Enhancing solar energy harvesting with quantum-dot monolayer on a photonic crystal

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Sandia National Laboratories

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MESA — Microsystems and Engineering Sciences Applications

CINT — Center for Integrated Nanotechnologies — Sandia — Los Alamos

LDRD — LABORATORY DIRECTED RESEARCH & DEVELOPMENT

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Outline

- Motivation
- PV paradigms
- Photonic crystal

- Photonics: Radiative control
  - Solar photovoltaic
  - Solar thermal photovoltaic
  - Near-field thermal photovoltaic

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Motivation

Explore photonic density of states control
for
solar energy applications

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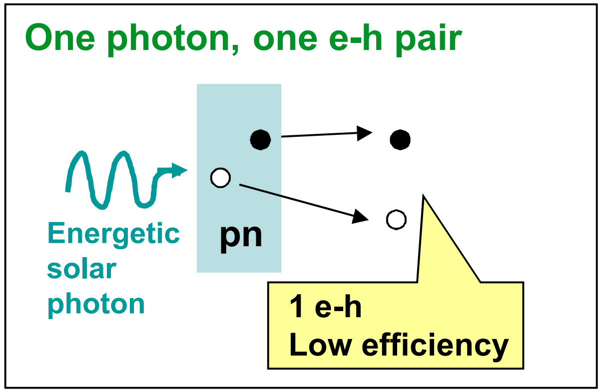
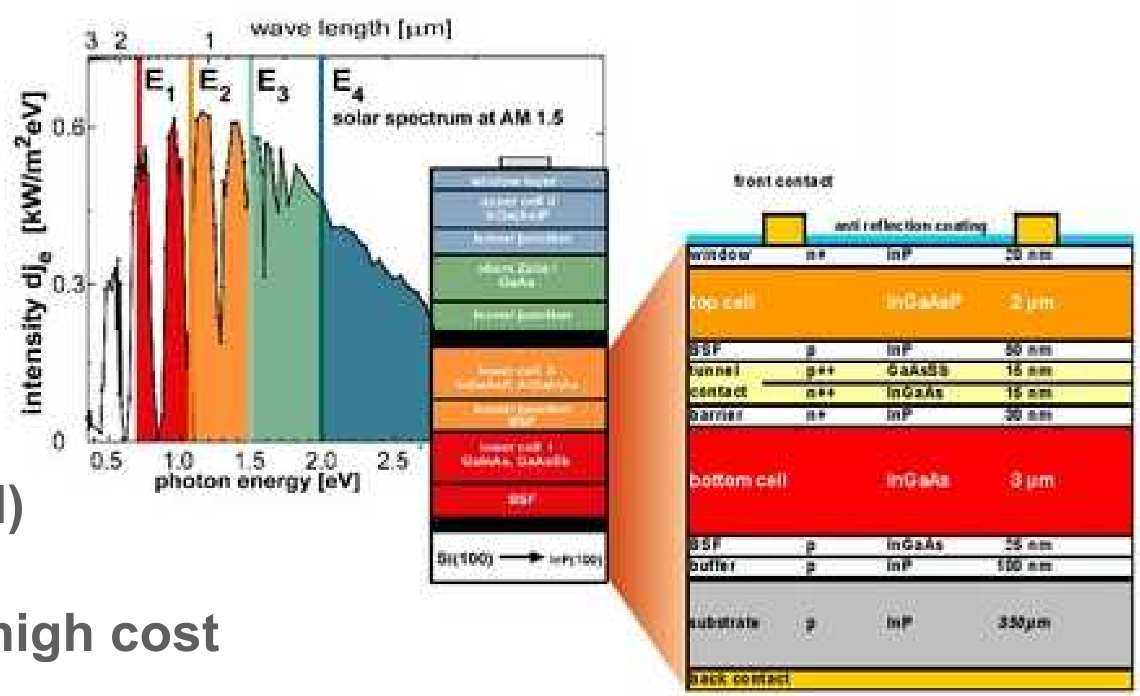
PV cell paradigms

- Holy grail of photovoltaic cell is high performance and low cost

  [Third generation photovoltaics by Martin Green, Springer 2003]



- 1photon produces 1e-h pair
- Efficiency tops at 32% for single junction



- Tandem cells (multi-energy-threshold)
- >40% efficiency
- high performance and high cost

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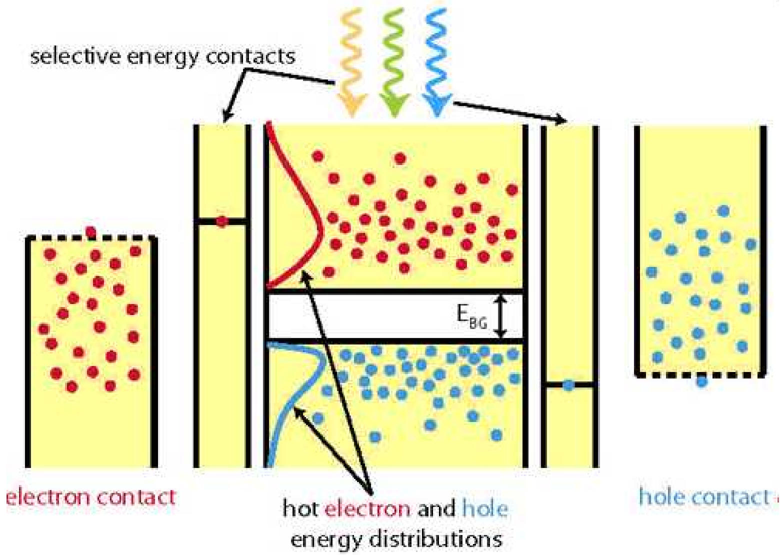
PV cell paradigms

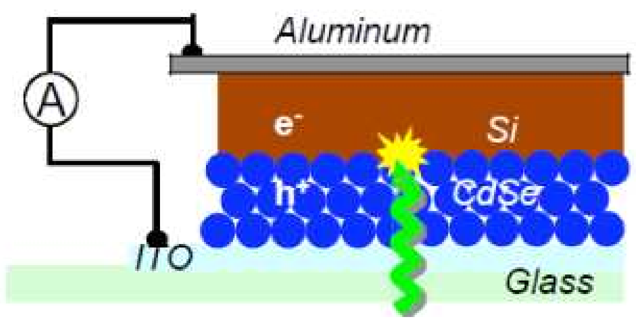
➢ Hot carriers harvesting
➢ Use quantum mechanical resonant tunneling structures

Martin Green



➢ Quantum dot based solar cell
➢ Low cost potential

Klimov



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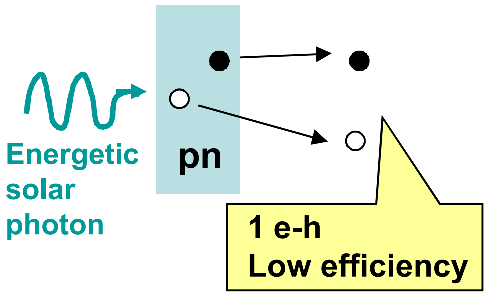
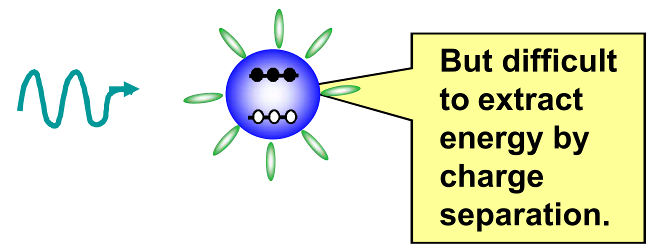
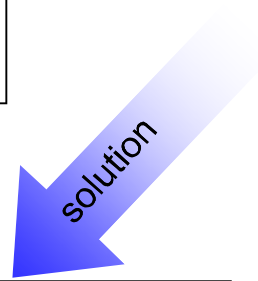
Photonic crystal solution to the energy extraction problem from quantum dots

One photon, one e-h pair



Multi-exciton generation (MEG)





- **Mechanism**

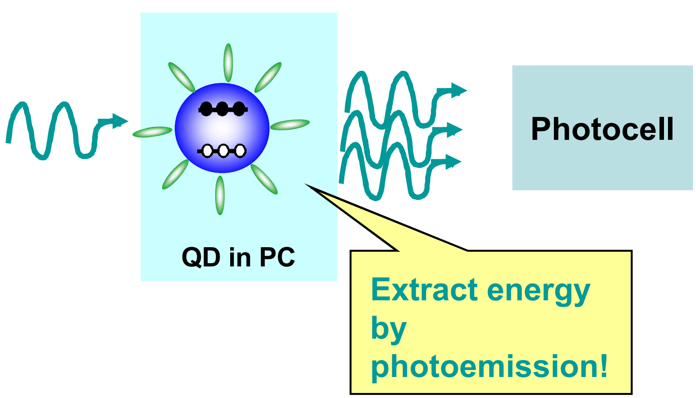
  Impact ionization, inverse Auger effect, quantum confinement effect of multi-e-h interaction..?

- **Impact to solar cell efficiency:**

36-42% for MEG threshold from $3E_g$ to $2E_g$.

[V. I. Klimov, "Detailed-balance power conversion limits of nanocrystal-quantum-dot solar cells in the presence of carrier multiplication," Applied Physics Letters **89** (12) (2006).]

Optical stepdown transformer approach



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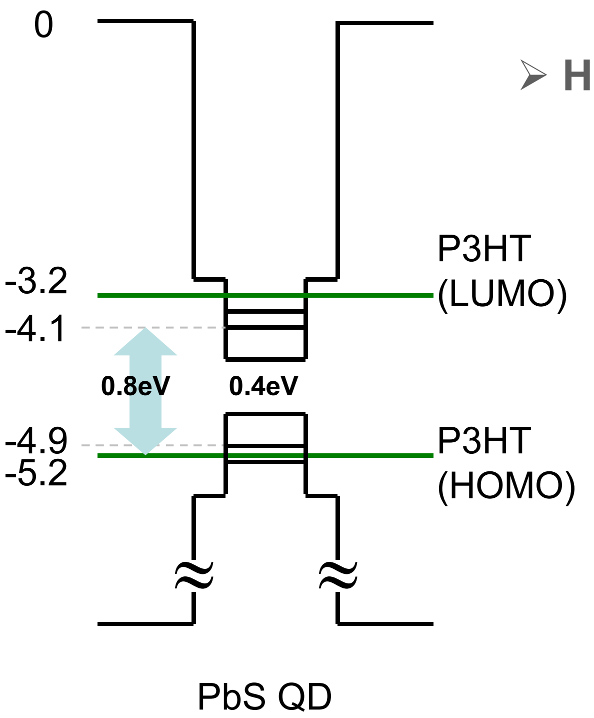
Why PbS quantum dots?



- **Harvest IR part of solar spectrum**
  Low exciton energy (0.8eV), size tunable to 0.6eV.
- **7 excitons generation reported**
- **Good photonic crystals can be fabricated**
- **Good potential solar cell candidate**

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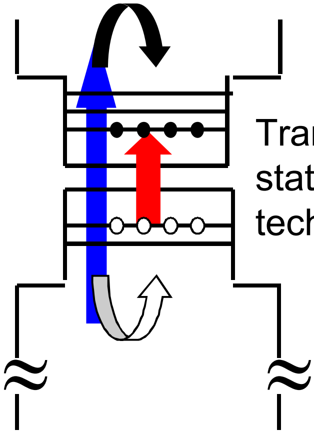
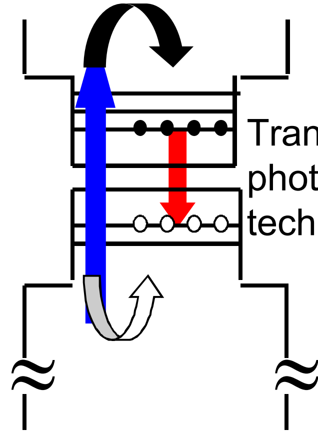
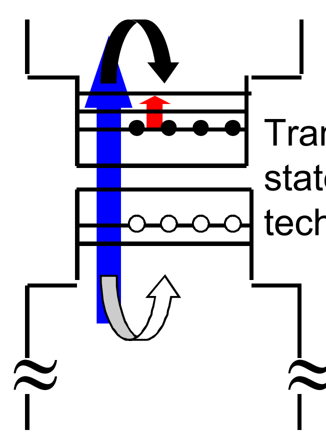
Is MEG real or not?



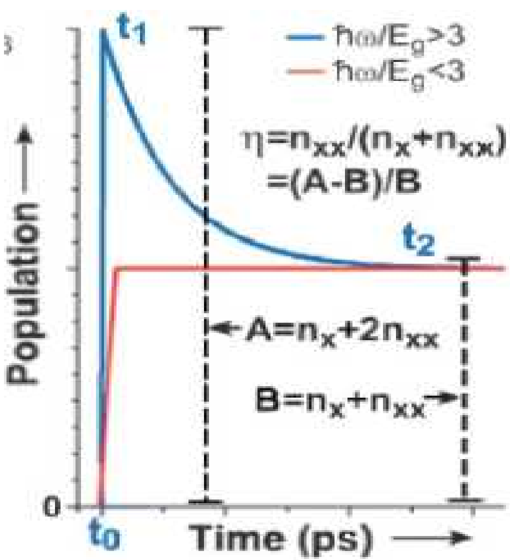
Transient ground state absorption technique



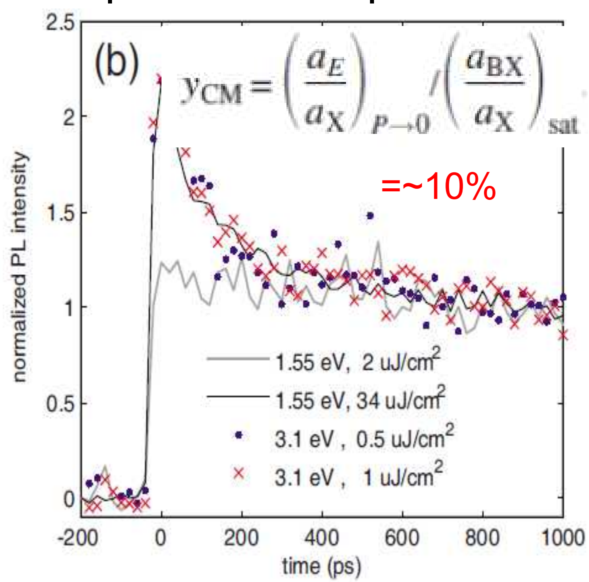
Transient photoluminescence technique



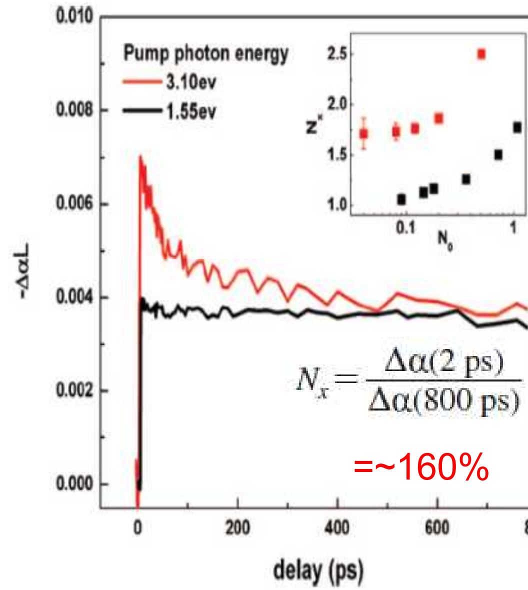
Transient excited state absorption technique



R. D. Schaller and V. I. Klimov, "High Efficiency Carrier Multiplication in PbSe Nanocrystals: Implications for Solar Energy Conversion," Physical Review Letters **92** (18), 186601 (2004).



G. Nair, S. M. Geyer, L. Y. Chang et al., "Carrier multiplication yields in PbS and PbSe nanocrystals measured by transient photoluminescence," Physical Review B **78** (12) (2008).



Minbiao Ji, Sungnam Park, Stephen T. Connor et al., "Efficient Multiple Exciton Generation Observed in Colloidal PbSe Quantum Dots with Temporally and Spectrally Resolved Intraband Excitation," Nano Lett. **9** (3) (2009).

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Planck radiation law and density of states

Planck distribution: Mean energy density within k and k+dk is

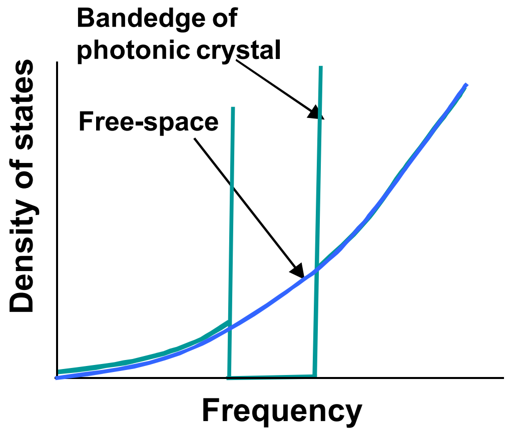
$$u(\omega,T)d\omega = 2\hbar\omega f(k)d^3k = \hbar\omega \frac{\omega^2 d\omega}{\pi^2 c^3} \frac{1}{e^{\hbar\omega/kT}-1} = \frac{8\pi hc}{\lambda^5} \frac{d\lambda}{e^{hc/\lambda kT}-1}$$

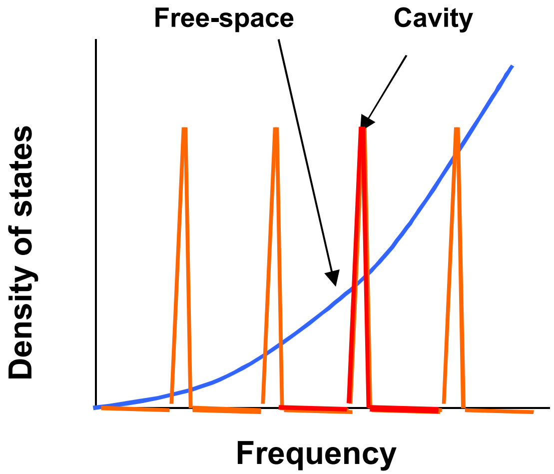
Free space density of states – no. of modes per unit frequency per unit volume

Mean photon number

In photonic crystal, size of void are less than the characteristic wavelength dimension

Non-free-space density of states





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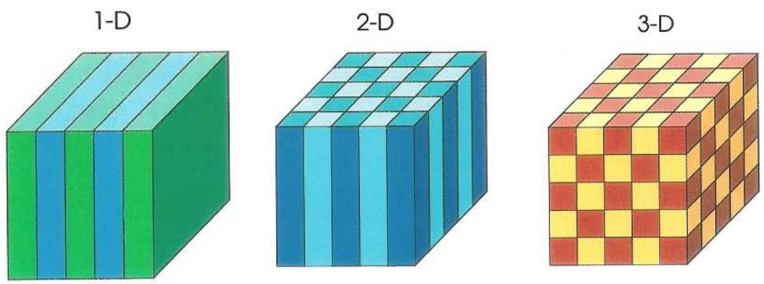
Photonic crystals

Photonic crystal: In this context: a~λ

- A periodic array of electromagnetic scattering centers **with period comparable to wavelength** results in the creation of regions of forbidden propagation commonly termed "spectral gaps" or "band-gaps"
- Bragg condition alone is insufficient to create an omni-directional bandgap.
- Threshold value of ε(r) is also needed.



Principle of PBG Operation:

An electromagnetic wave passing throughout an array of periodic scatterers will undergo destructive interference for certain combinations of wave-vectors at certain frequencies, thus forbidding propagation for such wave-vectors at these frequencies.

$$\left[\nabla \times \left(\frac{1}{\varepsilon(\vec{r})} \nabla \times\right)\right] \vec{H}(\vec{r}) = \left(\frac{\omega}{c}\right)^2 \vec{H}(\vec{r})$$

Metamaterial:

A metamaterial (or meta-material) is a material which gains its properties from its structure rather than directly from its composition. To distinguish metamaterials from other composite materials, the *metamaterial* label is usually used for a material which has unusual properties.

In general: a<< λ

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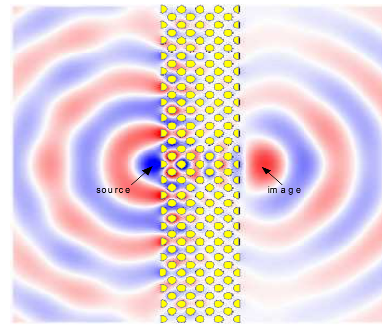
Photonic crystal bandstructure anatomy

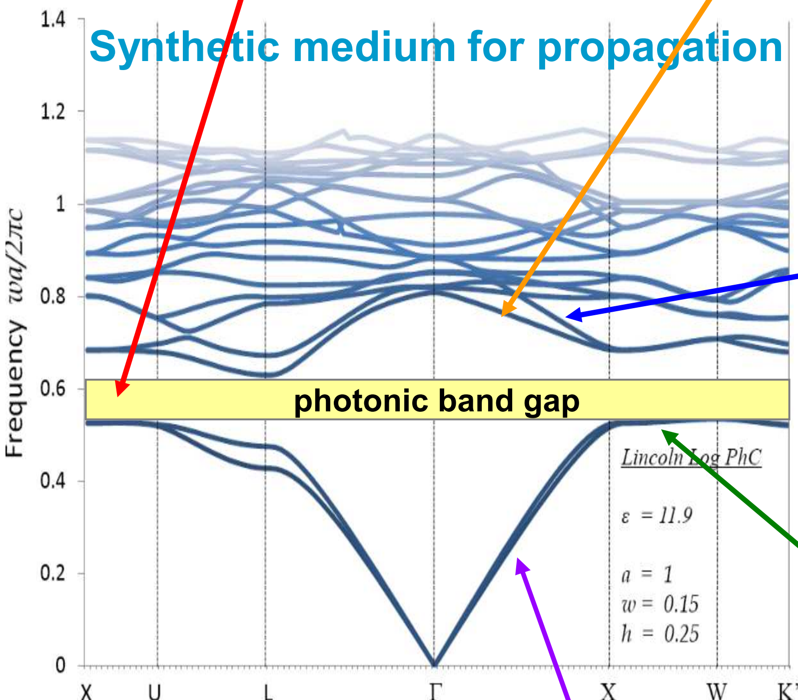
Bandgap region
- Inhibit spontaneous emission
- Nanocavity
- Waveguides

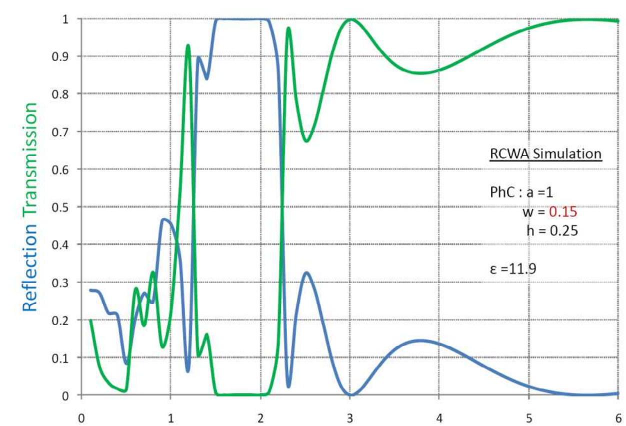
Backwards slope means
- Negative refraction
- Subwavelength imaging



Strong curvature:
- Super-prisms
- Dispersion control

Synthetic medium for propagation





Linear dispersion region
- Optical waveguide and circuits

$d\omega/dk \rightarrow 0$:
- Slow light,
- High photon density of states
- Enhance spontaneous emission

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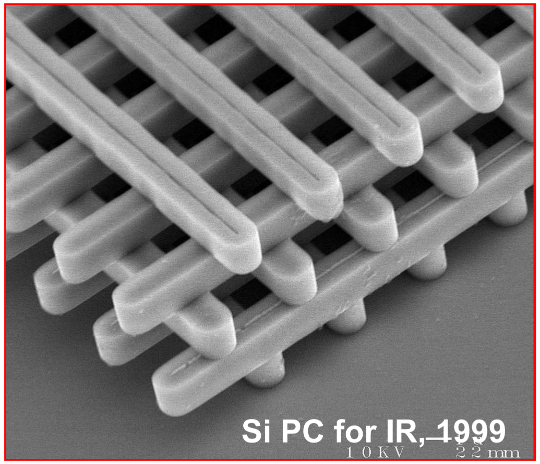
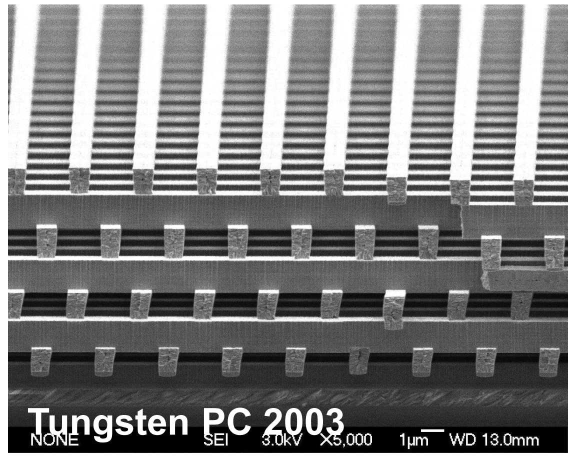
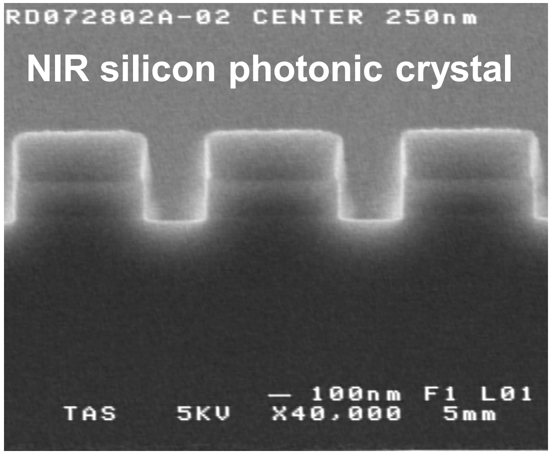
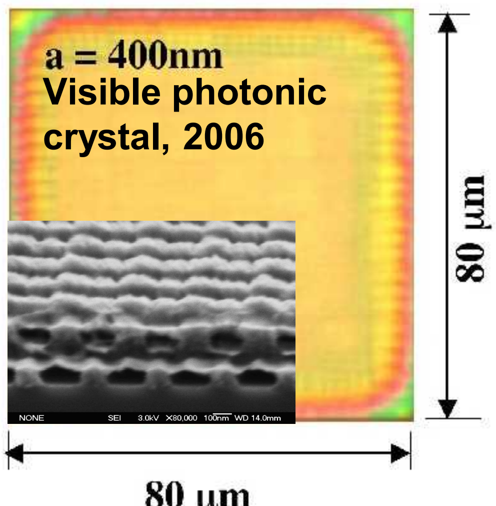
Photonic Crystal Fabrication at SNL

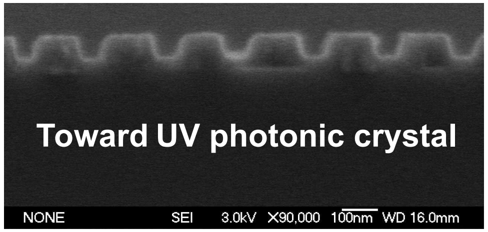
- Enabling fabrication technologies - Low temperature, low stress CMOS compatible and large volume fabrication.

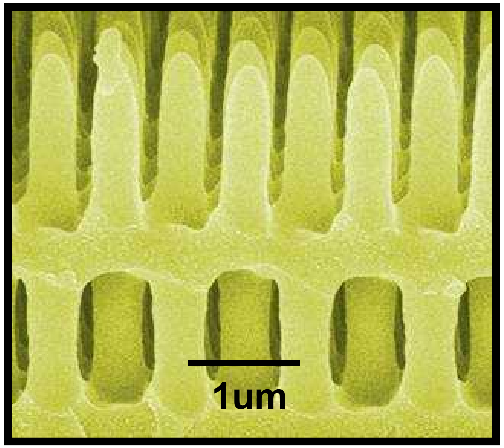












- Other fabrication techniques: proximity field lithography, DXRL, EBL.

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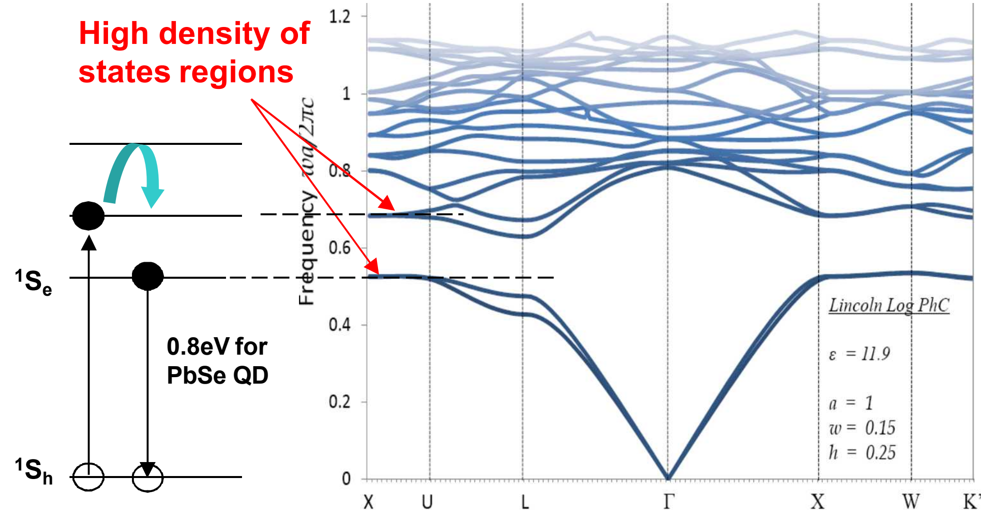
Radiative properties control by photonic crystals: Band edge effect

Band edge effect

Slower group velocity means high density of states which increases absorption coefficient.



Out-compete non-radiative process

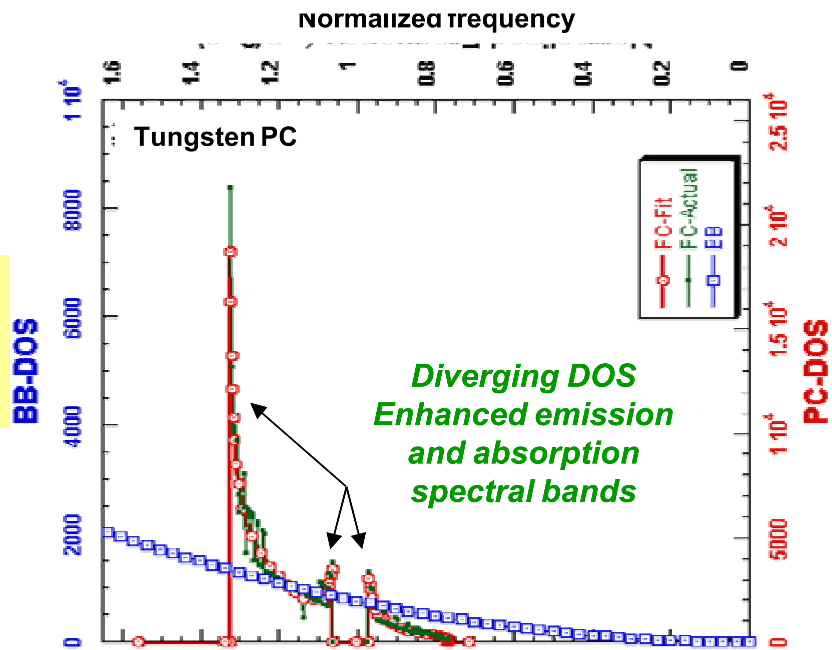
Radiative transition rate

$$\Gamma_{eg} = \frac{2\pi}{\hbar^2}\sum_{k,n}\left|\left\langle g,1_{k,n}\left|\vec{d}\cdot\vec{E}(\vec{x}_0)\right|e,vac\right\rangle\right|^2\delta\left(\omega_{k,n}-\omega_0\right)$$

$$\Gamma_{eg} = \frac{\pi\omega_0\,|\vec{d}\,|^2}{\varepsilon_0\hbar}\rho\left(\vec{x}_0,\frac{\vec{d}}{|\,d\,|},\omega_0\right)$$

Photonic density of states engineering

$$\hat{\vec{E}}(\vec{x}_0) = \sum_{k,n}\sqrt{\frac{\hbar\omega_{k,n}}{2\varepsilon_0 V}}\left(\vec{E}_{\vec{k},n}(x_0)\,\hat{a}_{\vec{k},n} + h.c.\right)$$



I. El-Kady et. al. "Emission from an active photonic crystal", Phys. Rev. B, vol. 72, 195110 (2005)

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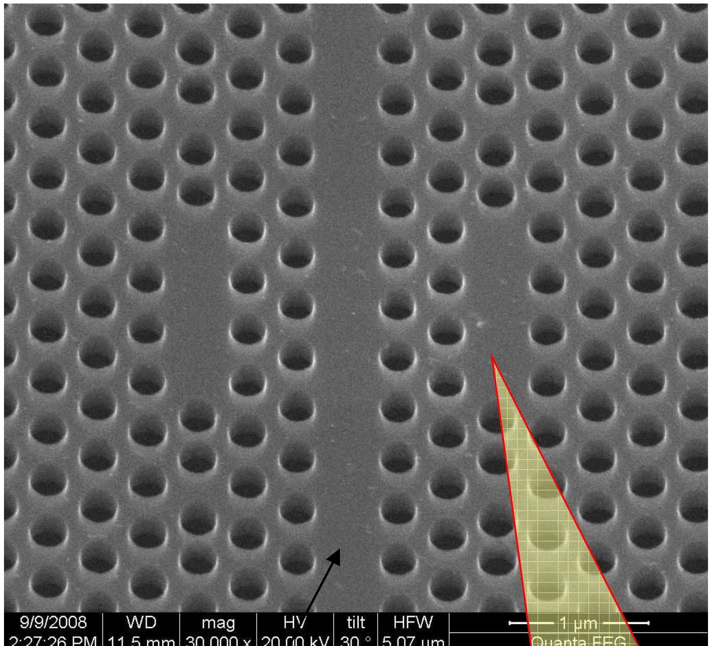
Radiative properties control by photonic crystals: Purcell effect



Waveguide

Purcell enhancement factor

$$P = \frac{3}{4\pi^2}\left(\frac{\lambda}{n}\right)^3 \frac{Q}{V}$$

Spontaneous emission enhancement factor

$$\frac{\tau}{\tau_0} = \left(\frac{3\lambda_C^3 Q}{4\pi^2 n^3 V_m}\right) \times \zeta_r \times \left(\frac{\Delta\lambda_C^2}{\Delta\lambda_C^2 + 4(\lambda_C - \lambda_X)^2}\right) + f,$$

Enhancement factor

Purcell factor (interaction strength with vacuum field in a small confined volume)

Cavity resonance effect

- **Mode volume is less than $(\lambda/n)^3$.**
- **$Q = 10^6$ demonstrated.**
- **Q of 10^9 is possible.**
- **Purcell factor of 10^5.**

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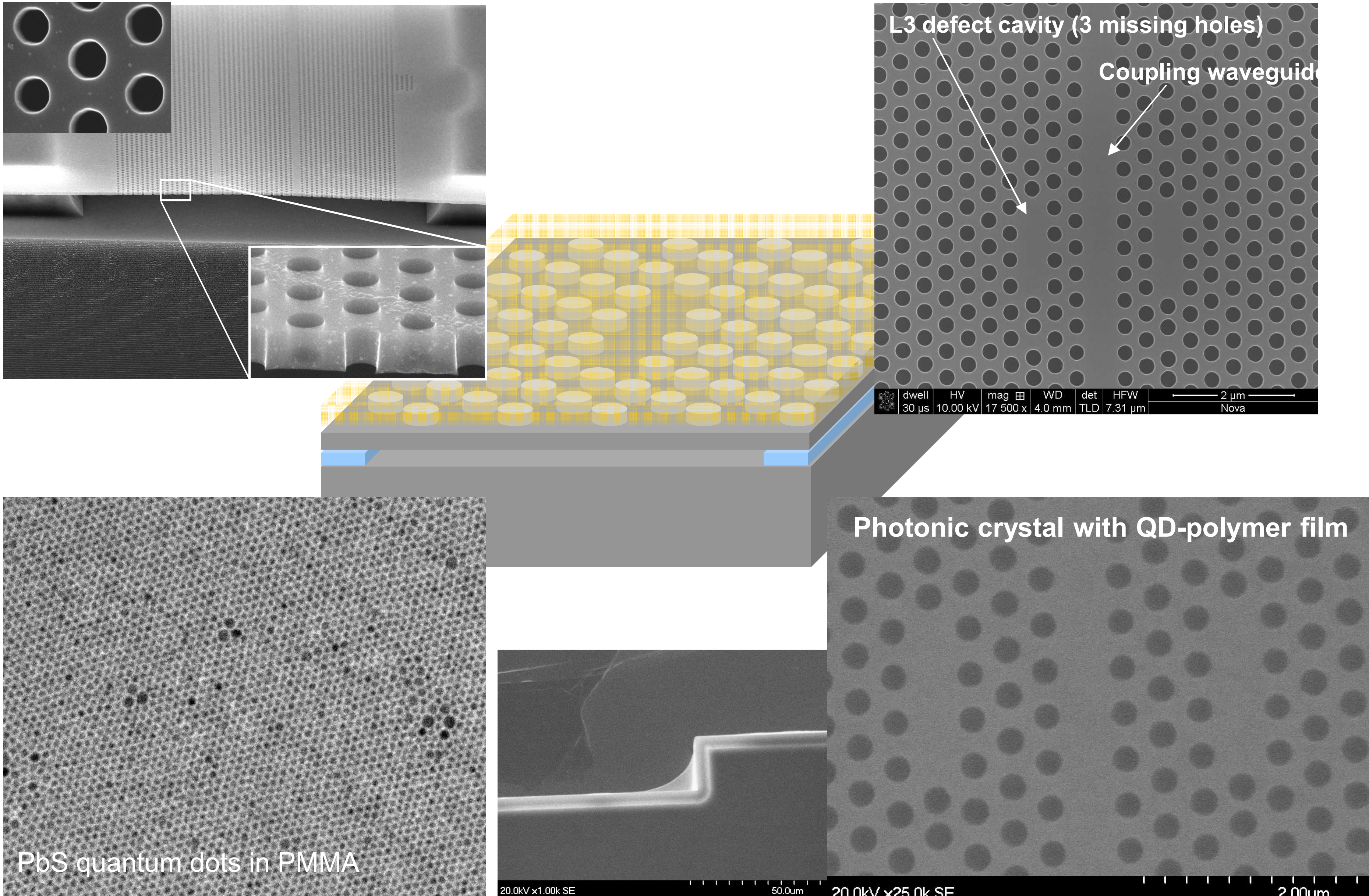
Photonic crystal fabrication



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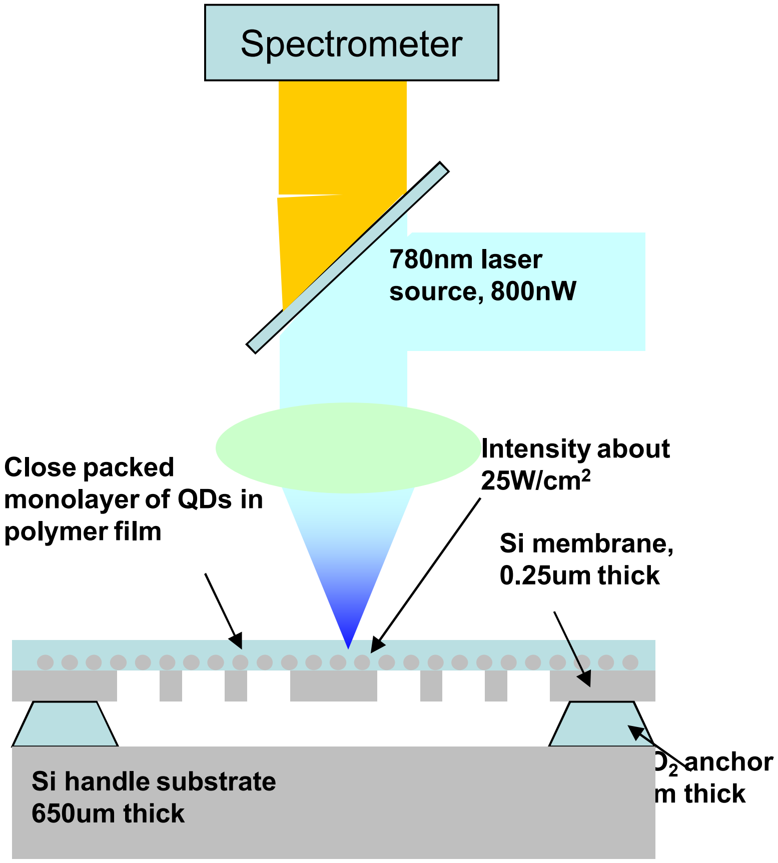
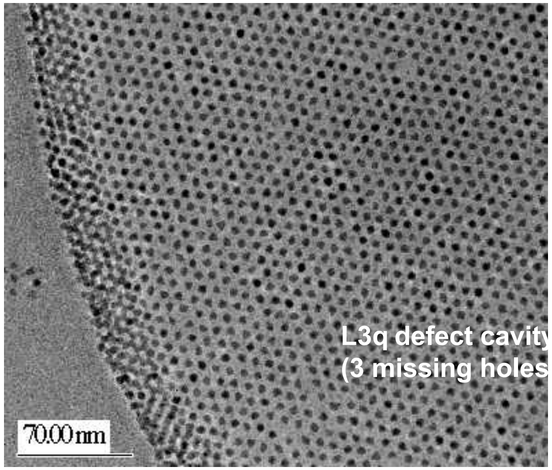
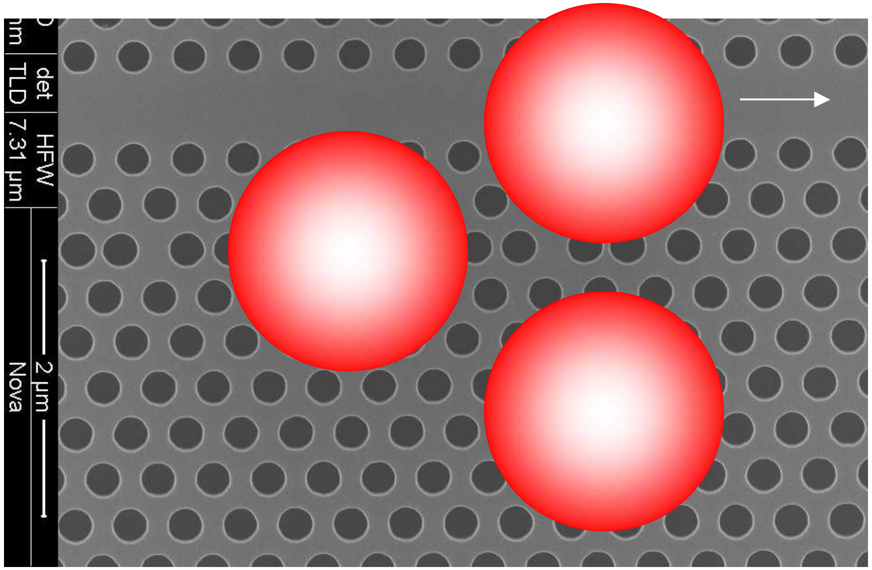
Photoluminescence experiment from Quantum dots



Side view





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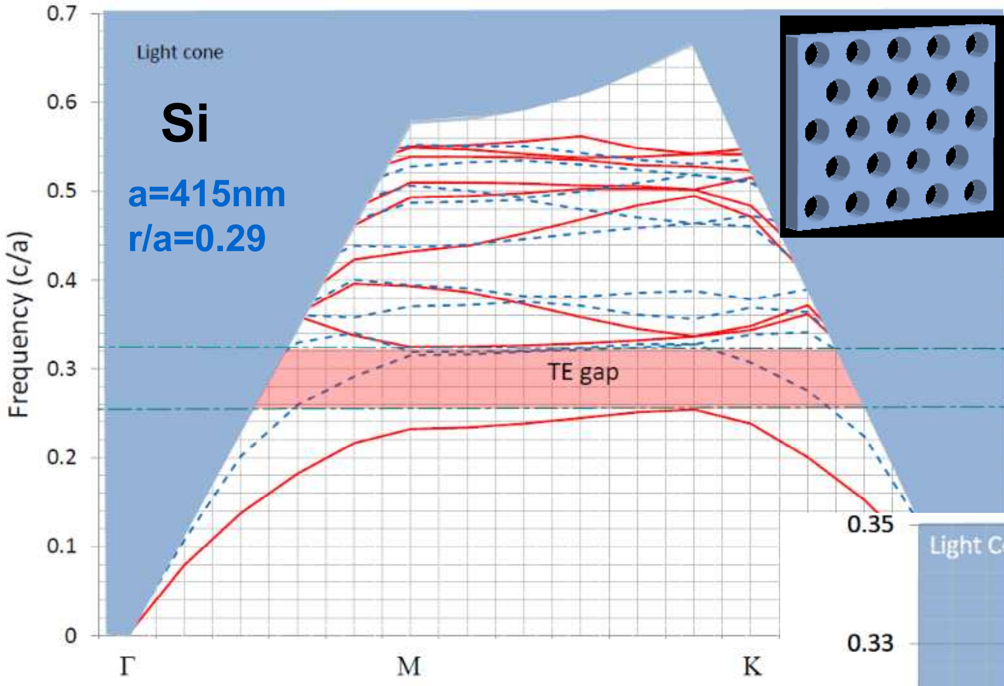
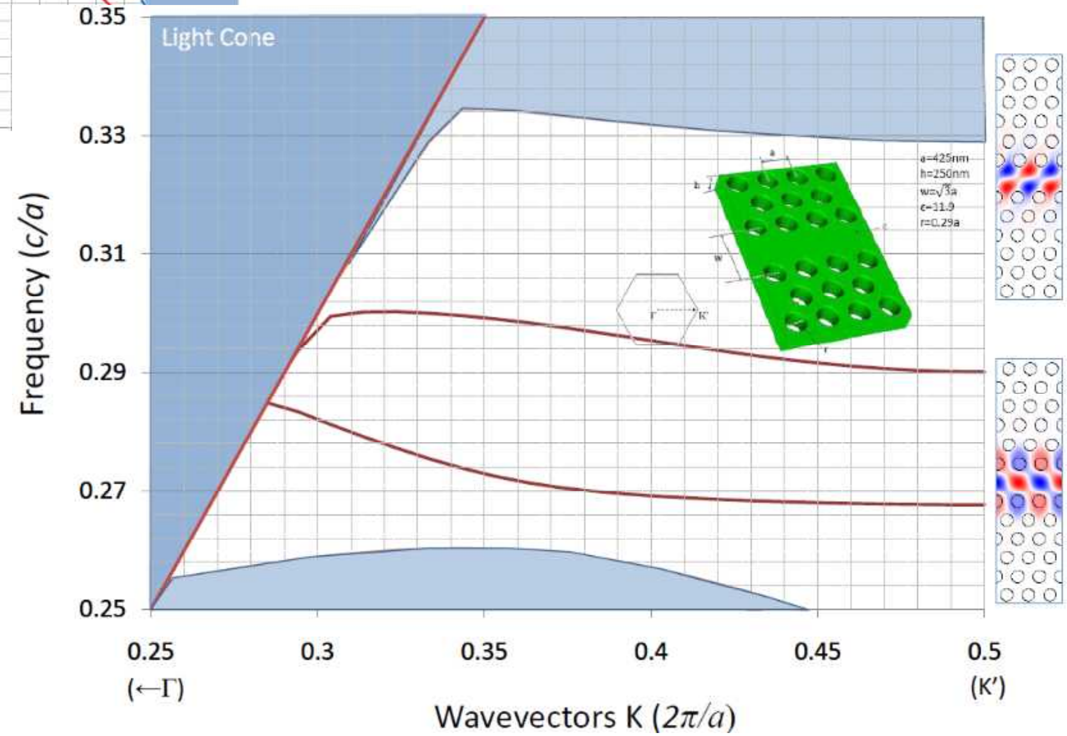
Bandstructure and waveguide defect of air-clad Si photonic crystal





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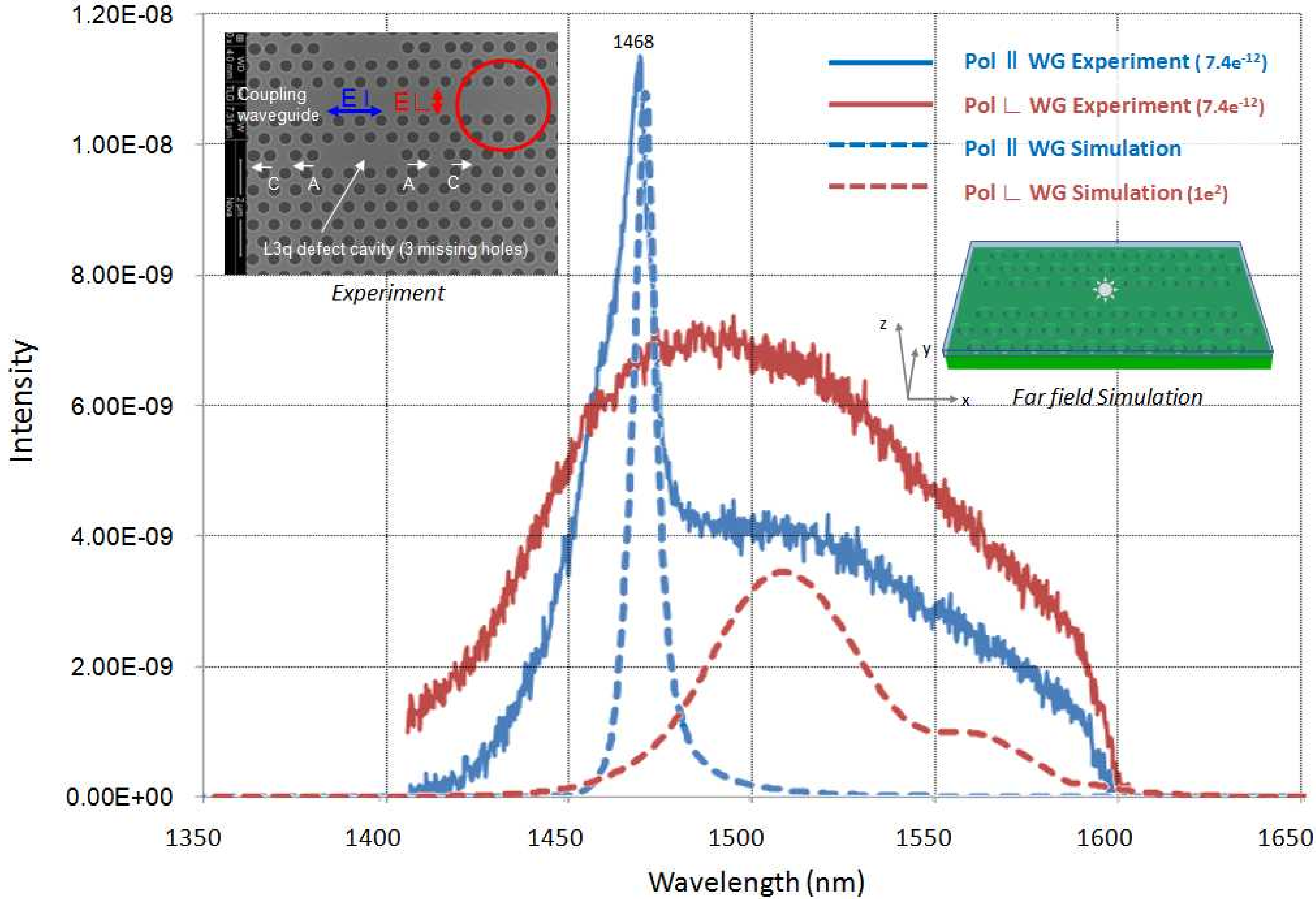
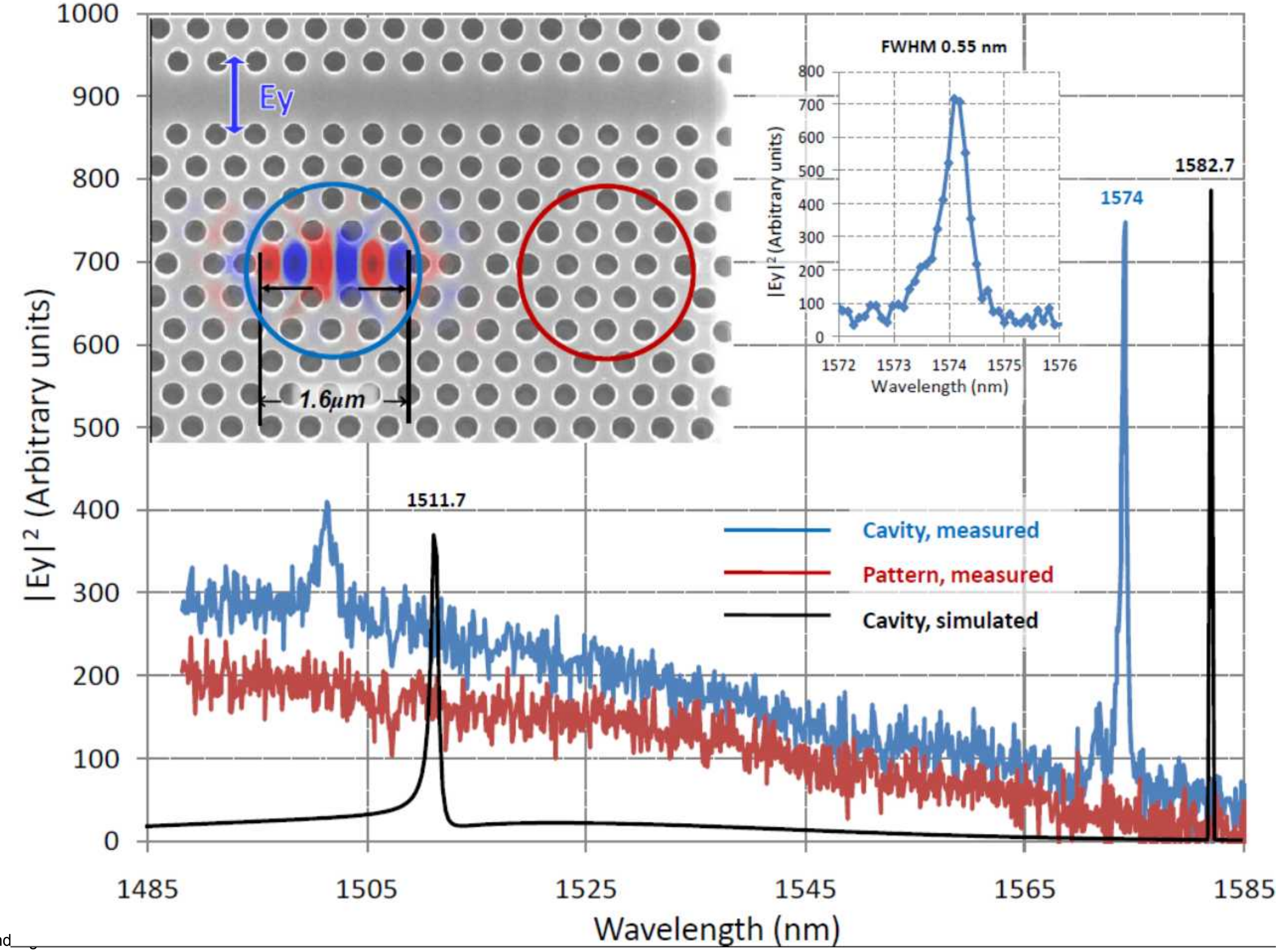
Enhanced Emission from Quantum dots on photonic crystal *waveguide*



Enhanced Emission from Quantum dots on photonic crystal *cavity*



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Enhanced Factor determination

Enhancement factor = $\dfrac{\text{Emission observed from the cavity at resonant frequency}}{\text{Reference emission}}$

What is a good reference?

Emission from the pattern surface,

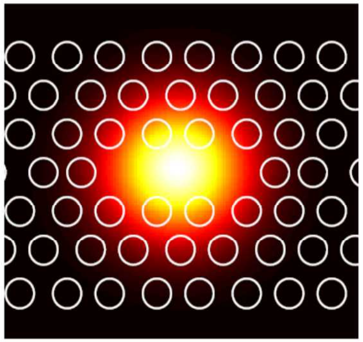
Emission from the cavity but wrong polarization,

What about the contributions from dots that are not in the cavity or at the cavity node?

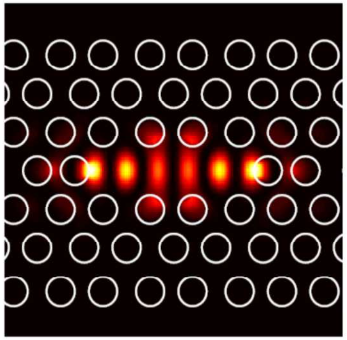
Enhancement factor = $\dfrac{8}{0.157} = 51$

$$\frac{\Gamma_c}{\Gamma_0} = \frac{Q}{V}\left(\frac{3\lambda^3}{4\pi^2 n^3}\right)\left(\frac{E_{surface}}{E_{mode}^{\max}}\right)^2$$ **= 73**

$$Contributing \quad fraction = \frac{\int \left|E_{ex}(x,y,surface)\right|^2 \left|E_{mode}(x,y,surface)\right|^2 dxdy}{\left|E_{mode}^{\max}(x,y,surface)\right|^2 \int \left|E_{ex}(x,y,surface)\right|^2 dxdy} = 0.157$$



Excitation field
Gaussian: $c = (0\ 0)$
$\sigma = 0.8\ \mu m$



Resonance mode :
Polarization = Ey (|_)
Freq = 0.262
Wavelength = 1582.7

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Summary

- We have shown photonic crystal cavity can enhance spontaneous emission of quantum dots significantly.
- We hope to use photonic density of states to control relaxation dynamics of carriers in semiconducting materials.

Acknowledgements:
Funded by SNL Lab Directed Research Development (LDRD)
Theory – Weng Chow
Modelling – Ihab El-Kady (SNL) and Bernardo Farfan (UNM student)
QDs in polymer – Jeff Brinker (SNL), Shisheng Xiong (UNM student)
Fabrication – Paul Resnick (SNL)
Measurements – Shisheng Xiong (UNM student) and myself

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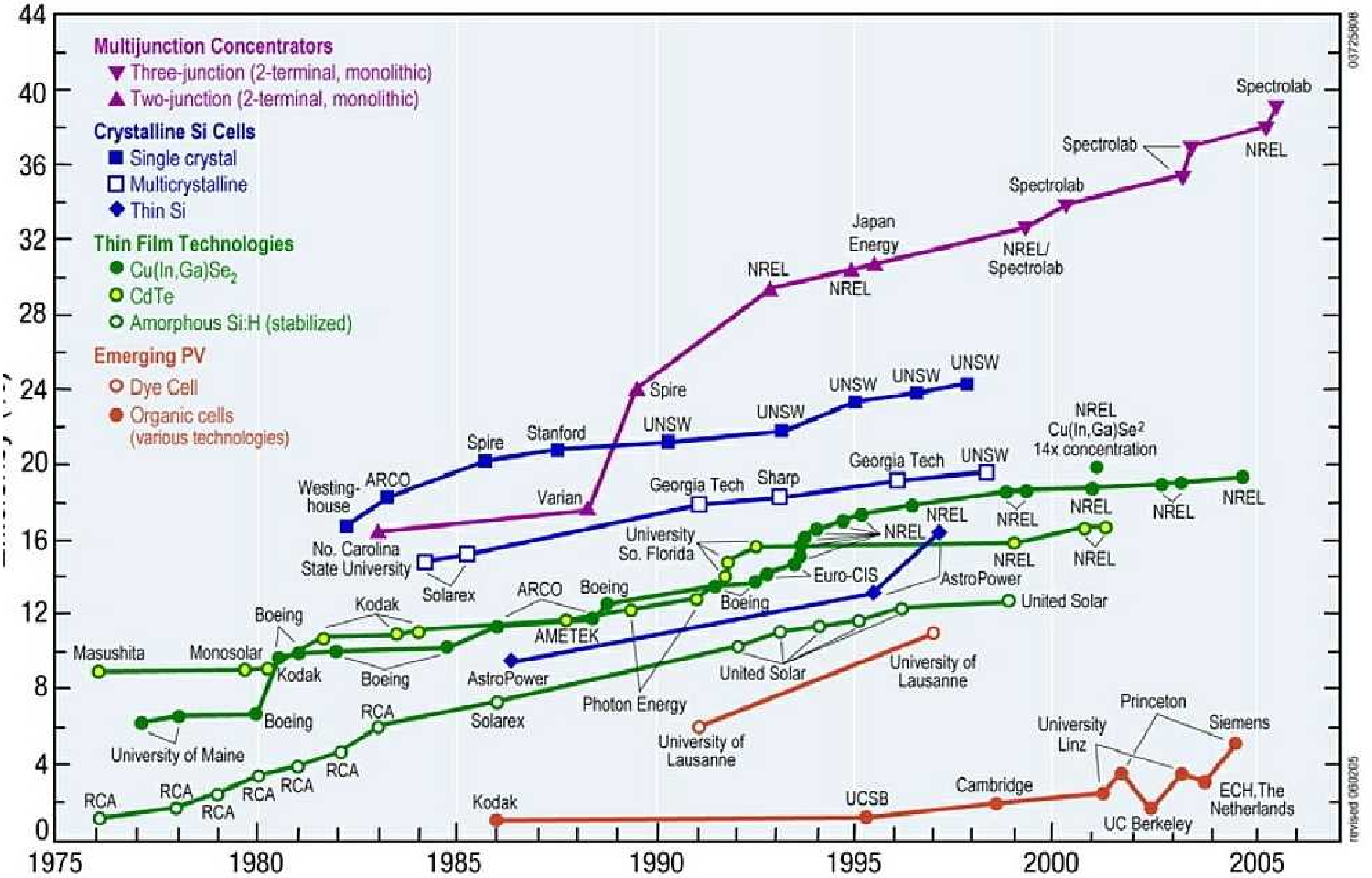

Improvements in solar cell efficiency, by system, from 1976 to 2004

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