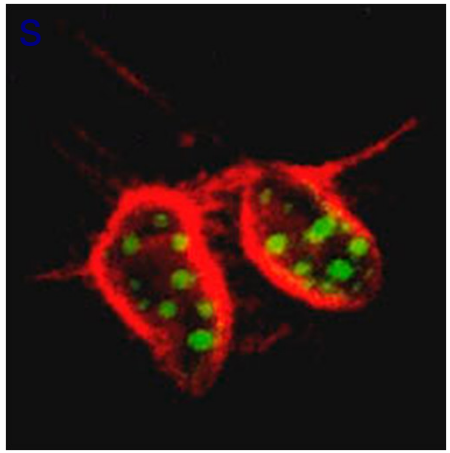
direct injection LEDs



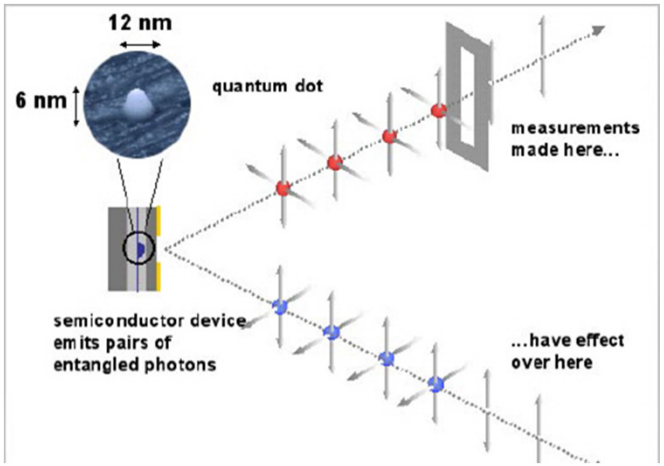
lasers



biolabel



Quantum emitters



lighting

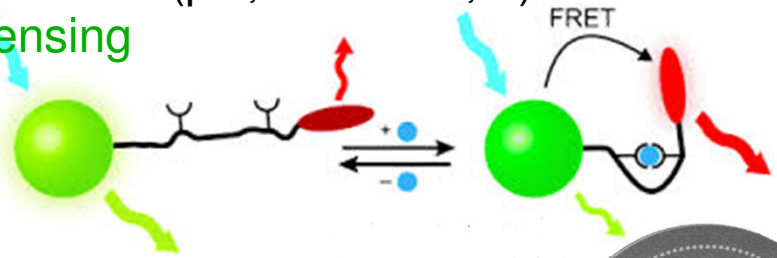


phosphors for displays



TRILUMINOS DISPLAY for mobile

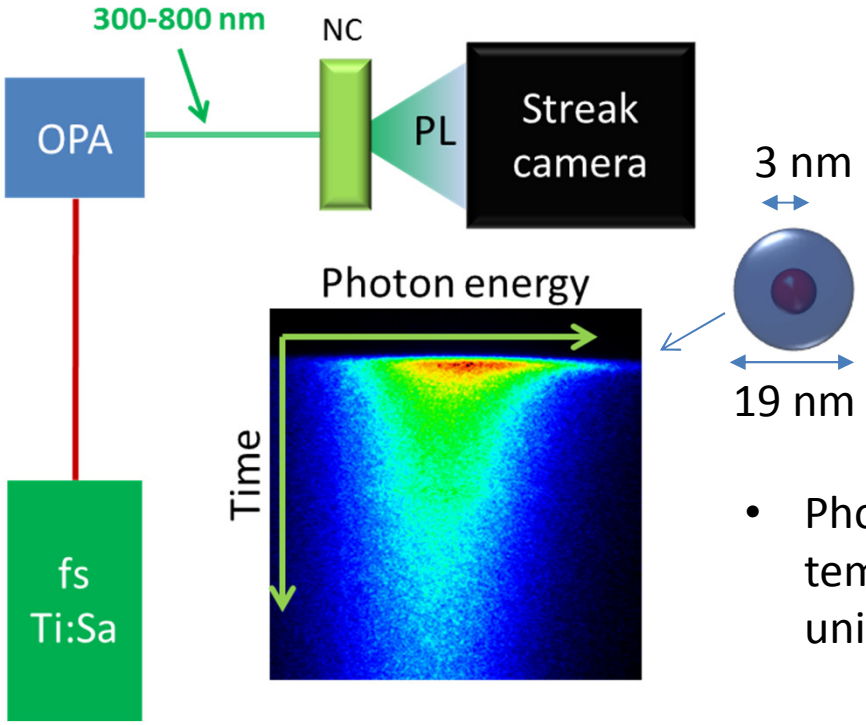
chemical (pH, molecule,...) sensing

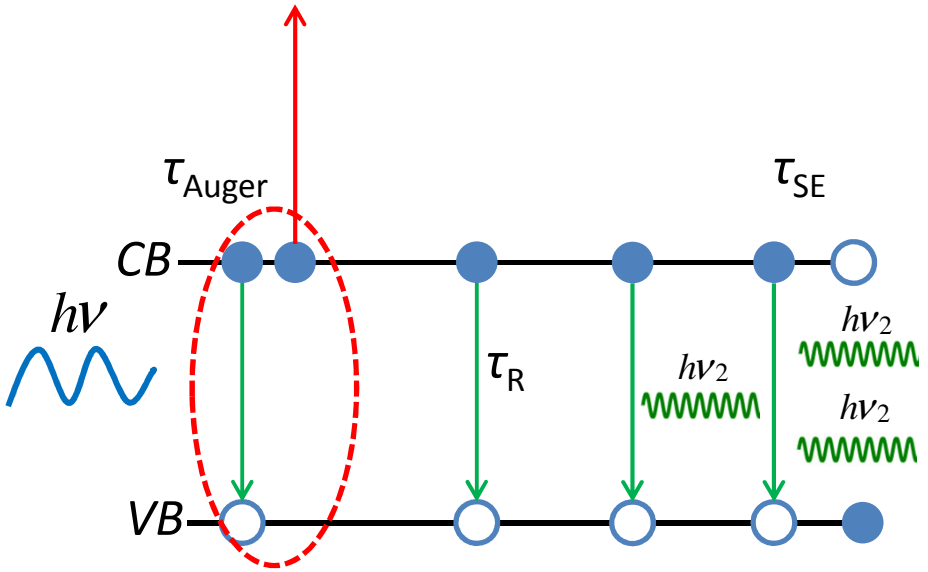


Colour-converters for energy applications
physical (pressure, temperature) sensing
memory/storage devices

Carrier dynamics probed with ultrafast spectroscopy

Measurements of PL excited with ultrafast amplified laser pulses



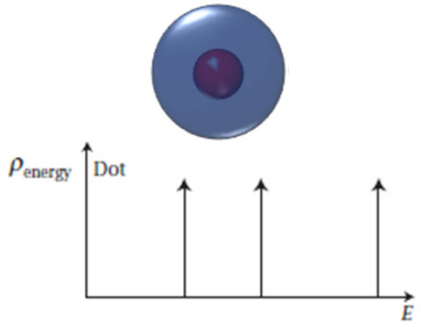


- Photo-excited luminescence at low (4K) to room temperature provides understanding of the physics in the unique nanocrystal geometries produced in our lab.
- Measured properties are used to inform future synthesis efforts based on desired application-specific properties.

Synthesis ⟷ **Probing/understanding basic physics** ⟷ **Applications**

Quantum dot lasing: a paradox

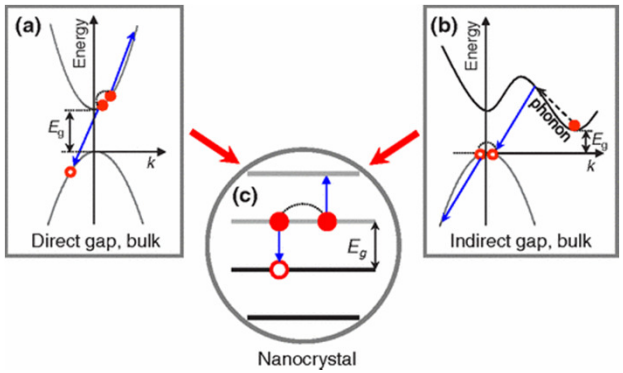
Advantages

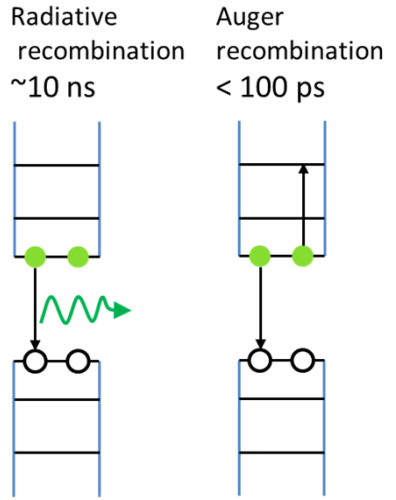


Delta-like density of states
High band-edge DOS
Color tunability
Solution-based synthesis

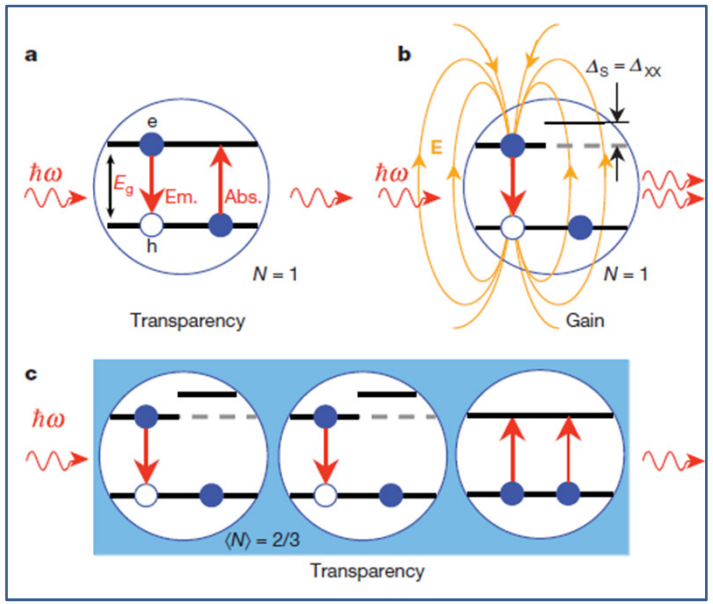
Disadvantage

Enhanced Auger rates





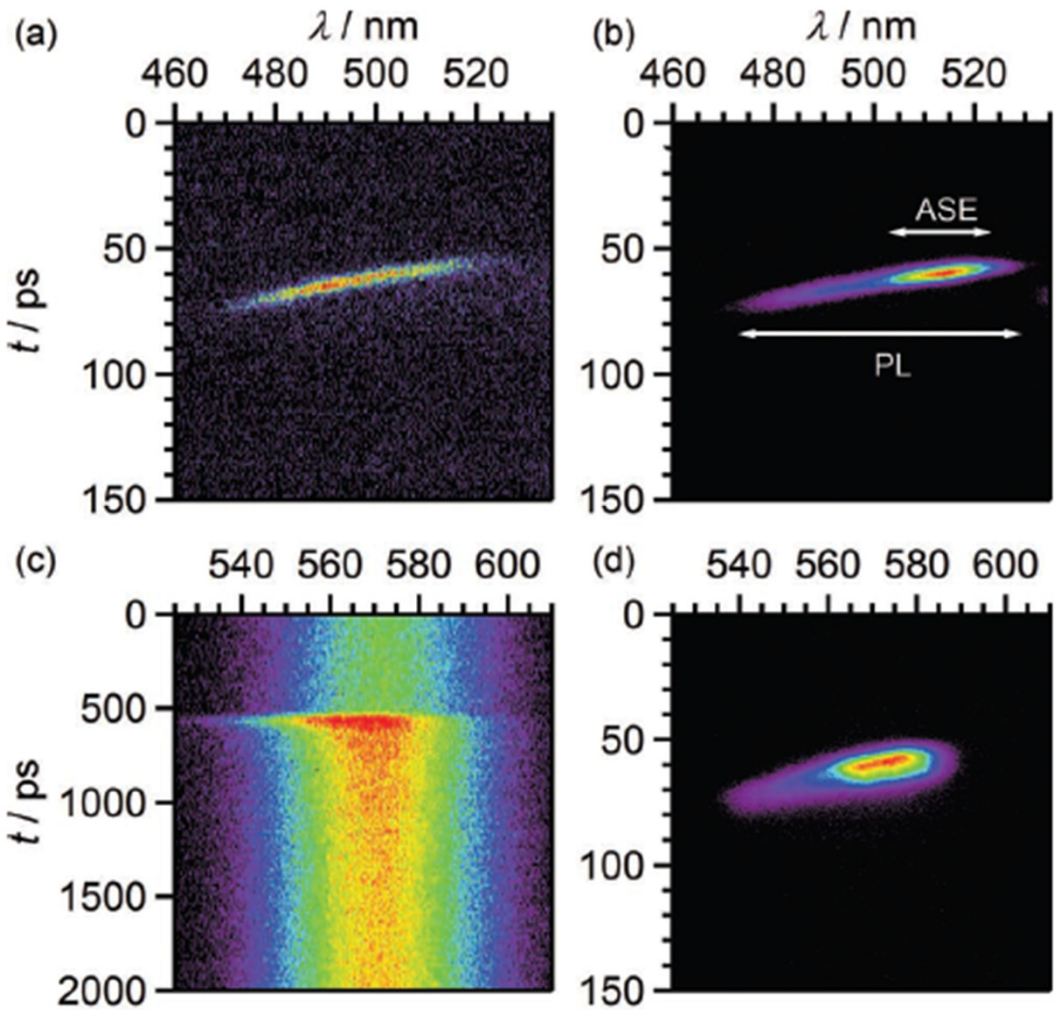
One solution: single-exciton gain



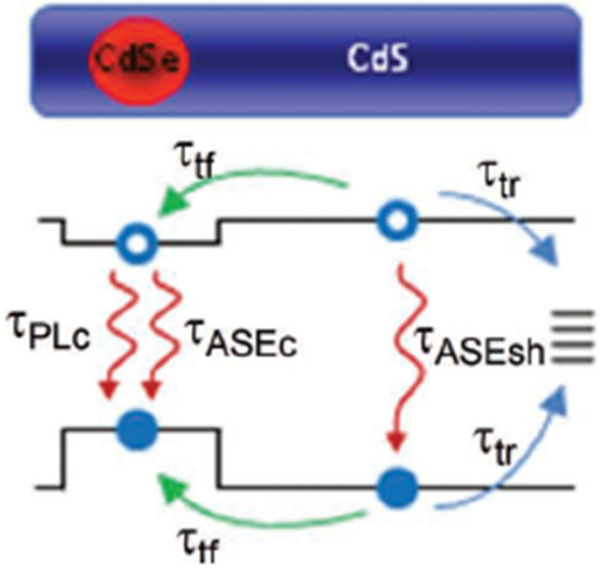
Klimov et al., *Nature* **447**, 441-446 (2007)

Temperature-independent stimulated emission

SE mechanism: carrier relaxation rates



I. Moreels et al., *Adv. Mater.*, **24**, OP231-OP235 (2012)

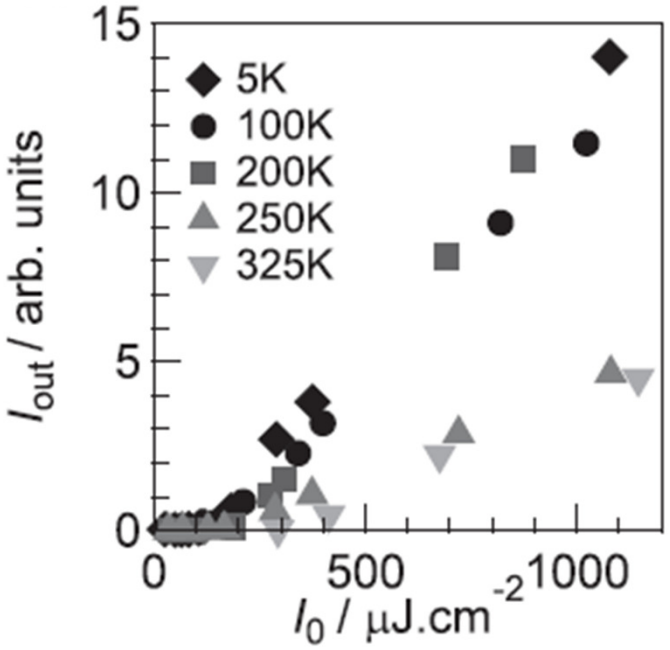


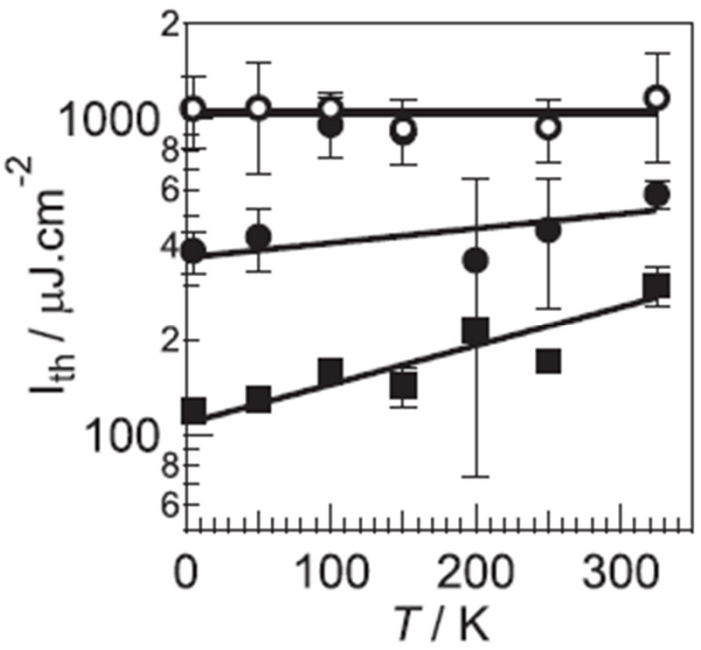
$\tau_{PL,sh}$ = 6 ps

$\tau_{ASE,sh}$ < 2 ps

Both comparable to ps electron transport timescale in 50 nm CdS rods. Rod length determines carrier relaxation.

Temperature-independent stimulated emission

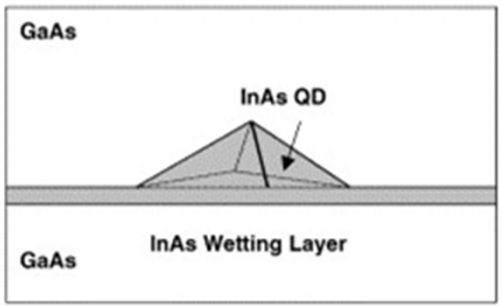




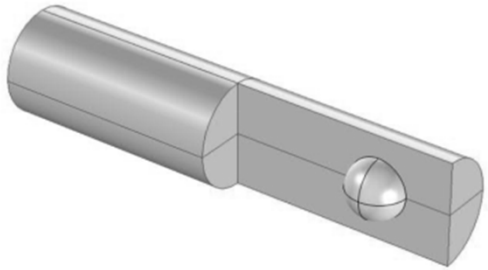
$$I_{th} = I_{th,0} \cdot \exp\left((T - T_0) / T_0\right)$$

$$T_0 = I_{th} / (\partial I_{th}/\partial T)$$

T_0 = 350K



http://www.sciencedirect.com/science/article/pii/S0022024801009848



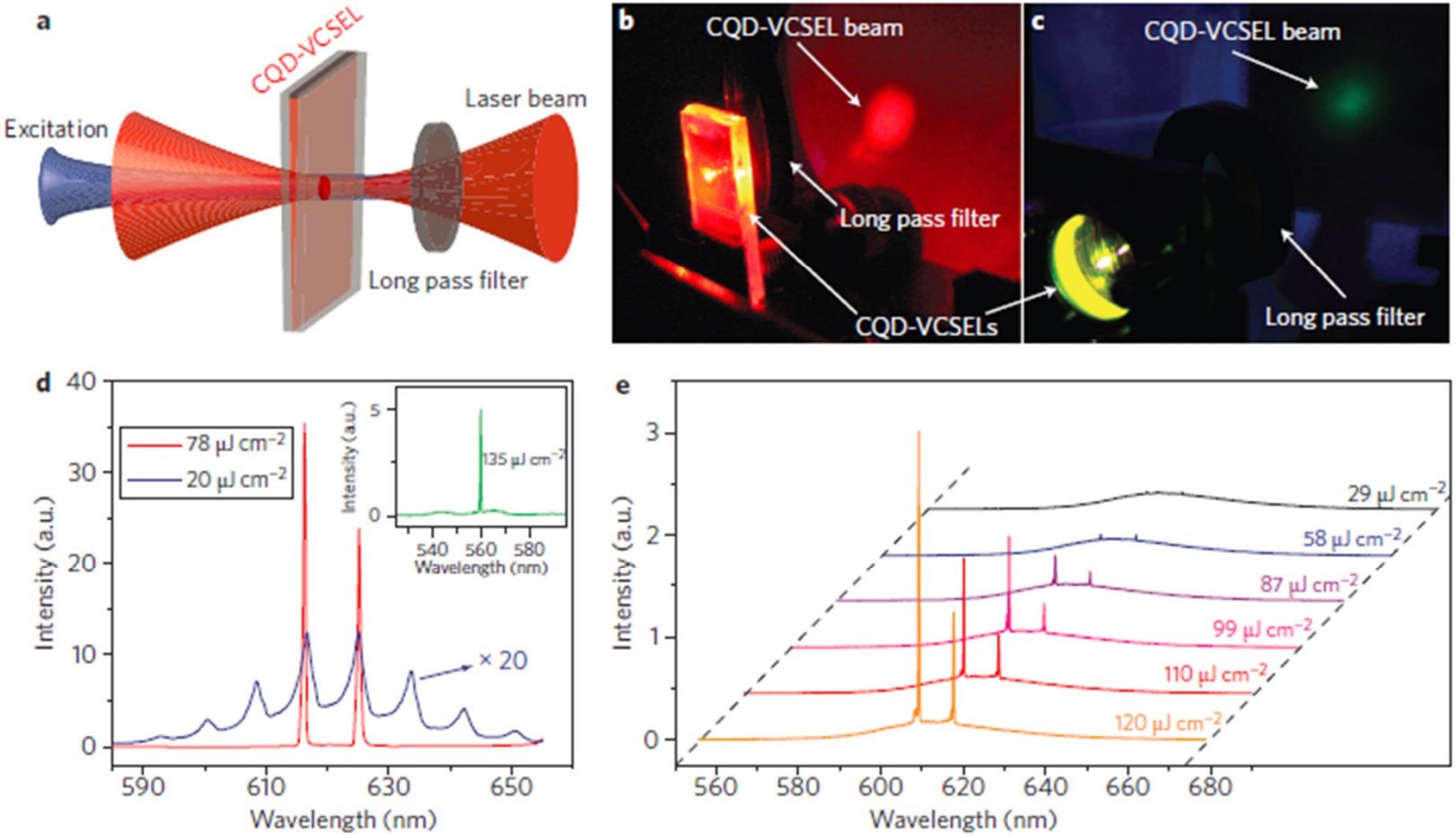
Epitaxial quantum dot lasers: thermal escape (T_0 100 K). Here true strong confinement and thus T-independent SE.

I. Moreels et al., *Adv. Mater.*, **24**, OP231-OP235 (2012)

Multicolor quantum dot single-exciton lasing

CdSe/CdZnS core/shells:





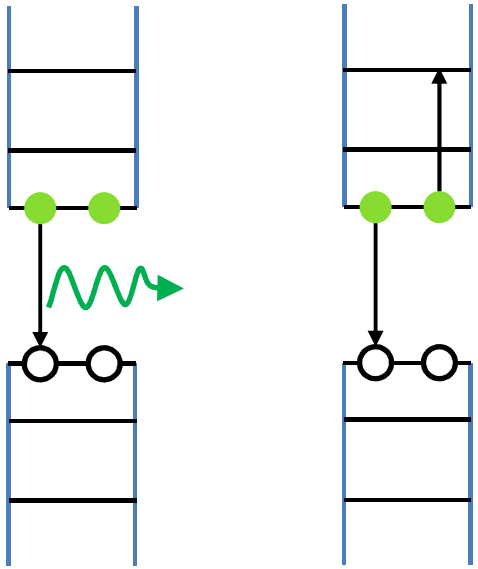
C. Dang et al., *Nature nanotech.* **7**, 335 (2012)

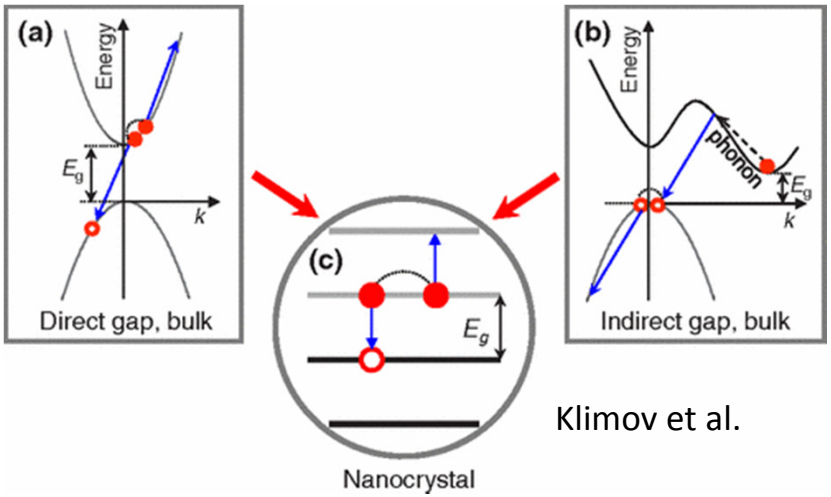
Red, green, and blue lasing achieved by changing Qdot size

Practical demonstrations not yet achieved for Qdot lasers

Radiative recombination
~10 ns

Auger recombination
< 100 ps





Klimov et al.

Amplified femtosecond laser



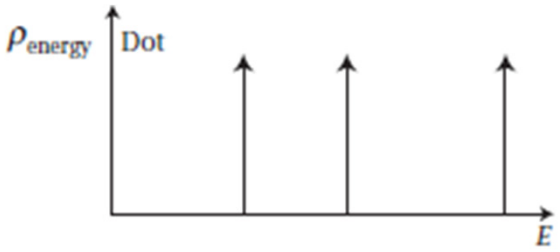


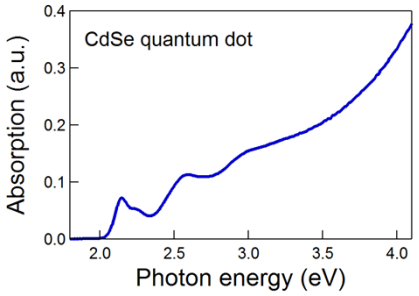
For realistic consumer applications, cw-pumped lasing is required!

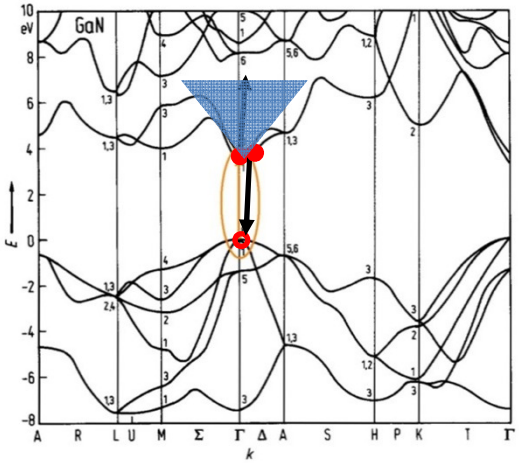
Another solution: change geometry

$$E_0 = \frac{3\hbar^2\pi^2}{2m_e L^2}$$

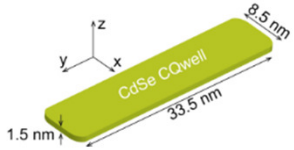


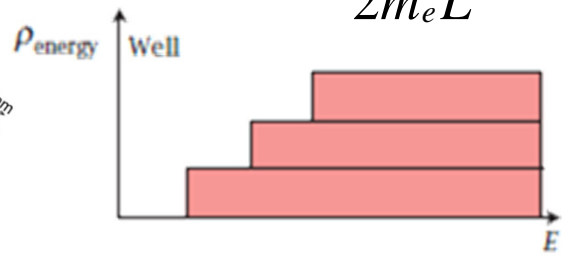


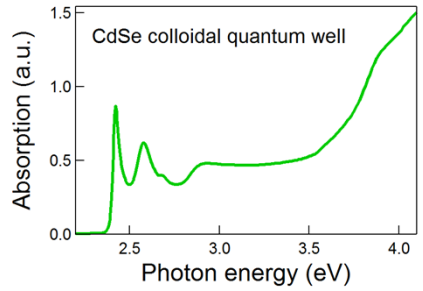


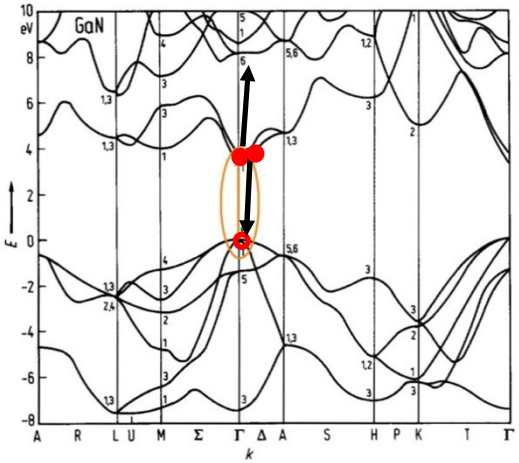


$$E_0 = \frac{\hbar^2\pi^2}{2m_e L^2}$$

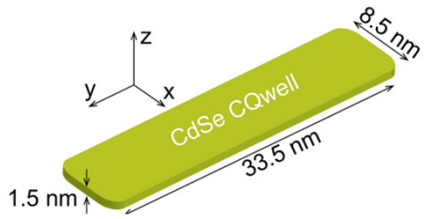


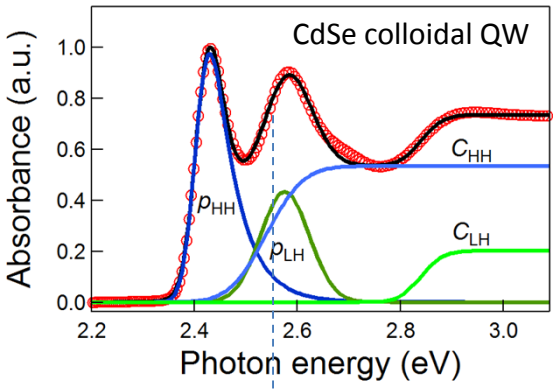




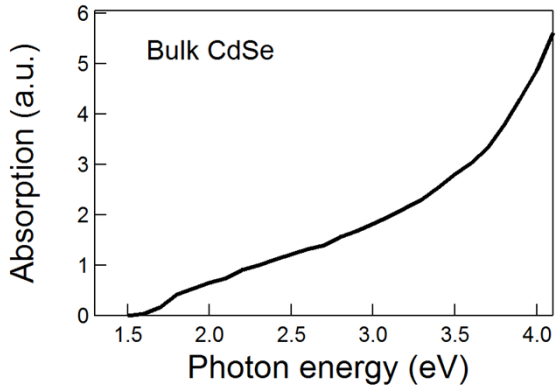


Properties of colloidal quantum wells



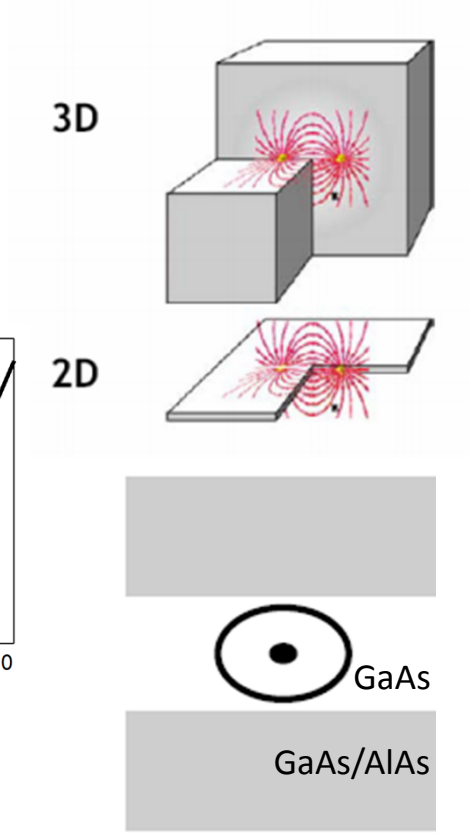


$$E_b^X = 132 \text{ meV}$$



$$E_b^X = 15 \text{ meV}$$

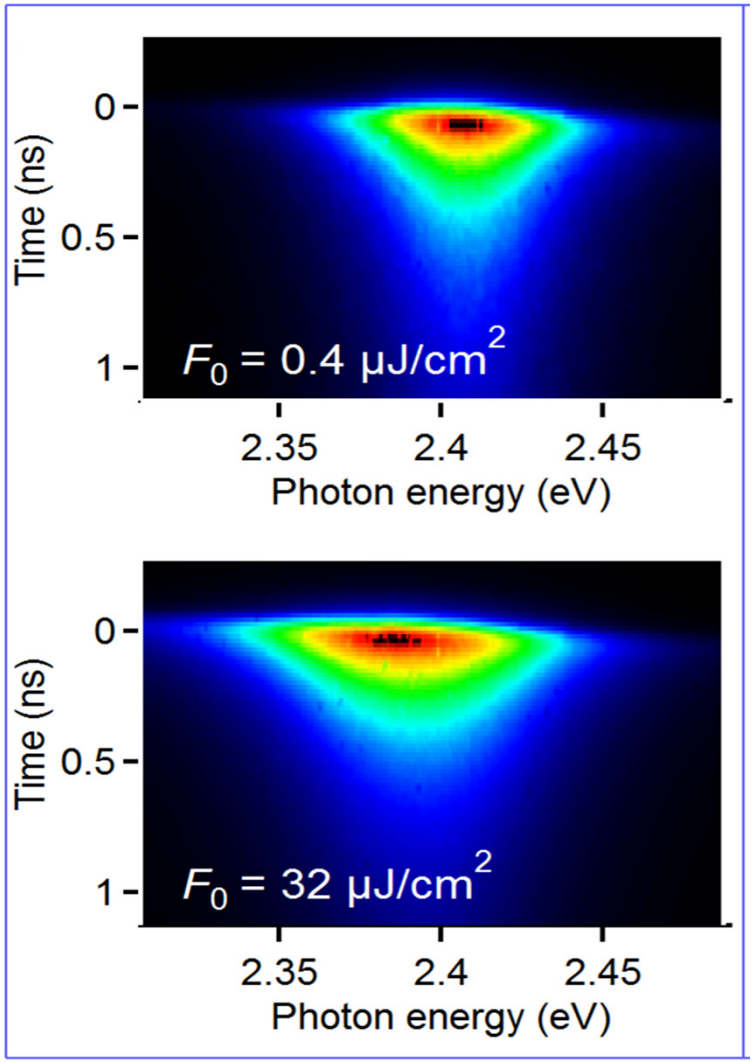




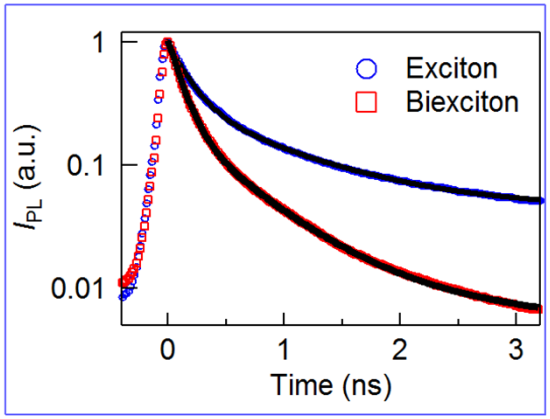
$$E_b^{2D} \approx 9E_b^{3D}$$

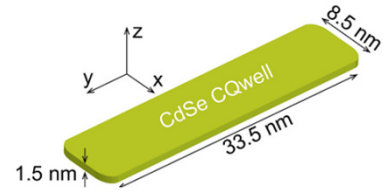
Qwell absorption model: Ali Naeem et al., arXiv:1403.7798 (2014)

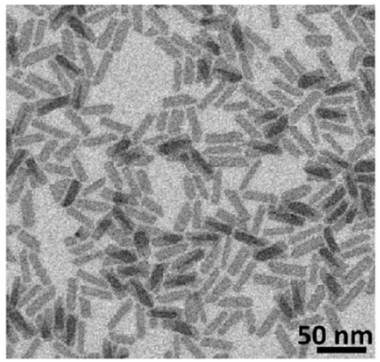
Properties of colloidal quantum wells



Exciton lifetime: **440 ps**
Biexciton lifetime: **125 ps**







- 1D Quantum confinement implies strict selection rules
- In-plane delocalization implies fast exciton recombination rate (giant oscillator strength transition).
- Strongly suppressed Auger recombination in 2D CQwells.

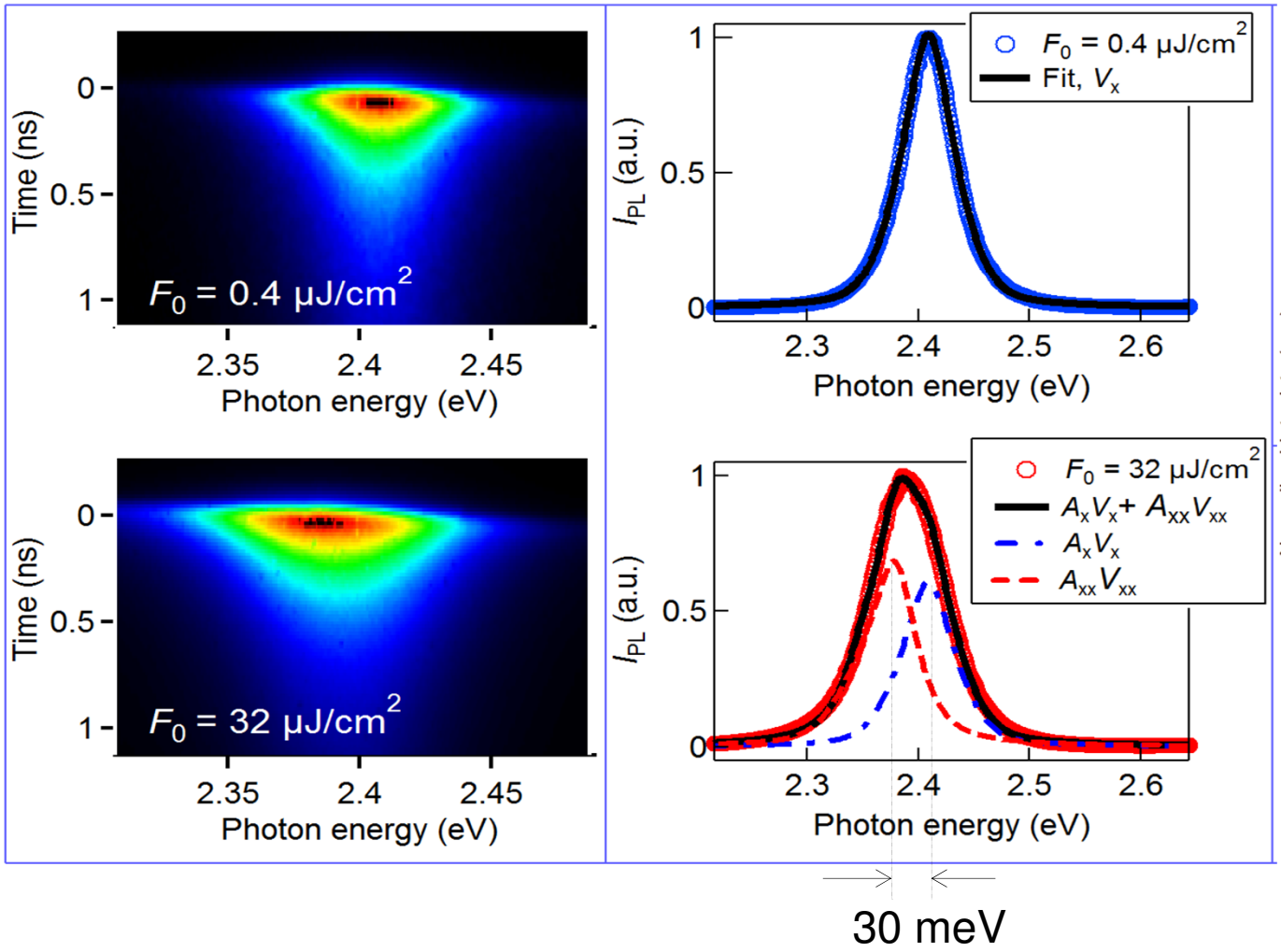
Increased exciton coherence volume → Reduced Auger

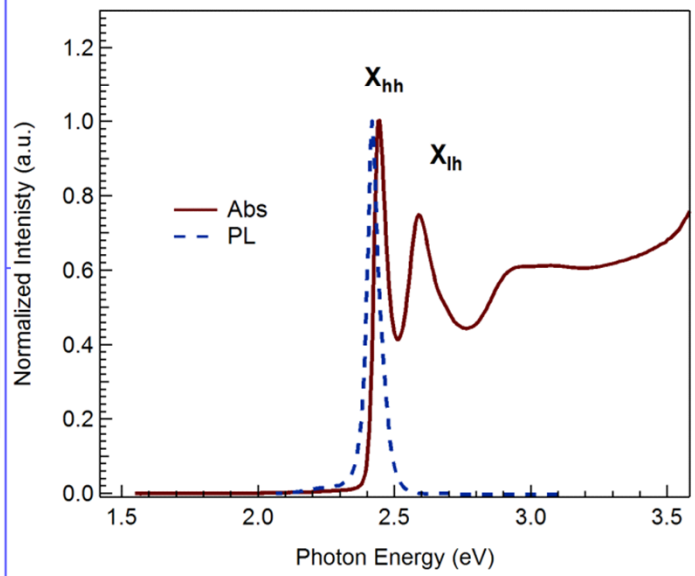
$$\tau_R^{-1} > \tau_{Auger}^{-1}$$

Giant Oscillator Strength: $F_{2D} = Nf_{2D} = f_0 \times 2A / \pi a_0^2$

J. Grim et al. *Nature nanotech.* **9**, 891–895 (2014)

Biexciton binding energy

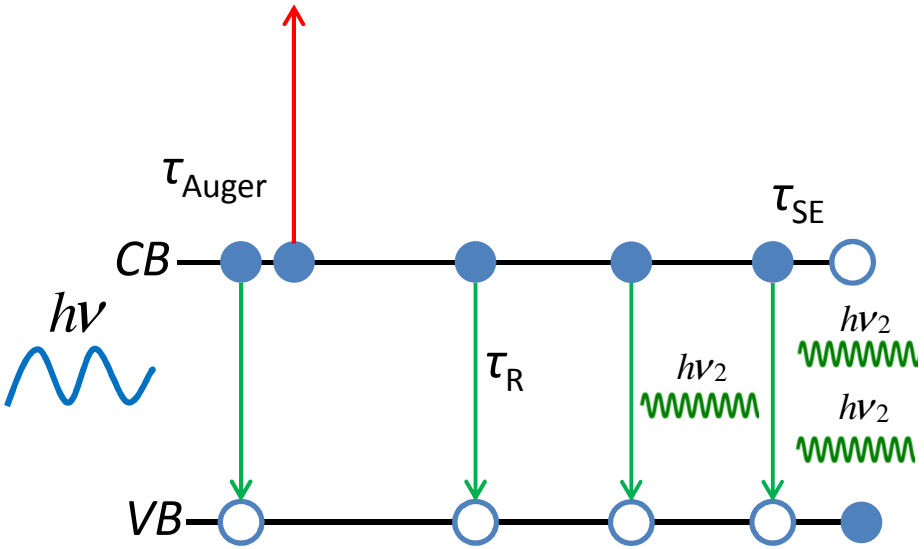




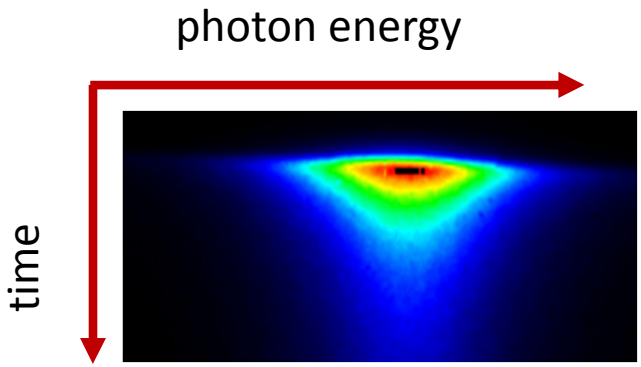
- Strong biexciton binding energy of 30meV.
- Biexciton PL outside of the absorption band (FWHM of absorption is ~30meV)
- Stable biexcitons at room temperature.

J. Grim et al. *Nature nanotech.* **9**, 891–895 (2014)

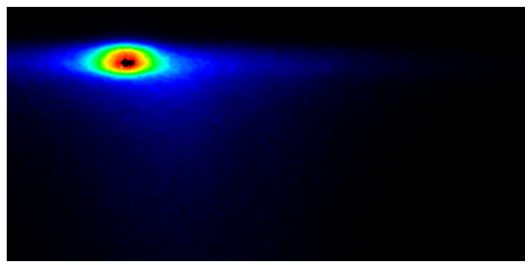
Harnessing confinement: colloidal quantum well lasers



Lasing under femtosecond excitation

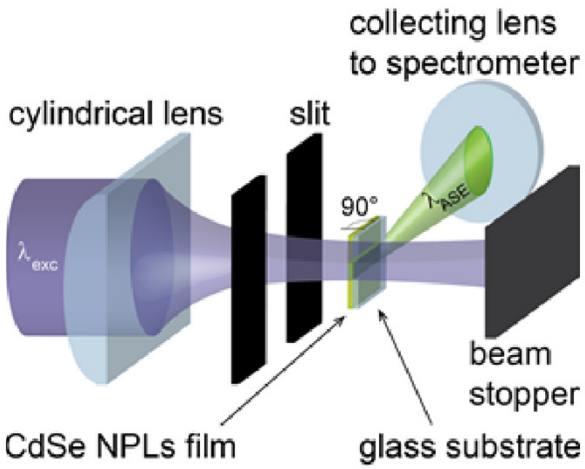


Spontaneous PL
Lifetime > 100 ps

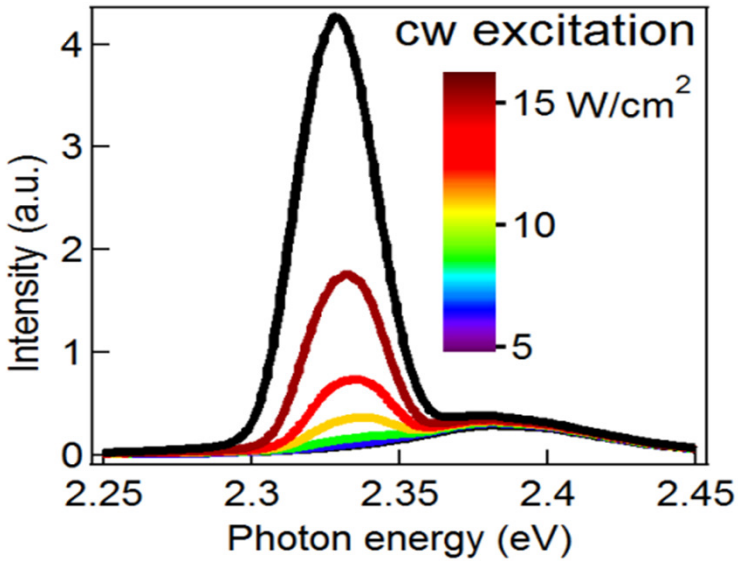


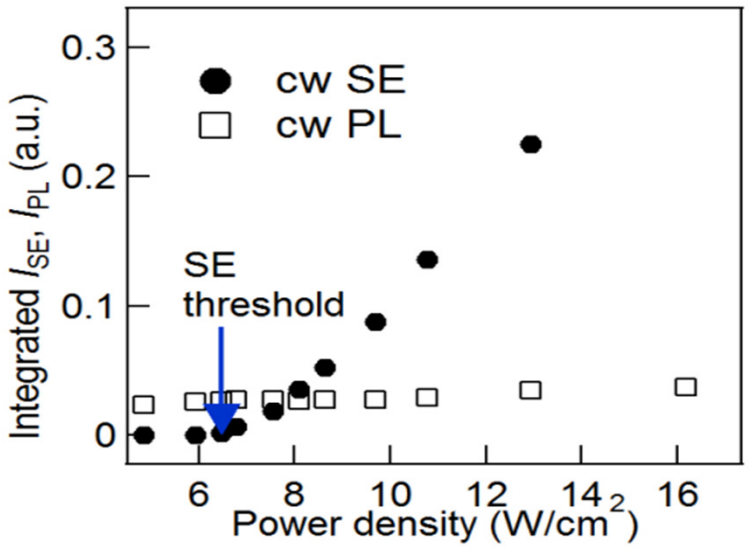
Fast stimulated emission
Lifetime < 1 ps

Harnessing confinement: colloidal quantum well lasers

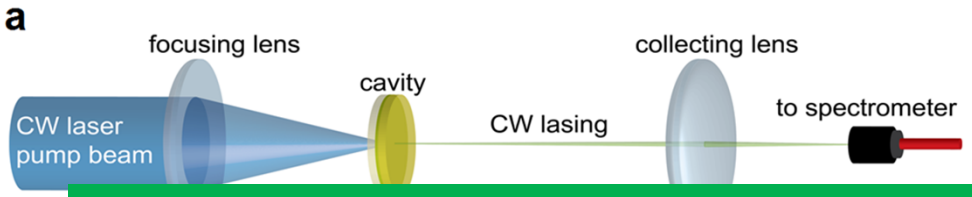


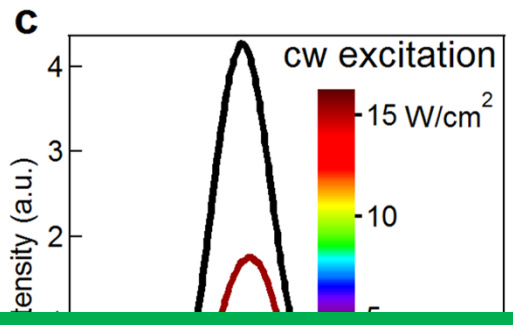
Continuous-wave pumped lasing:
no realistic applications without it



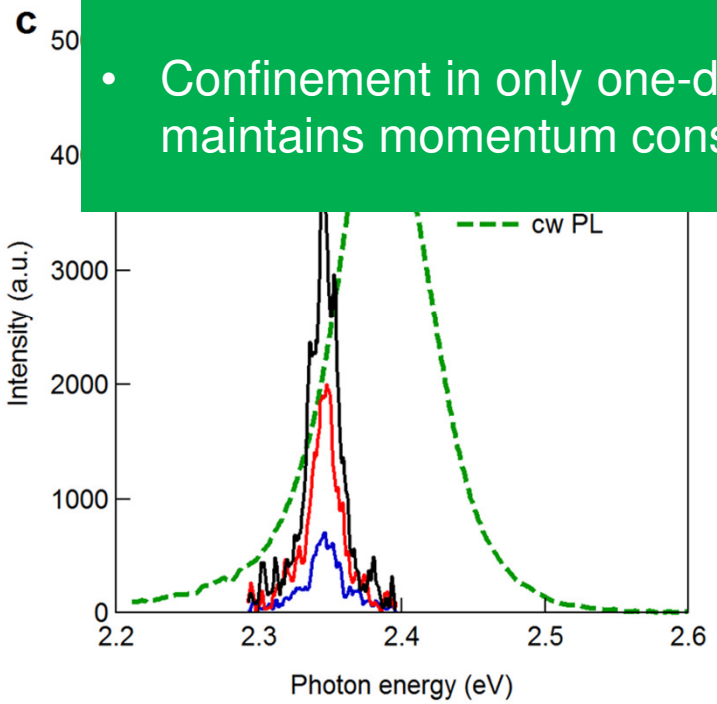


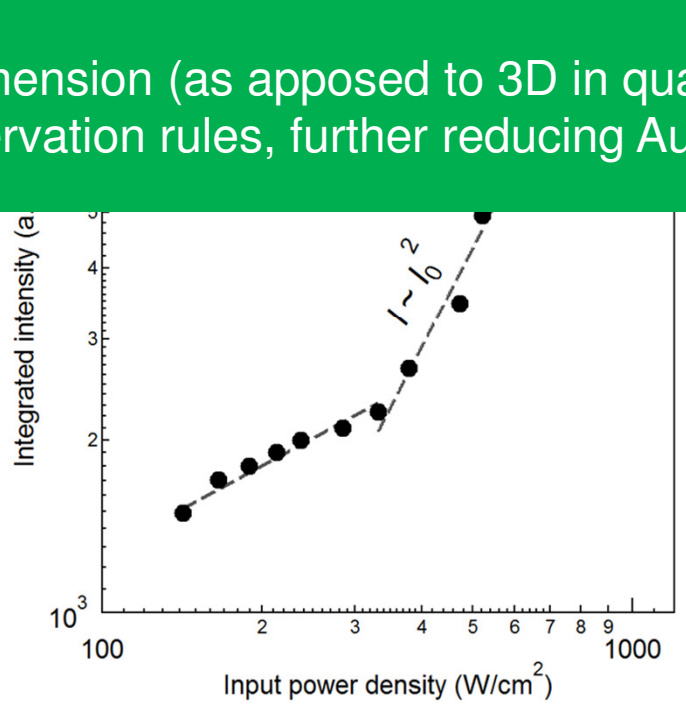
CW-pumped biexciton lasing using CQwells





- 2D nanocrystals have short PL lifetimes, providing a fast radiative channel to compete with nonradiative recombination.
- Confinement in only one-dimension (as apposed to 3D in quantum dots) maintains momentum conservation rules, further reducing Auger recombination.





ASE threshold of 6 μJ/cm^2

cw-excitation:
ASE threshold of 6.5 W/cm^2

CW-pumped lasing
with threshold of 440 W/cm^2

J. Grim et al. *Nature nanotech.* **9**, 891–895 (2014)

Conclusions

- Quantum dot lasers can be realized by engineering heterostructures to spatially separate electrons-hole wavefunction overlap, mitigating the effects of nonradiative Auger recombination.

- Separating carriers reduces the exciton oscillator strength, increasing lasing thresholds and maximum gain.

- CQwells provide a system with minimized Auger and enhanced radiative emission rates, enabling the first demonstration of colloidal nanocrystal continuous-wave pumped lasing.

