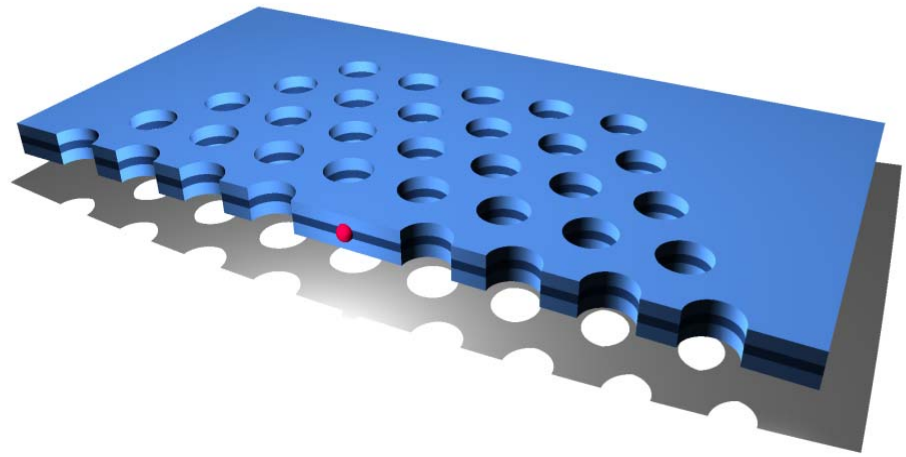
Quasimodes and Purcell factors for leaky optical cavities and plasmonic nanoresonators

S. Hughes

Queen's University, Kingston, Ontario, Canada

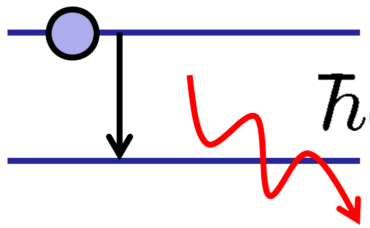


Rongchun Ge, P. T. Kristensen, J.F. Young, NSERC ($)

Kristensen and Hughes, Perspectives, ACS Photonics 1 (2014)

Traditional route to cavity-QED and local field enhancements

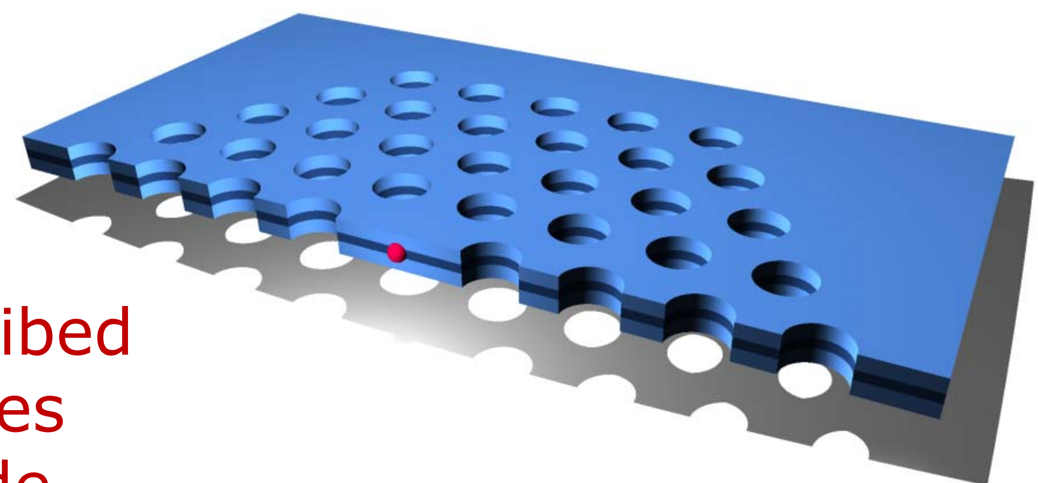
- General scaling for light-matter interactions



$$\hbar\omega \quad \propto \left(\frac{\lambda}{n}\right)^3 \frac{Q}{V}$$

.... Quality factor

.... Volume



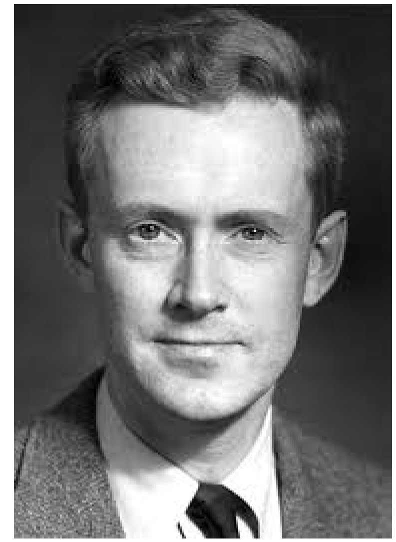
Enhancement described in terms of properties of single cavity mode

Cavity-modified spontaneous emission

B10. Spontaneous Emission Probabilities at Radio Frequencies. E. M. PURCELL, *Harvard University*.—For nuclear magnetic moment transitions at radio frequencies the probability of spontaneous emission, computed from

$$A_\nu = (8\pi\nu^2/c^3)h\nu(8\pi^3\mu^2/3h^2)\ \text{sec.}^{-1},$$

is so small that this process is not effective in bringing a spin system into thermal equilibrium with its surroundings. At 300°K, for $\nu=10^7$ sec.$^{-1}$, $\mu=1$ nuclear magneton, the corresponding relaxation time would be 5×10^{21} seconds! However, for a system coupled to a resonant electrical circuit, the factor $8\pi\nu^2/c^3$ no longer gives correctly the number of radiation oscillators per unit volume, in unit frequency range, there being now *one* oscillator in the frequency range ν/Q associated with the circuit. The spontaneous emission probability is thereby increased, and the relaxation time reduced, by a factor $f=3Q\lambda^3/4\pi^2V$, where V is the volume of the resonator. If a is a dimension

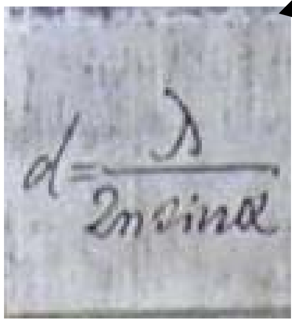


Edward Purcell

E. M. Purcell, Phys. Rev. 69, 681 (1946).

Cavity size limits?

U. Jena

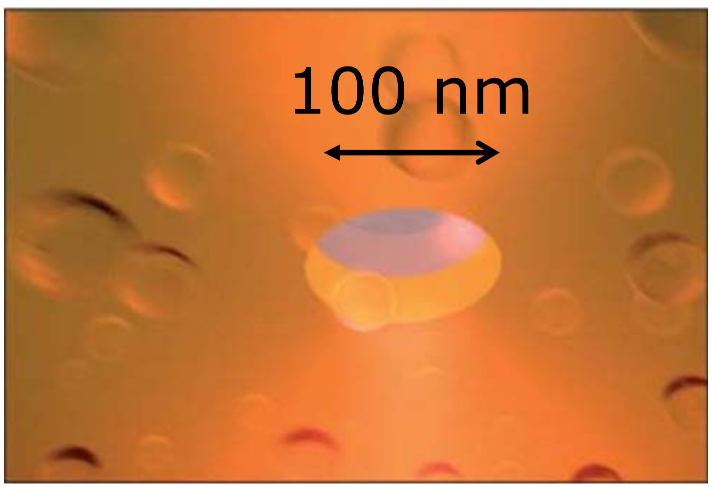


$$V_{\text{min}} \sim \left(\frac{\lambda}{n}\right)^3$$ limited by diffraction

Extreme confinement: Nanoplasmonics

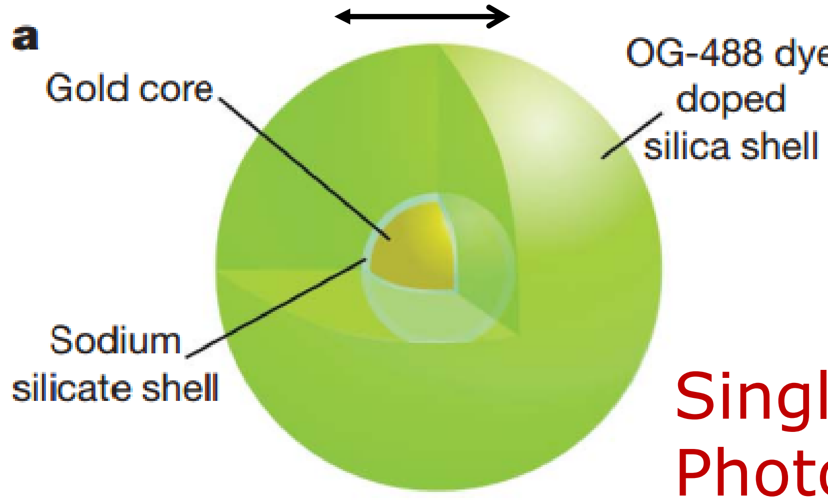
Lycurgus Cup

Nanoscale Light Trap

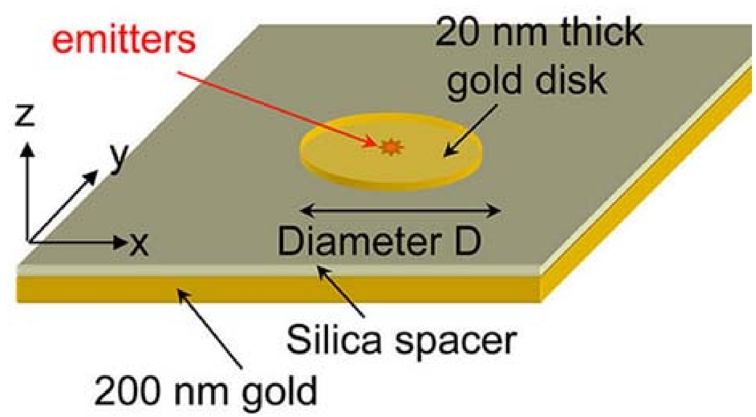


14 nm

Spaser

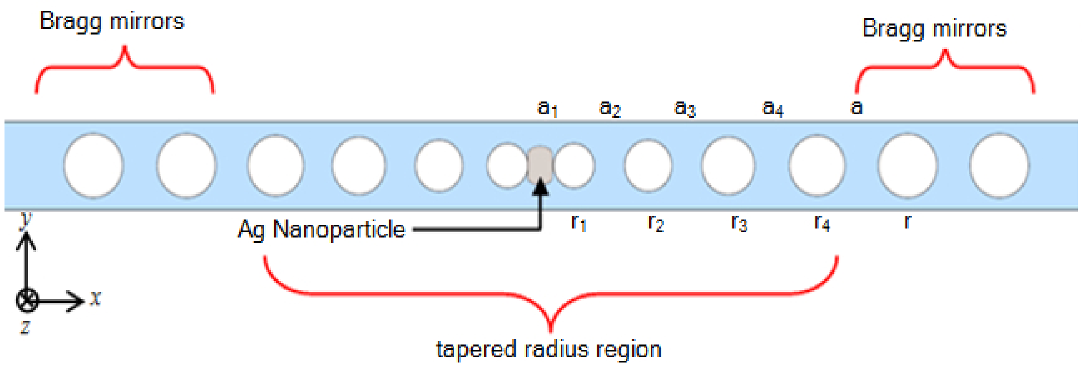


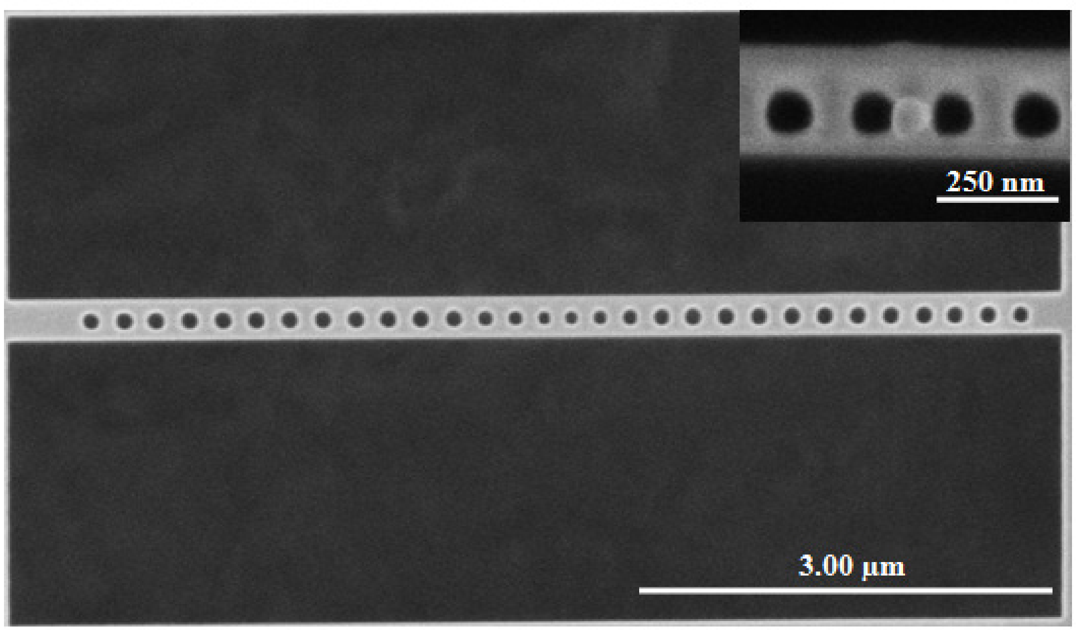
Single Photon Emitter



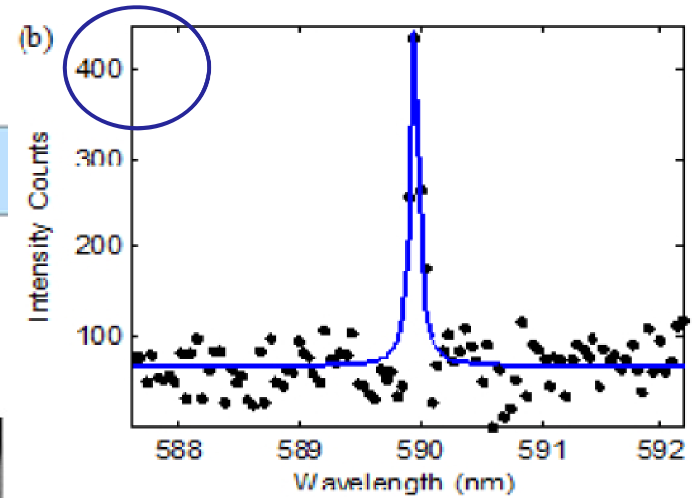
Example works of Noginov et al., Gordon et al., and Maitre et al.

Coupling metal particles with dielectric cavities

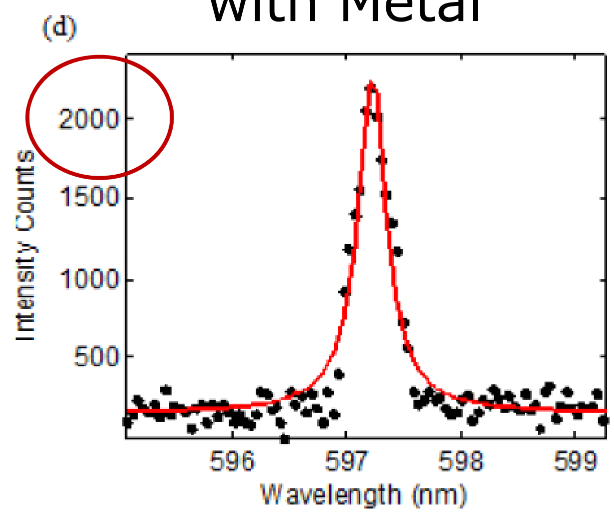




no metal

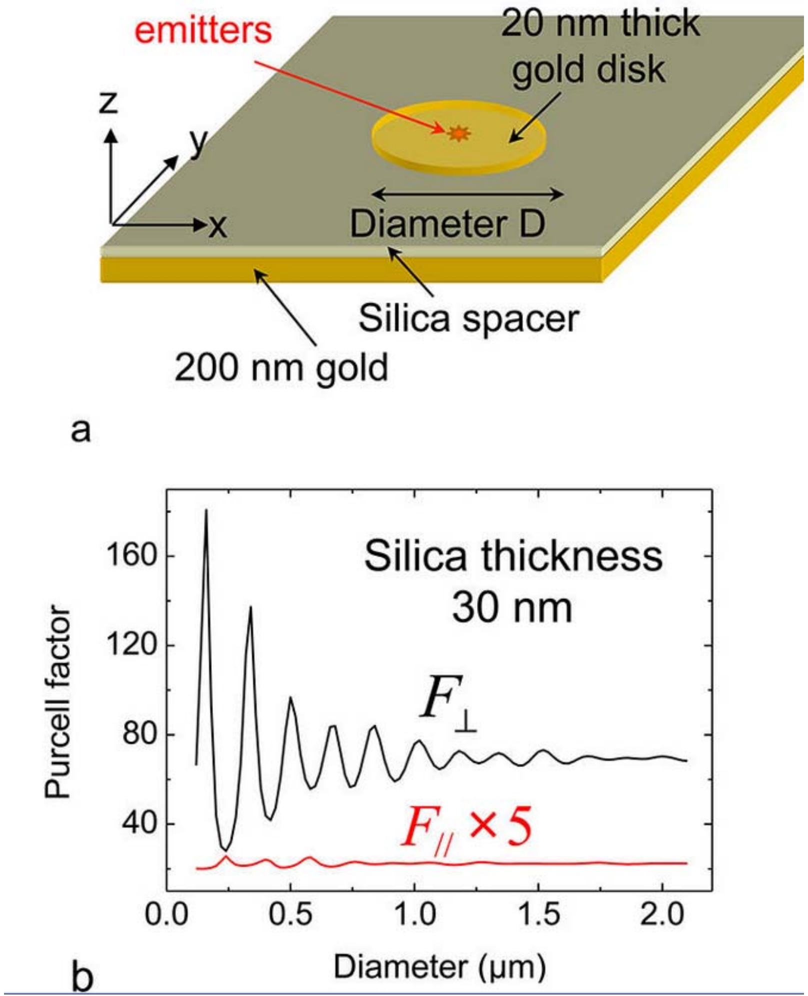


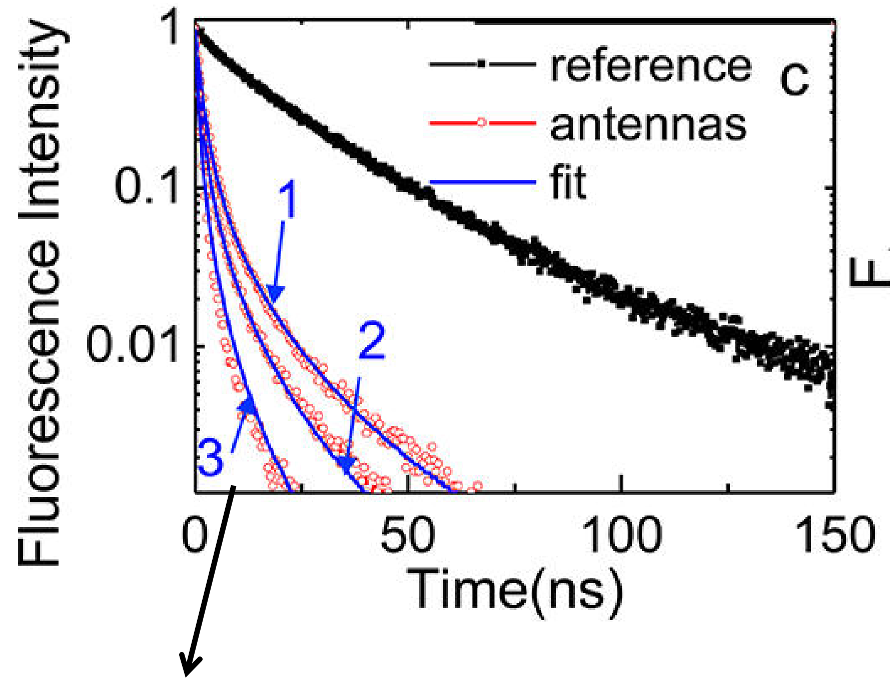
with Metal



*Mukherjee, Hajisalem, and Gordon, Opt. Express 2011.

Enhanced spontaneous emission with plasmonic *nanoscale* resonators





Enhanced emission for various nanoantenna designs

*Belacel et al., Nano Letters (2013).

Problem definition and motivation

- Modelling light-matter interactions in *open* cavity systems, especially metals, is very time consuming and numerically challenging (almost tedious!).
- However, the LDOS enhancements in cavity structures are directly attributable to one or just a few local resonances of the cavity modes.
- Unfortunately there is a disturbing lack of a precise definition for what constitutes an open cavity mode, and so their mathematical properties are somewhat unspecified and frequently ambiguous.

Outline

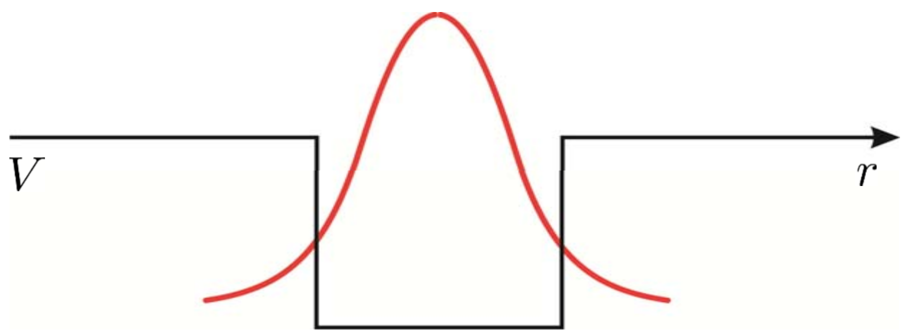
- How to fix Purcell formula for an dissipative cavity – a quasinormal mode (QNM) theory

- Examples:
  I. simple dielectric cavity
  II. plasmonic nanoresonators
  III. single photon emitter from metal dimer

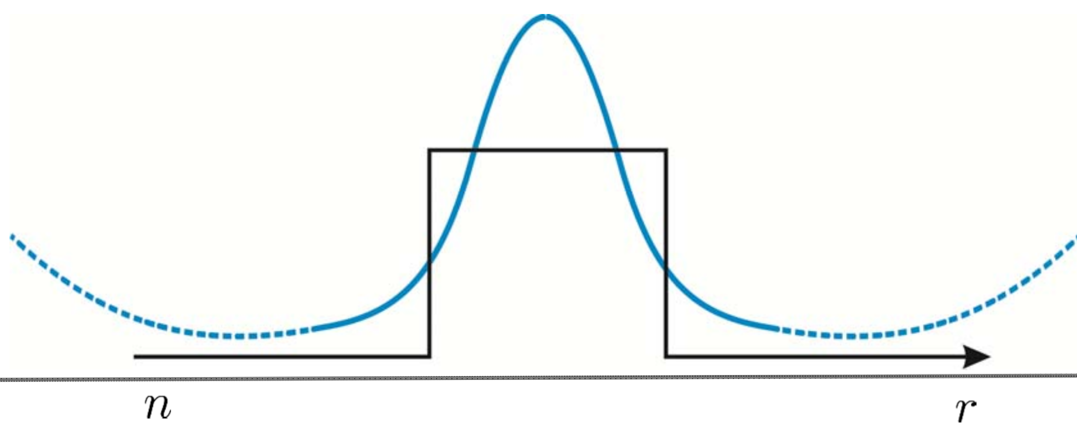
- Summary

Bound modes versus *open* cavity modes

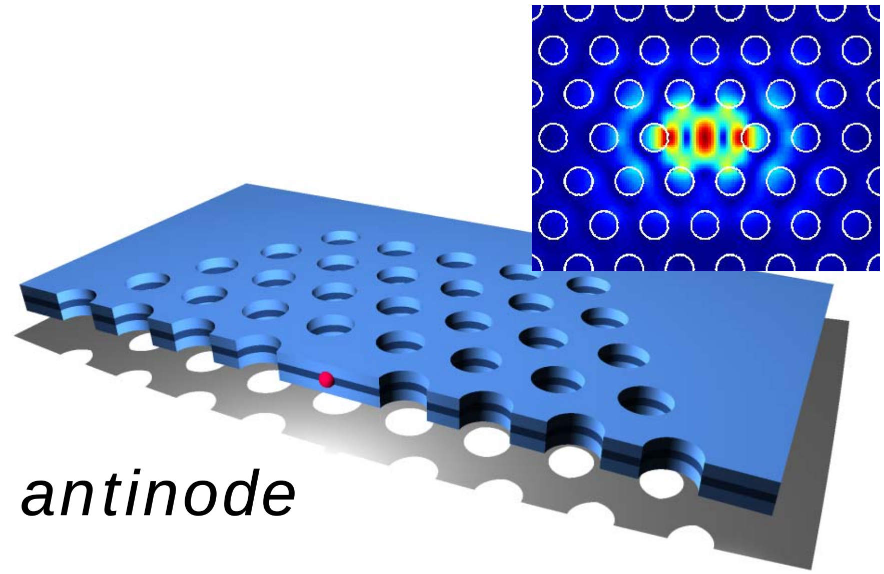
Electronic wavefunction, finite potential well:



Electric field, high-index cavity



Purcell effect in an optical cavity



- SE rate

$$\Gamma_{\mathrm{R}} = \alpha_{\mathrm{QD}}\, \rho(\mathbf{r}, \omega)$$

- Purcell factor at cavity *antinode*

$$F_{\mathrm{P}} = \frac{\rho(0,\omega)}{\rho_0} = \frac{3}{4\pi^2}\left(\frac{\lambda_{\mathrm{C}}}{n_{\mathrm{C}}}\right)^3\left(\frac{Q}{V_{\mathrm{eff}}}\right)$$

- Effective ("normal") mode volume

$$V_{\mathrm{eff}}^{\mathrm{N}} = \int_V \frac{\epsilon_{\mathrm{r}}(\mathbf{r})\,|\mathbf{f}_\mu(\mathbf{r})|^2\,\mathrm{d}\mathbf{r}}{\epsilon_{\mathrm{r}}(\mathbf{r}_{\mathrm{c}})|\mathbf{f}_\mu(\mathbf{r}_{\mathrm{c}})|^2}$$

?!

Properties of normal modes

$$\nabla \times \nabla \times \mathbf{E}(\mathbf{r}) - \frac{\omega^2}{c^2}\epsilon_r(\mathbf{r})\mathbf{E}(\mathbf{r}) = 0$$

Normal modes have fixed or periodic boundary conditions:

$$\mathbf{E}(\mathbf{r}) = 0, r = R_{\max}$$

*Real frequencies, complete
*Continous spectrum (if large V)

Normalization:

$$\langle \mathbf{f}_\mu | \mathbf{f}_\lambda \rangle = \int_V \epsilon_{\mathrm{r}}(\mathbf{r})\, \mathbf{f}_\mu^*(\mathbf{r}) \cdot \mathbf{f}_\lambda(\mathbf{r})\, \mathrm{d}\mathbf{r} = \delta_{\mu,\lambda},$$

Properties of QNMs

$$\nabla \times \nabla \times \mathbf{E}(\mathbf{r}) - \frac{\omega^2}{c^2}\epsilon(\mathbf{r})\mathbf{E}(\mathbf{r}) = 0$$

QNMs[1] have outgoing boundary conditions:

$$\mathbf{E}(\mathbf{r}) \propto \frac{1}{r}e^{i\omega r/c}, r \to \infty$$

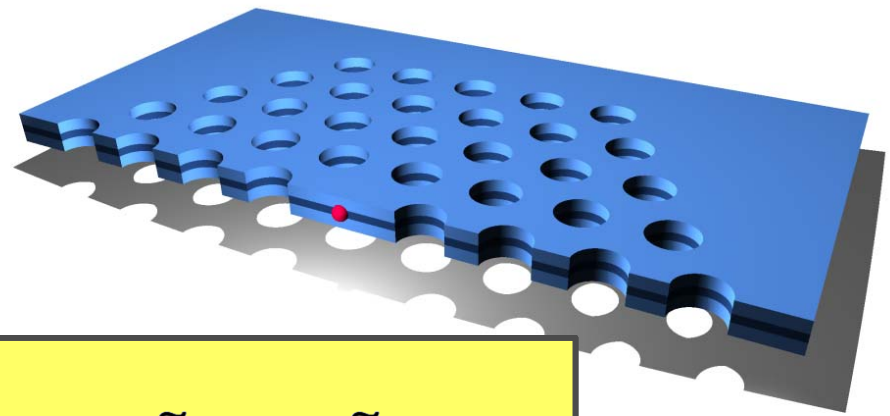
*Complex freqs, complete (?)
*Discrete spectrum

Normalization:

$$\langle\langle \tilde{\mathbf{f}}_\mu | \tilde{\mathbf{f}}_\mu \rangle\rangle = \lim_{V\to\infty} \int_V \left(\frac{1}{2\omega} \frac{\partial(\epsilon(\mathbf{r},\omega)\omega^2)}{\partial\omega} \right)_{\omega=\tilde{\omega}_{Q_1}} \tilde{\mathbf{f}}_\mu(\tilde{\mathbf{r}}) \cdot \tilde{\mathbf{f}}_\mu(\mathbf{r}) d\mathbf{r} + \frac{ic}{2\tilde{\omega}_{Q_1}} \int_{\partial V} \sqrt{\epsilon(\mathbf{r})} \tilde{\mathbf{f}}_\mu(\mathbf{r}) \cdot \tilde{\mathbf{f}}_\mu(\mathbf{r}) d\mathbf{r}.$$

* Lee et al. JOSA B (1999), Kristensen et al, OL (2012), Kristensen and Hughes, ACS Photonics (2013), Sauvan et al, PRL (2013)

Green function expansion inside scattering geometry (where QNMs are complete)



$$\mathbf{G}(\mathbf{r}_1, \mathbf{r}_2) = \sum_\alpha \frac{\omega^2}{2\tilde{\omega}_\alpha(\tilde{\omega}_\alpha - \omega)} \tilde{\mathbf{f}}_\alpha(\mathbf{r}_1)\tilde{\mathbf{f}}_\alpha(\mathbf{r}_2)$$

Complex eigenfrequencies:

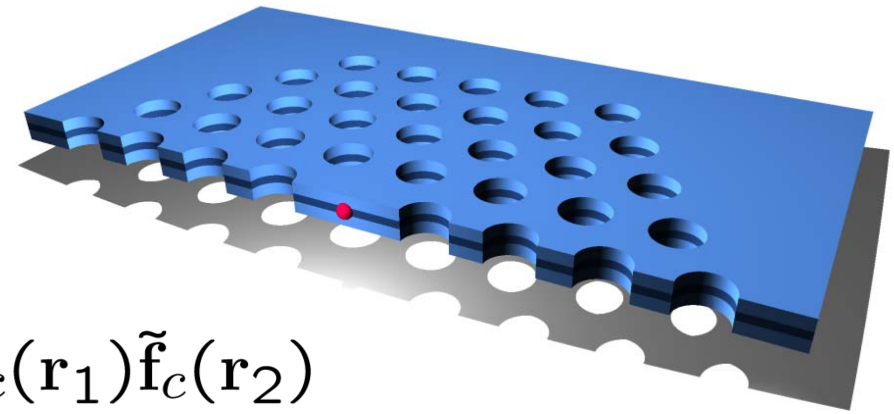
$$\tilde{\omega}_\alpha = \omega_\alpha - i\gamma_\alpha$$

Quality factor of each mode:

$$Q_\alpha = \frac{\omega_\alpha}{2\gamma_\alpha}$$

Single QNM Green function

$$\tilde{\omega}_c = \omega_c - i\gamma_c \qquad Q = \frac{\omega_c}{2\gamma_c}$$

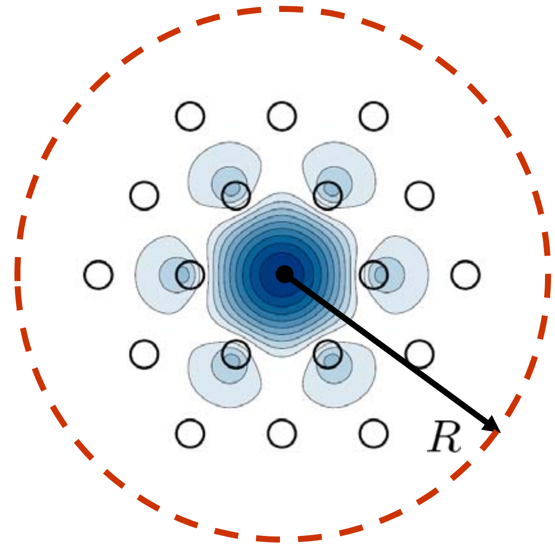


$$\mathbf{G}^{\mathsf{f}}(\mathbf{r}_1, \mathbf{r}_2) \approx \frac{\omega^2}{2\tilde{\omega}_c(\tilde{\omega}_c - \omega)} \tilde{\mathbf{f}}_c(\mathbf{r}_1)\tilde{\mathbf{f}}_c(\mathbf{r}_2)$$

$$F_{\mathsf{P}}(\mathbf{r}_d) = \frac{\mathbf{n}_d \cdot \mathrm{Im}\mathbf{G}^{\mathsf{f}}(\mathbf{r}_d, \mathbf{r}_d; \omega) \cdot \mathbf{n}_d}{\mathbf{n}_d \cdot \mathrm{Im}\mathbf{G}^{\mathsf{hom}}(\mathbf{r}_d, \mathbf{r}_d; \omega) \cdot \mathbf{n}_d} \propto \frac{Q}{V_{\mathsf{eff}}}$$

⇨ Rigorous definition of V_{eff} and $F_{\mathsf{P}}(\mathbf{r}_d)$

Effective mode volume



Normal-mode volume

$$V_{\text{eff}}^{N} = \frac{1}{\varepsilon(\mathbf{r}_c)} \frac{\langle \mathbf{f}_\mu | \mathbf{f}_\mu \rangle}{|\mathbf{f}_\mu^2(\mathbf{r}_c)|^2}, \quad \langle \mathbf{f}_\mu | \mathbf{f}_\mu \rangle = \int_V \varepsilon(\mathbf{r}) |\mathbf{f}_\mu^2(\mathbf{r})|^2 d\mathbf{r}$$

(Corrected) Quasi-mode volume [1]

$$V_{\text{eff}}^{Q} = \text{Re}\left\{\frac{1}{v_Q}\right\}, \quad v_Q = \frac{1}{\varepsilon(\mathbf{r}_c)} \frac{\langle\langle \tilde{\mathbf{f}}_\mu | \tilde{\mathbf{f}}_\mu \rangle\rangle}{\tilde{\mathbf{f}}_\mu^2(\mathbf{r}_c)}$$

$$\langle\langle \tilde{\mathbf{f}}_\mu | \tilde{\mathbf{f}}_\mu \rangle\rangle = \lim_{V \to \infty} \int_V \varepsilon(\mathbf{r}) \tilde{\mathbf{f}}_\mu \cdot \tilde{\mathbf{f}}_\lambda d\mathbf{r} + \frac{ic}{2\tilde{\omega}_\mu} \int_{\partial V} \varepsilon(\mathbf{r}) \tilde{\mathbf{f}}_\mu \cdot \tilde{\mathbf{f}}_\mu d\mathbf{r}$$

[1] Kristensen, Van Vlack, Hughes, Opt. Lett. 37, 1649 (2012)

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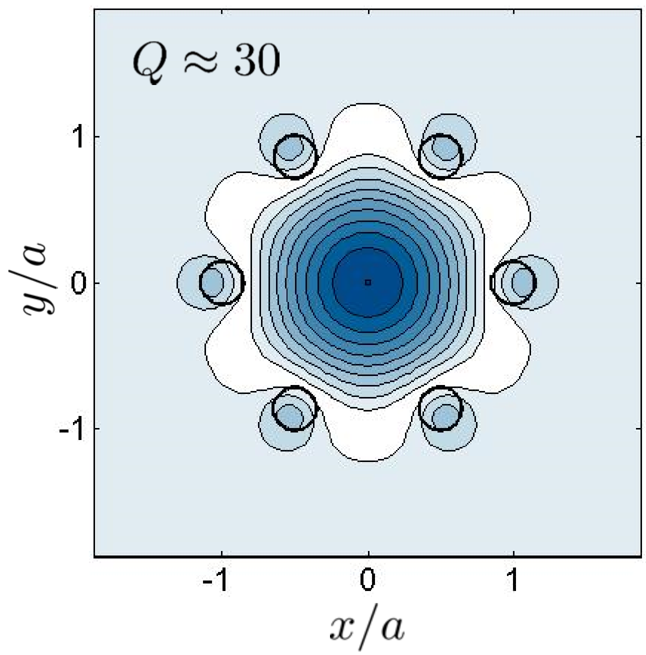
Quasi-modes for simple dielectric cavity

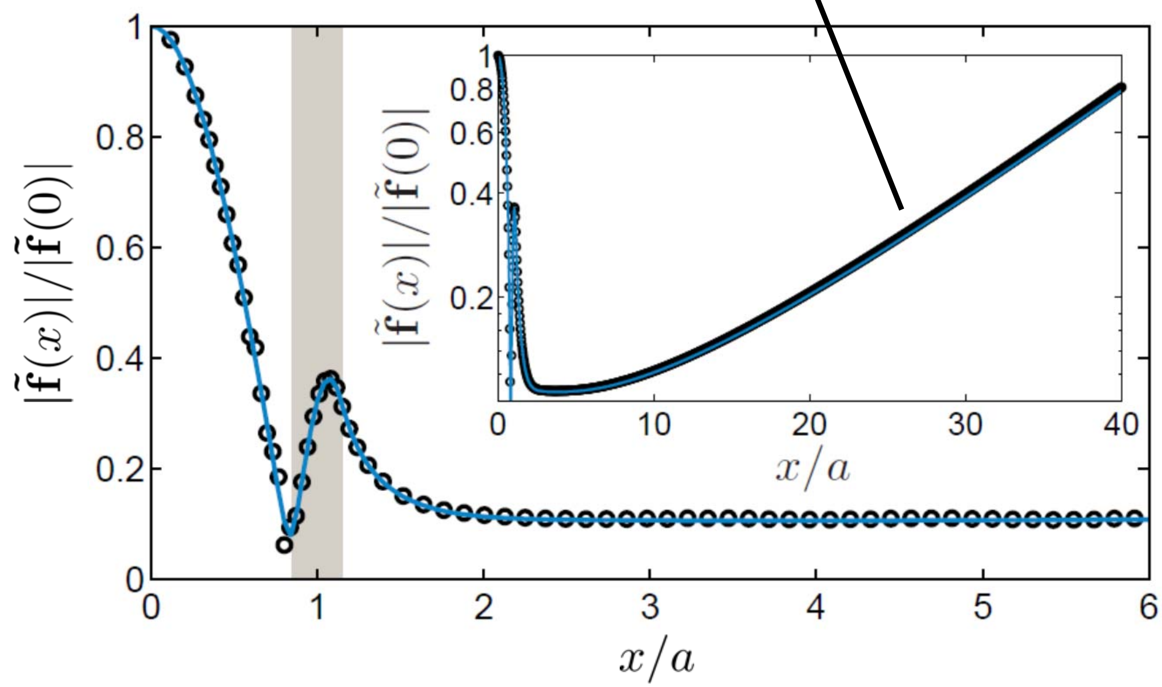
Can use a Fredholm-type equation:

$$\mathbf{E}(\mathbf{r},\omega) = \left(\frac{\omega}{c}\right)^2 \int_V \mathbf{G}^{\mathrm{B}}(\mathbf{r},\mathbf{r}',\omega)\,\Delta\varepsilon(\mathbf{r}')\,\mathbf{E}(\mathbf{r}',\omega)\mathrm{d}\mathbf{r}'.$$

… or use FDTD*

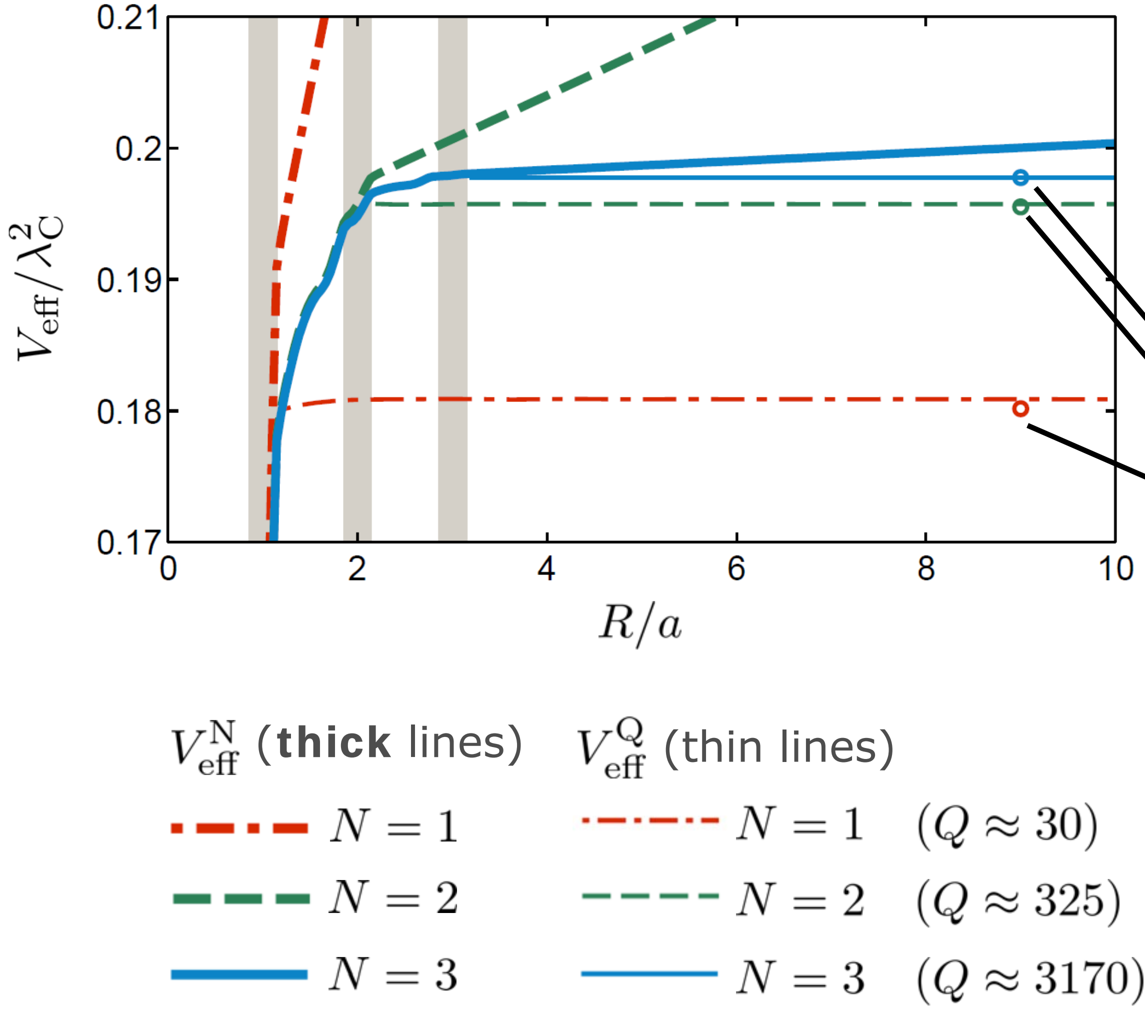
two different numerical techniques

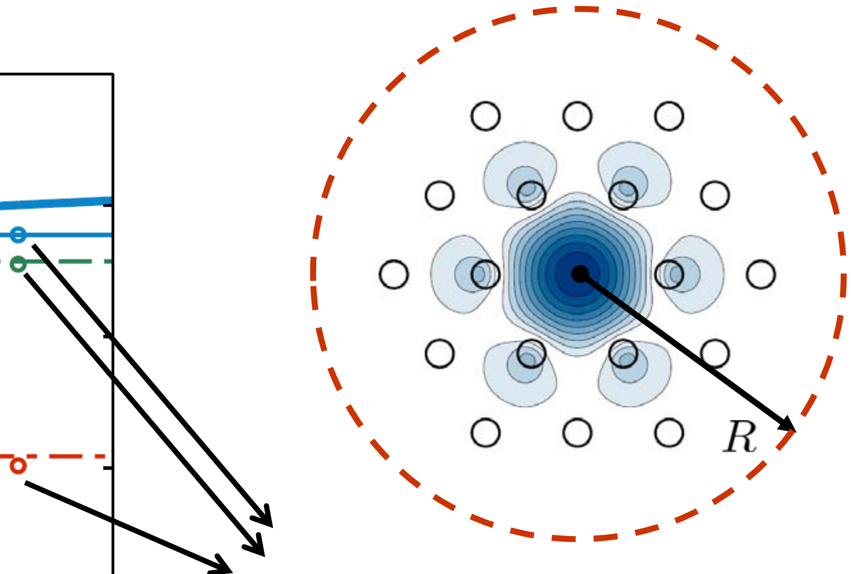




* We use Lumerical FDTD: www.lumerical.com

Corrected effective mode volumes





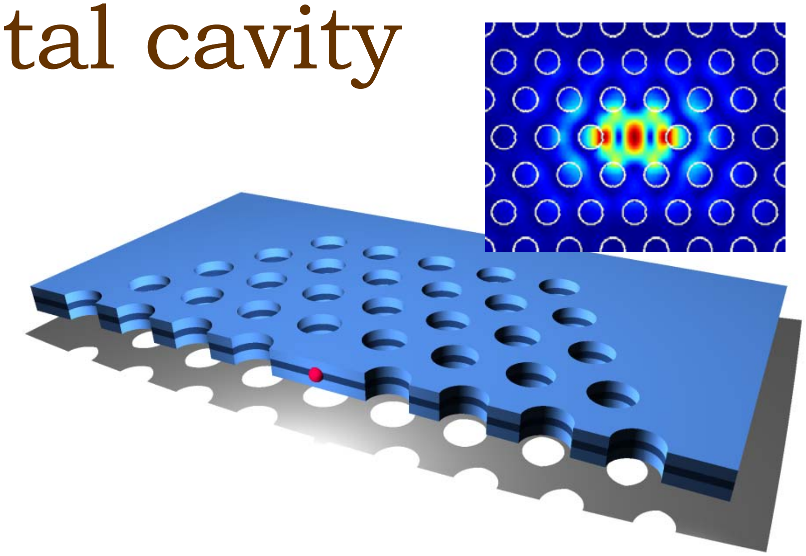
Exact solutions from numerical **G**

N = no of layers

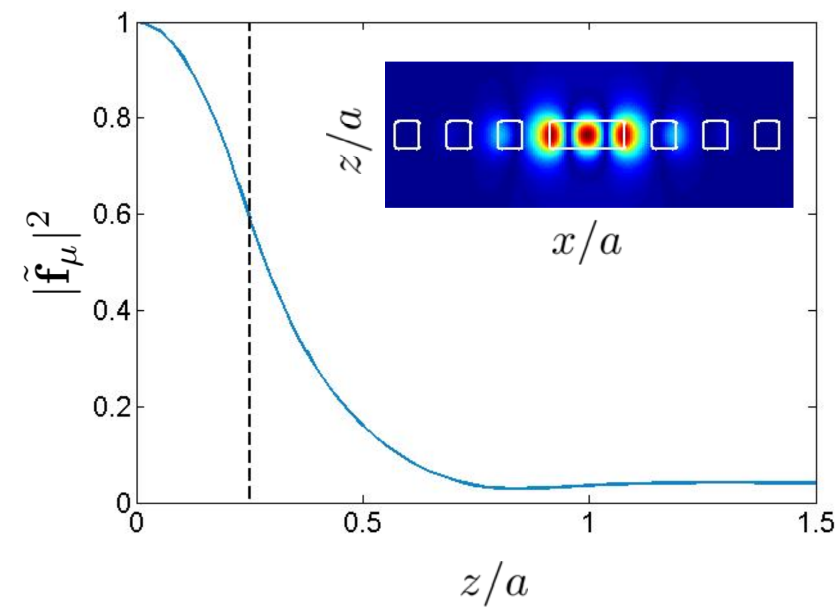
[1] Kristensen, Van Vlack, Hughes, Opt. Lett. 37, 1649 (2012)

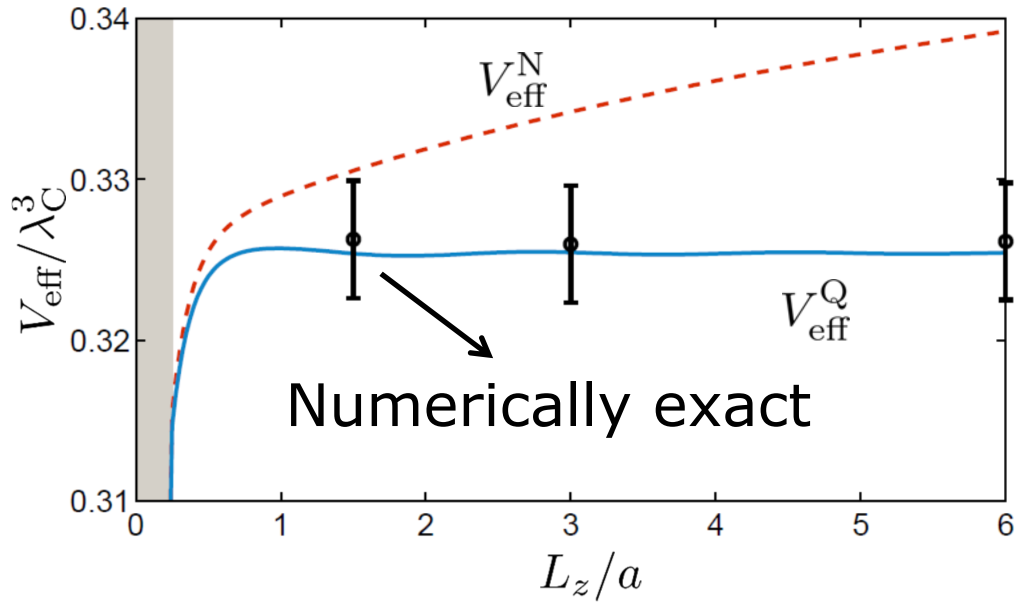
3D photonic crystal cavity

Using FDTD we get the resonance frequency, Q factor and mode profile.



Field along center:

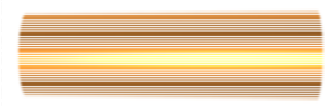


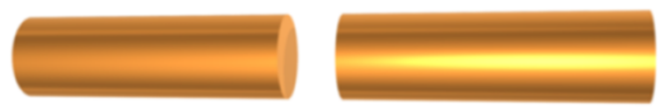


[1] Kristensen, Van Vlack, Hughes, Opt. Lett. 37, 1649 (2012)

Next consider a metal nanoparticle (MNP)

e.g.





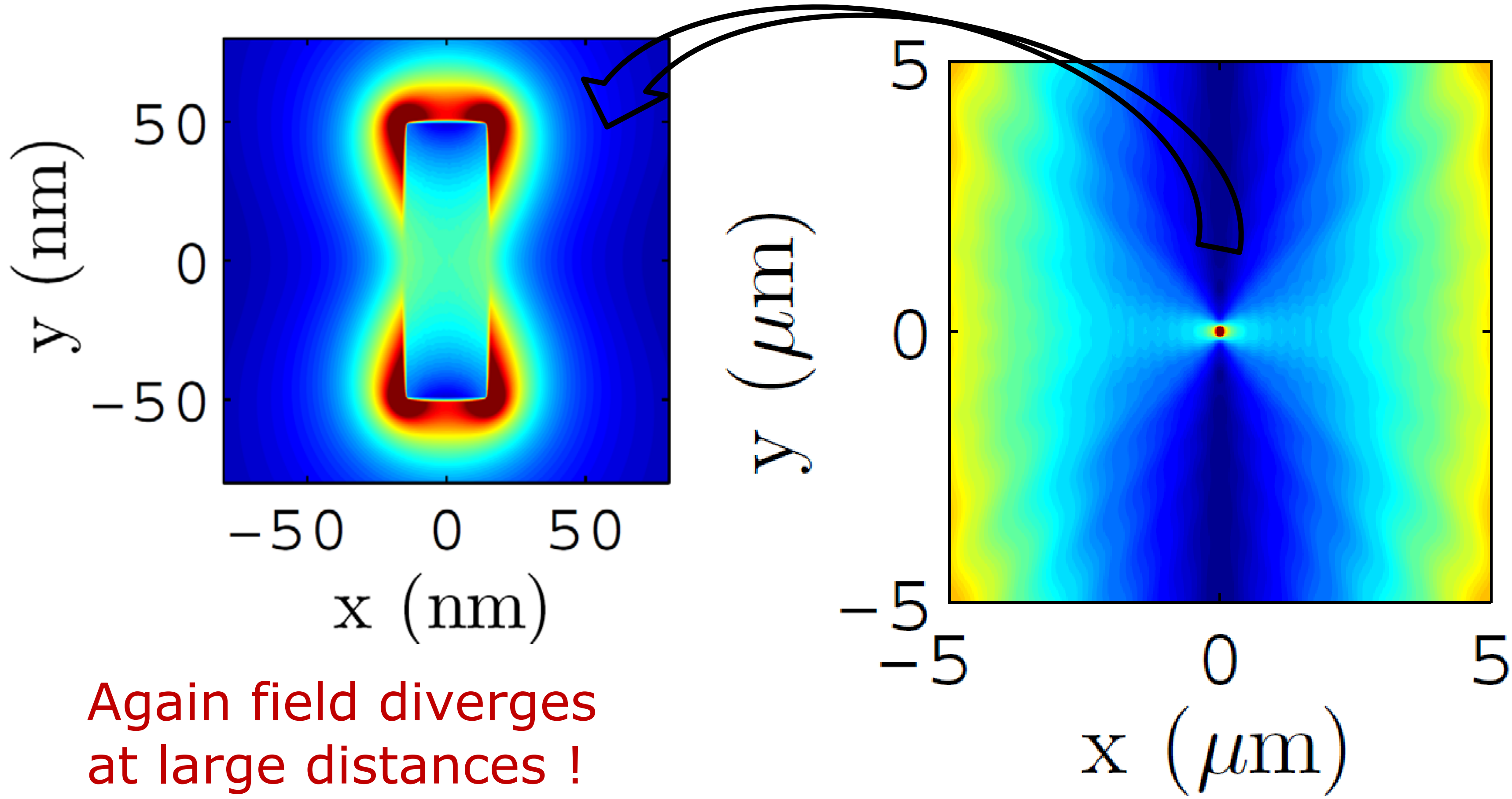
With a Drude model for the dielectric constant

$$\epsilon(\omega) = 1 - \frac{\omega_p^2}{\omega(\omega + i\gamma)}$$

Metal nanoresonator

2D picture of a quasi-mode for a metal cylinder

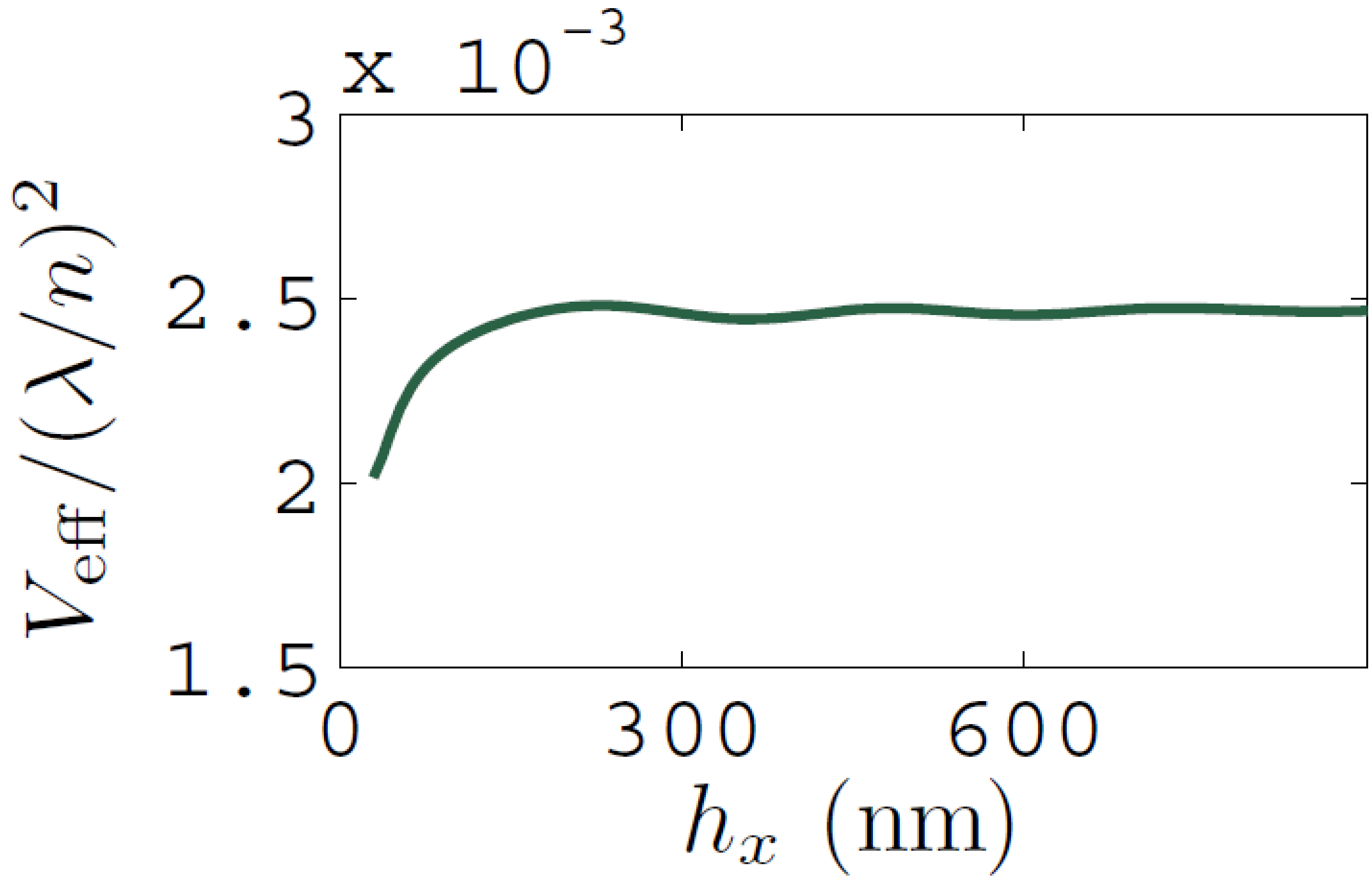
(10 by 80 nm gold nanorod)



Again field diverges
at large distances !

* See also related work by Sauvan et al, PRL (2013)

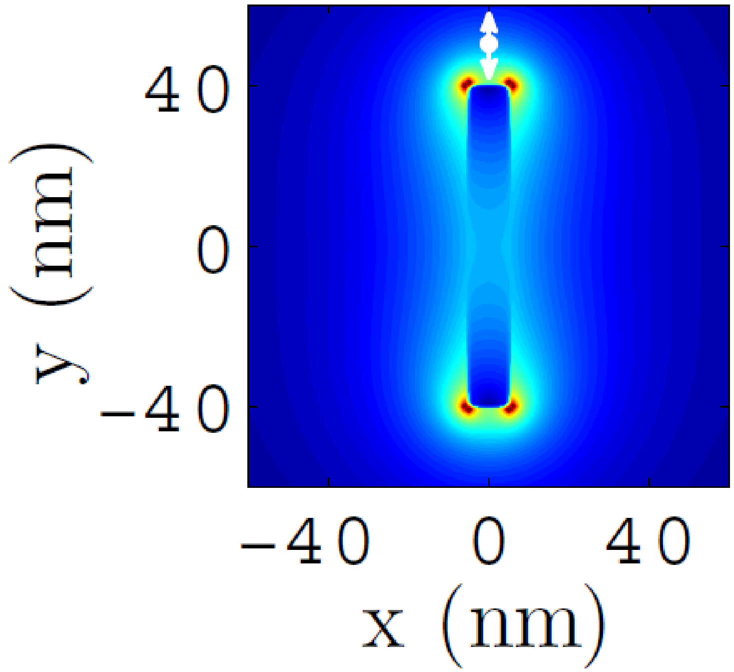
Mode volume versus domain size



Saturates very quickly, roughly at the caustic radius [1]

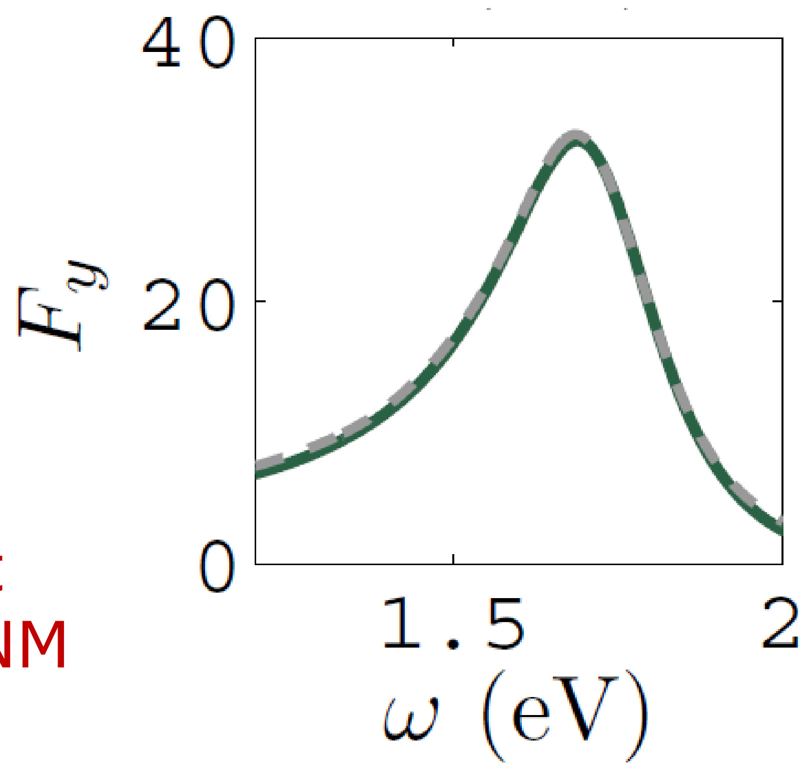
[1] See Snyder and Love, Optical Waveguide Theory (1983)

Exact emission factor vs single QNM



Single QNM theory

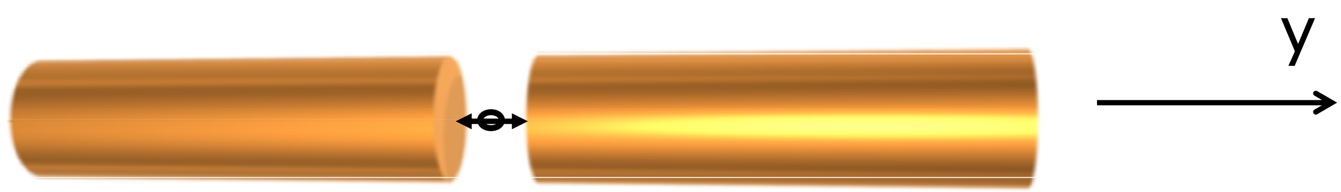
numerically exact (FDTD)



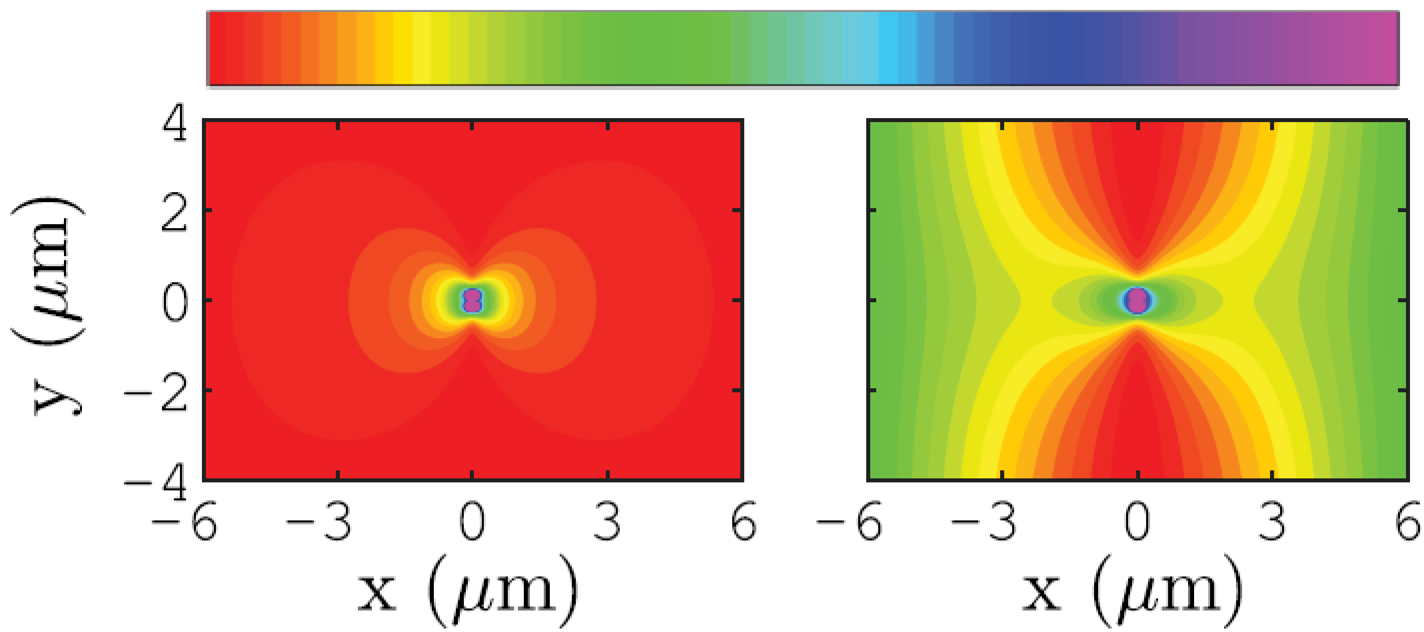
⇨ Excellent agreement with just a single QNM

Ge, Kristensen, Young, and Hughes, N. J. Phys (2014)

Single Photon Emitter from Metal Dimer



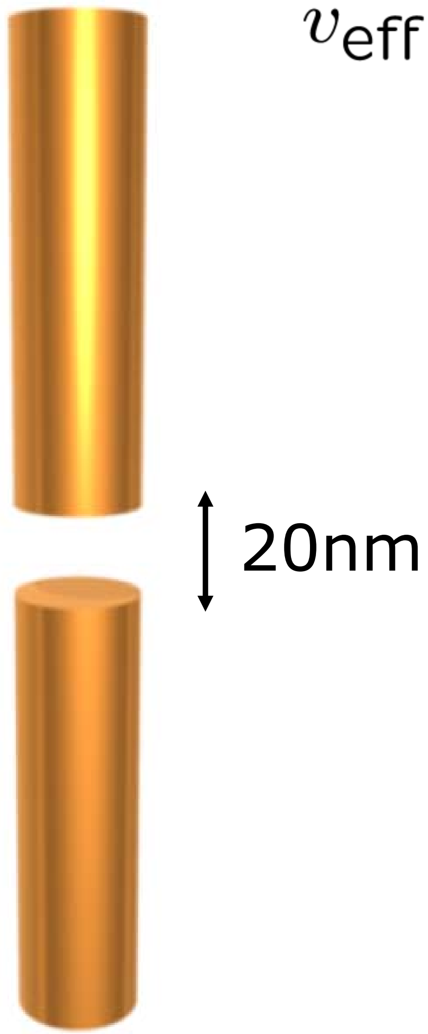
Scattered field: QNM field:

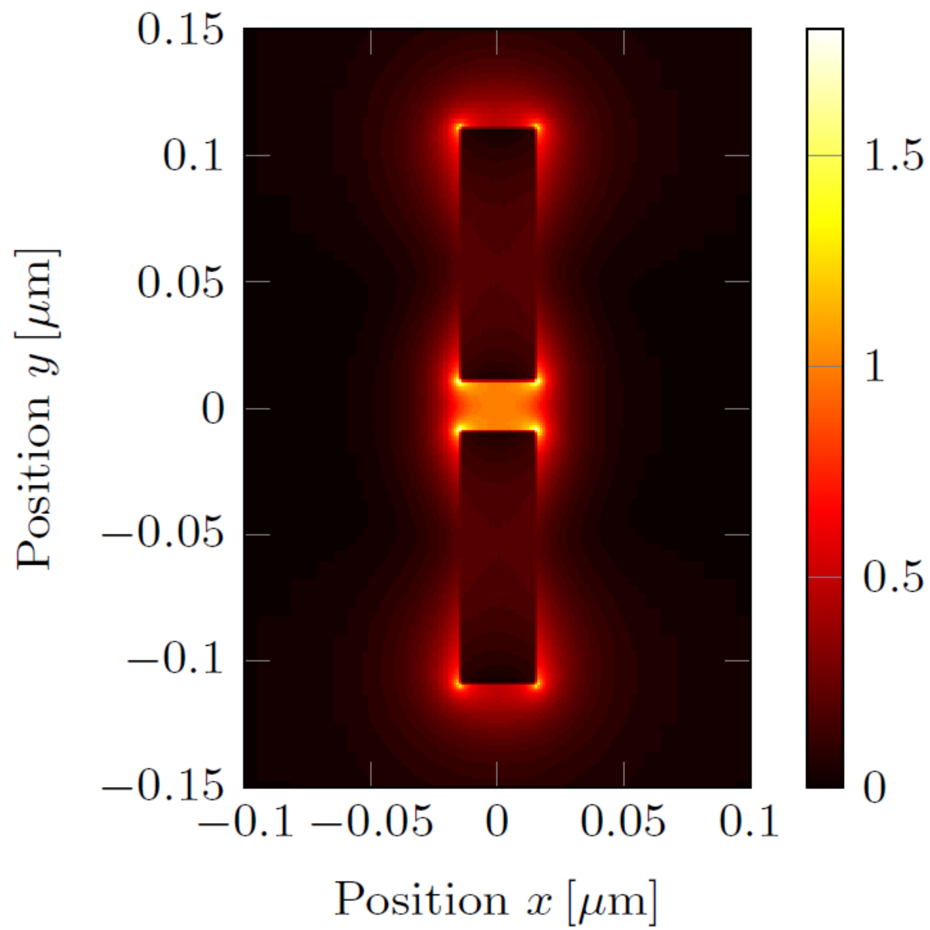


Ge and Hughes, Optics Letters (2014)

Single Photon Emitter from Metal Dimer

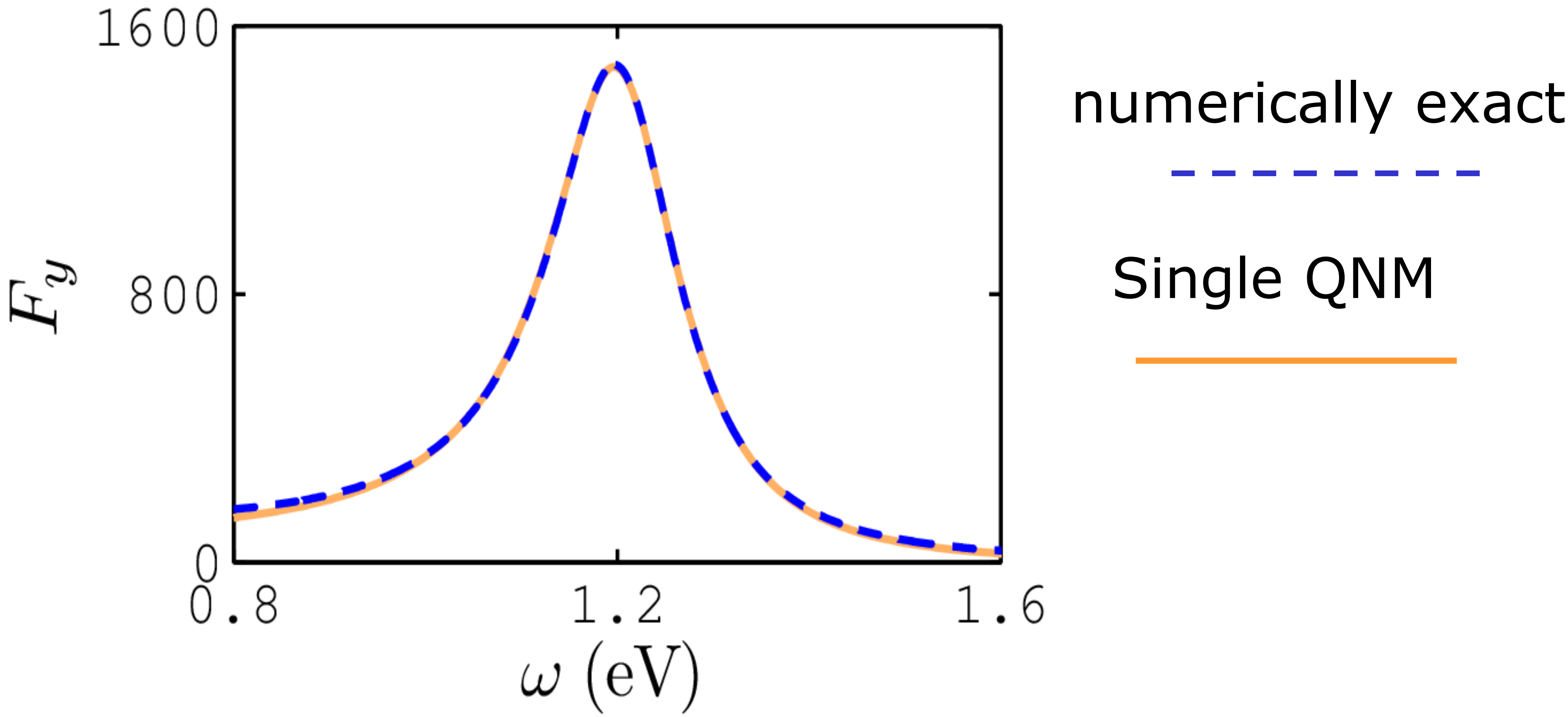
$$v_{\text{eff}} \approx 1.9 \times 10^{-4} \left(\frac{\lambda}{n_B}\right)^3$$





Ge and Hughes, Optics Letters (2014)

Enhanced emission at center of dimer



Single photon radiative output coupling:
beta factor ~ 60%

Ge and Hughes, Optics Letters (2014)

Comments

- Q/V of the dimer is about 3 times smaller than the single gold nanorod, yet the enhanced emission factor is about 3 times larger at an equivalent dipole position (10nm from surface)
  - Q/V is not the correct metric for emission factors away from the antinode!
- Quenching is also reduced in the dimer (beta factor is better) since more of the QNM lives outside the lossy metal.

Summary

- We have presented a QNM technique for dielectric and plasmonic resonators which largely fixes a bunch of ambiguities in the literature.
- Our approach allows for an accurate and clear definition of effective mode volume, the Purcell factor, and the propagator, at all spatial positions.
- Broad applications to a wide range of material systems, including nonlinear optics, quantum optics, quantum nanoplasmonics, and hybrid semiconductor-metal structures.

Refs: Kristensen, Van Vlack, Hughes, Opt. Lett. 37, 1649 (2012)
Ge, Kristensen, Young, and Hughes, N. J. Phys (2013)
Kristensen and Hughes, Perspectives Article, ACS Photonics (2014)