Quantum optics with metal nanoparticles

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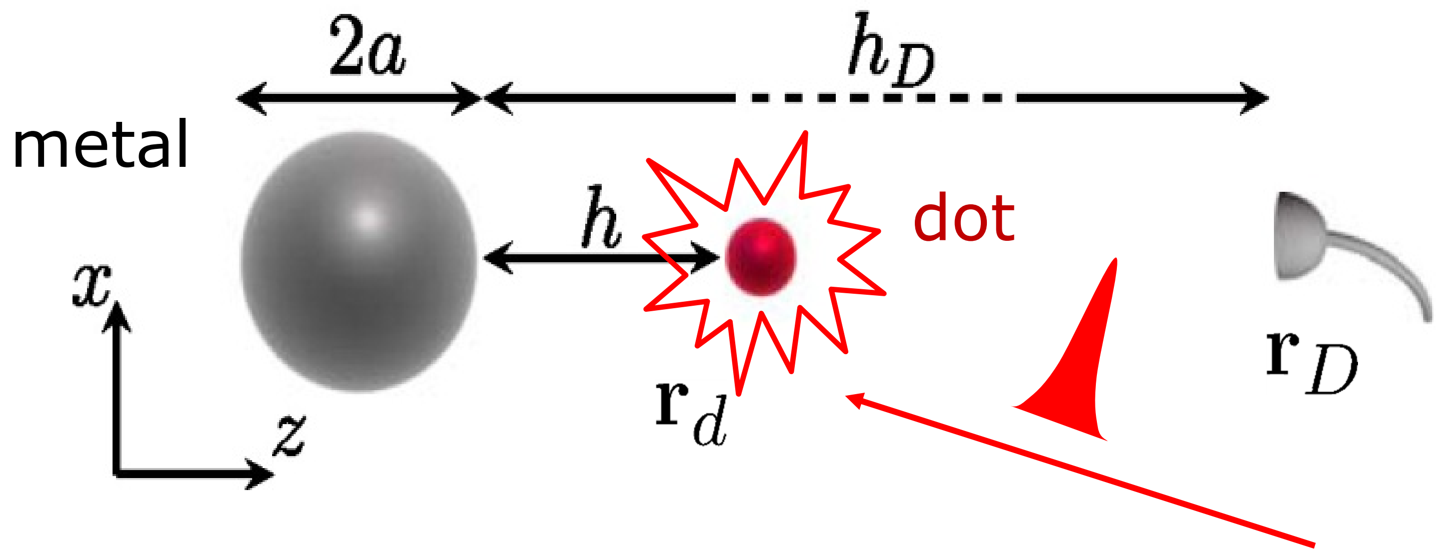


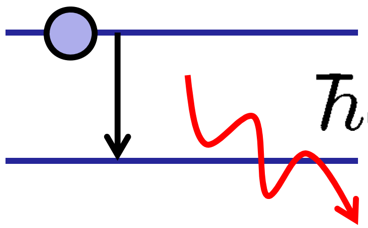
Rongchun Ge, P. T. Kristensen,
C. Van Vlack, J.F. Young, NSERC ($)

Outline

- Background
- Strong coupling regime
- Field-driven Mollow triplets
- "Local field problem" in quantum optics
  (*probably skip and summarize*)

Quick recap

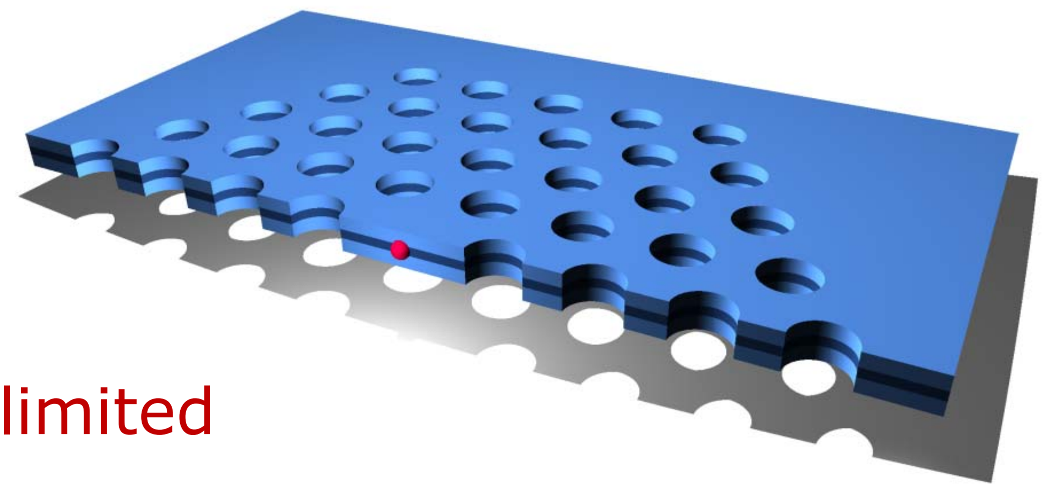
- General scaling for light-matter interactions



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

.... Quality factor

.... Volume

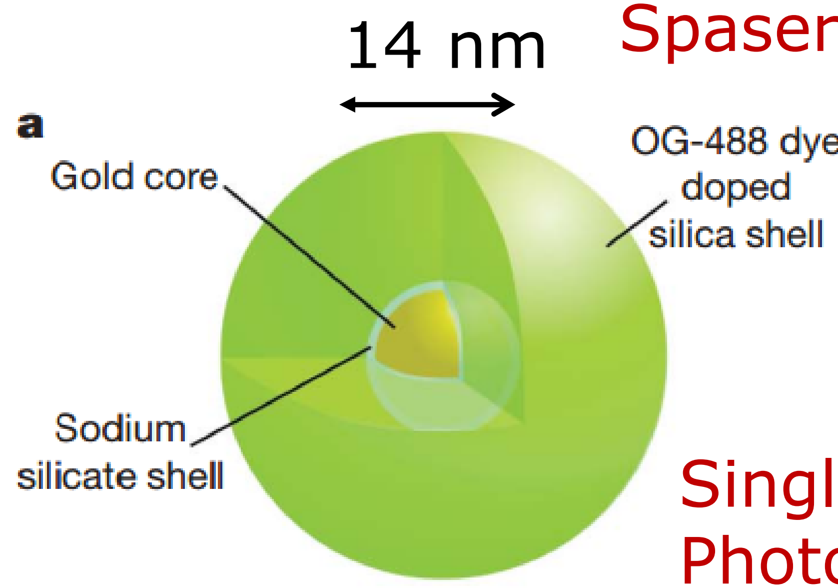


Size enhancements limited by diffraction

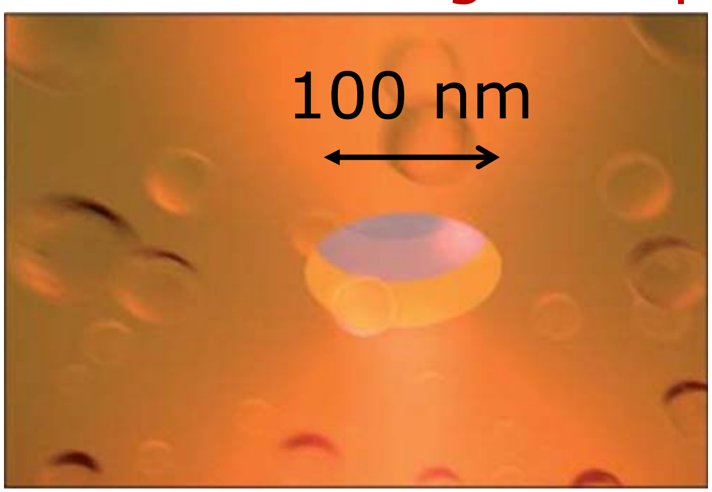
Nanoplasmonics offers a new platform for QO

Lycurgus Cup

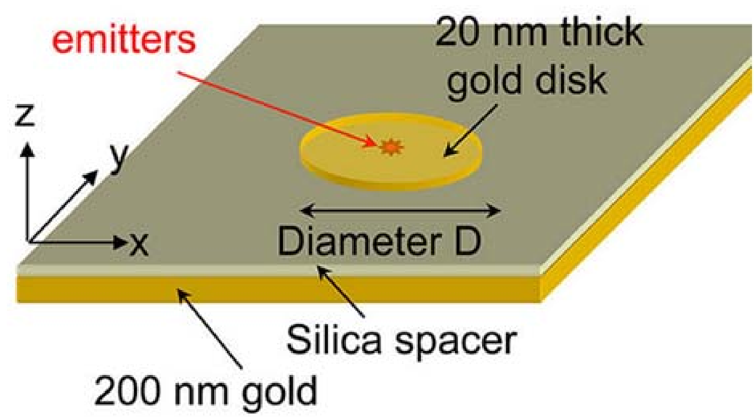
Spaser



Nanoscale Light Trap



Single Photon Emitter



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

Motivation for theory

- Textbook quantum optics usually not deal at all with metals
- The problem has some conception problems
- Theories and quantum optical regimes largely unexplored
- Lots of emerging experiments

What is quantum plasmonics?

nature physics

REVIEW ARTICLE

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Quantum plasmonics

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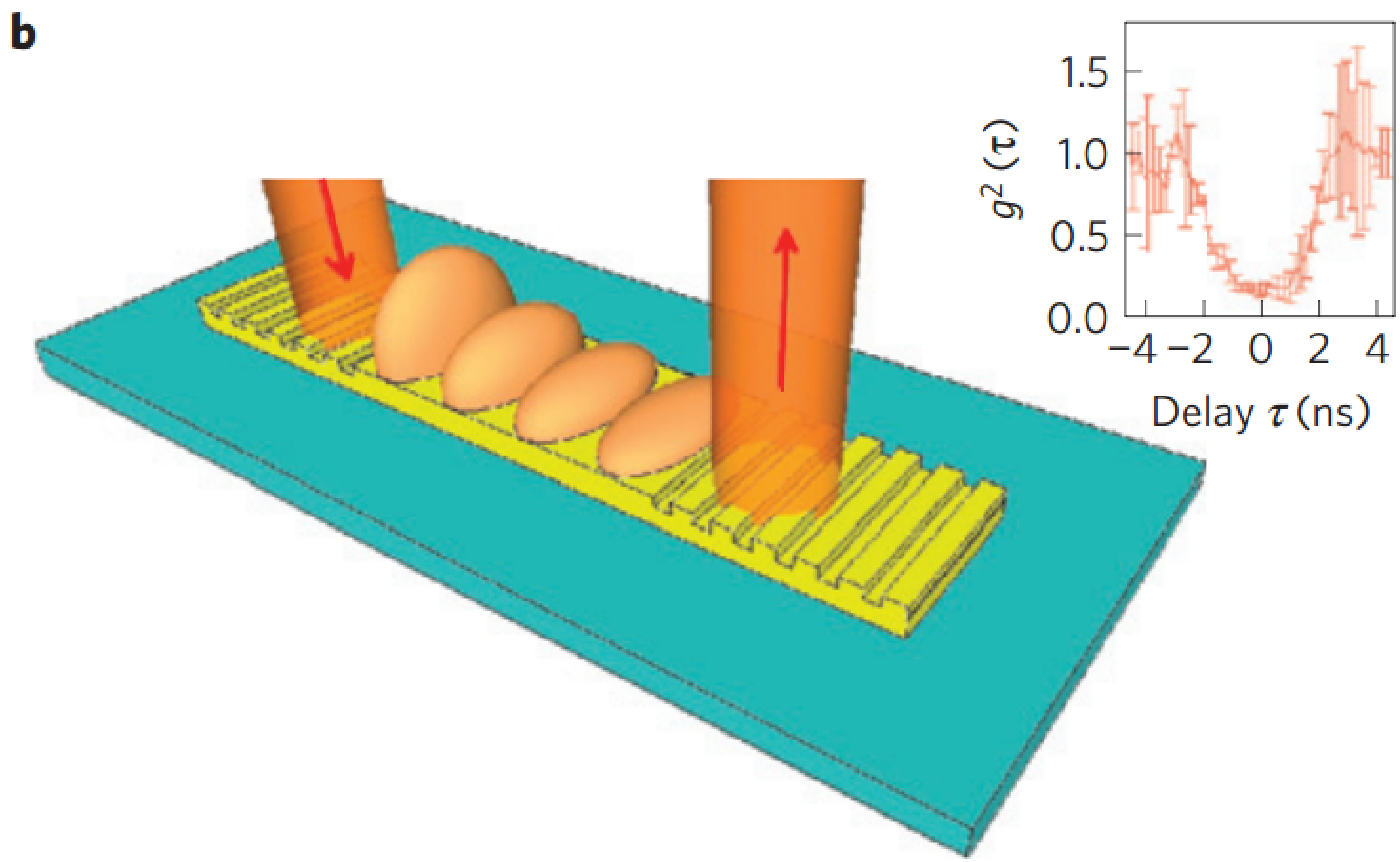
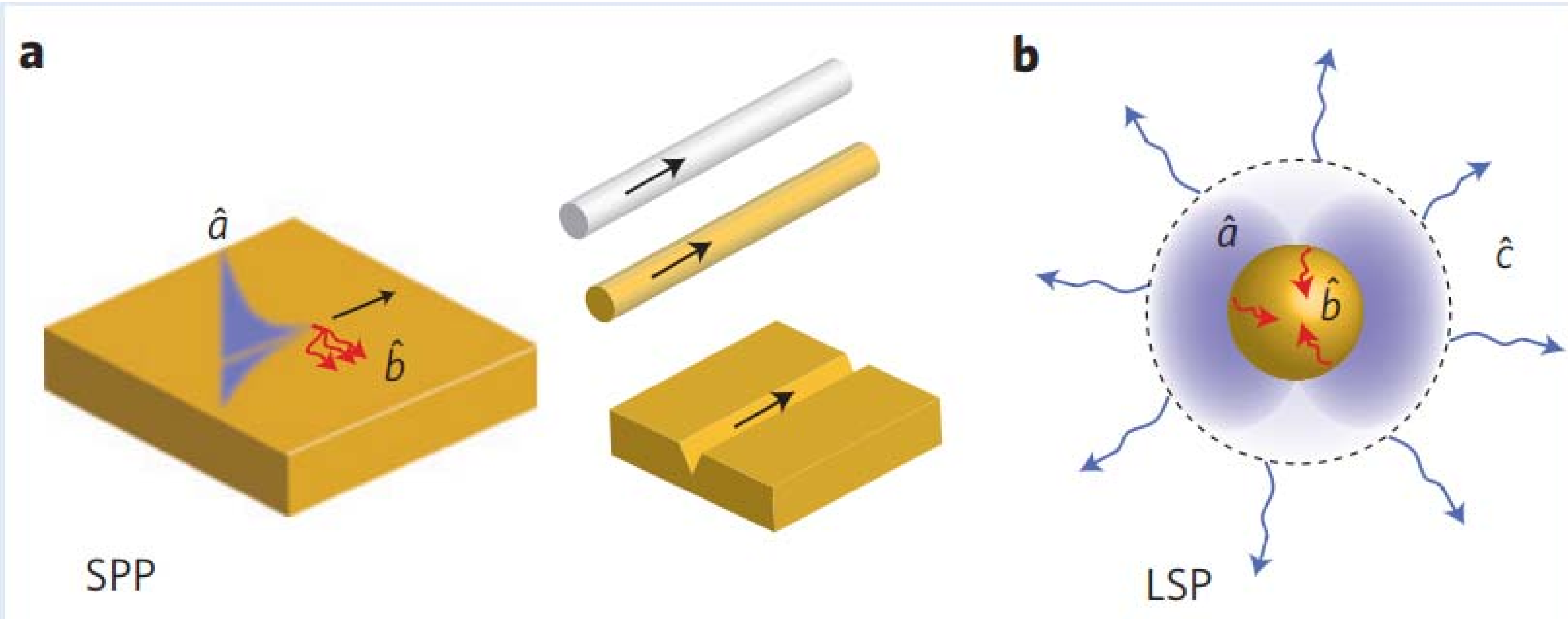
Quantum plasmonics is a rapidly growing field of research that involves the study of the quantum properties of light and its interaction with matter at the nanoscale. Here, surface plasmons—electromagnetic excitations coupled to electron charge density waves on metal-dielectric interfaces or localized on metallic nanostructures—enable the confinement of light to scales far below that of conventional optics. We review recent progress in the experimental and theoretical investigation of the quantum properties of surface plasmons, their role in controlling light-matter interactions at the quantum level and potential applications. Quantum plasmonics opens up a new frontier in the study of the fundamental physics of surface plasmons and the realization of quantum-controlled devices, including single-photon sources, transistors and ultra-compact circuitry at the nanoscale.

Single plasmon creation



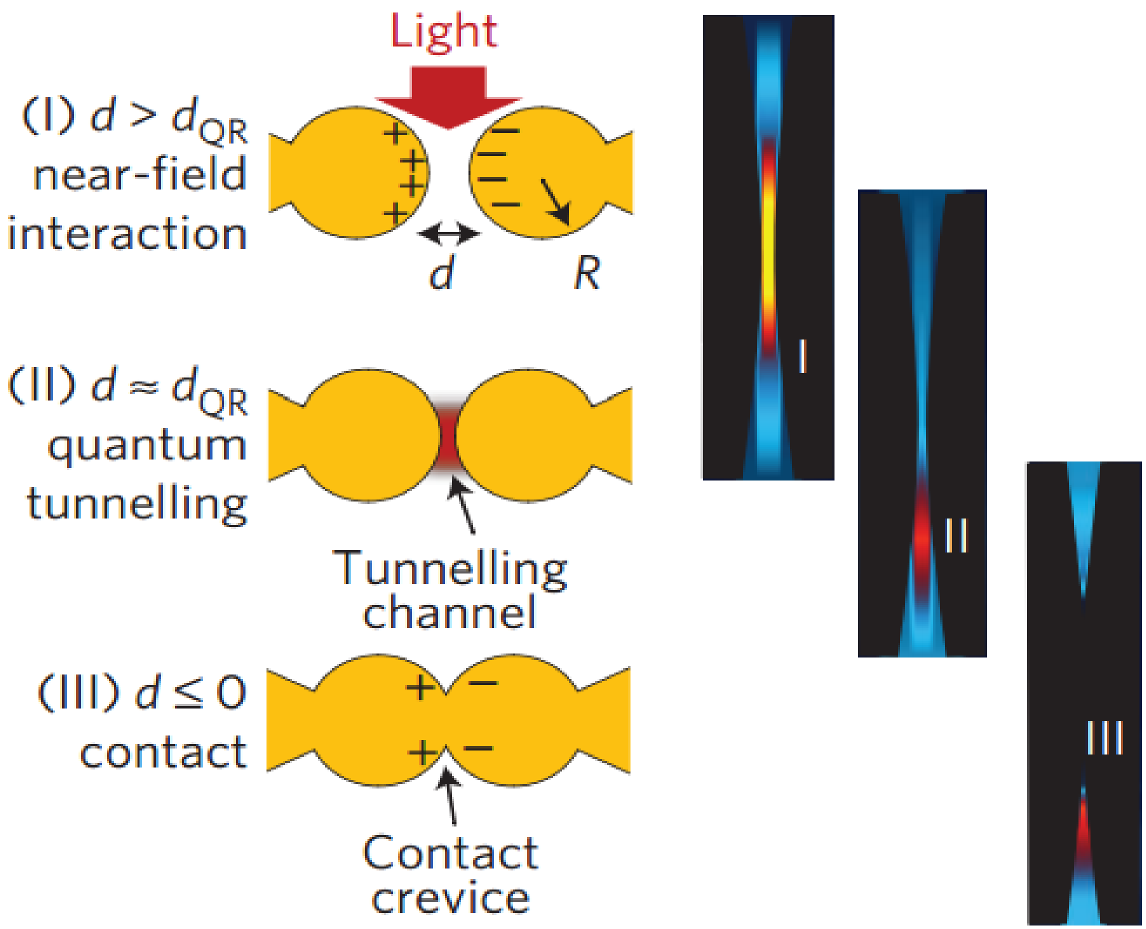
Tame et al, Nature Physics (2013)

Various geometries have been realized



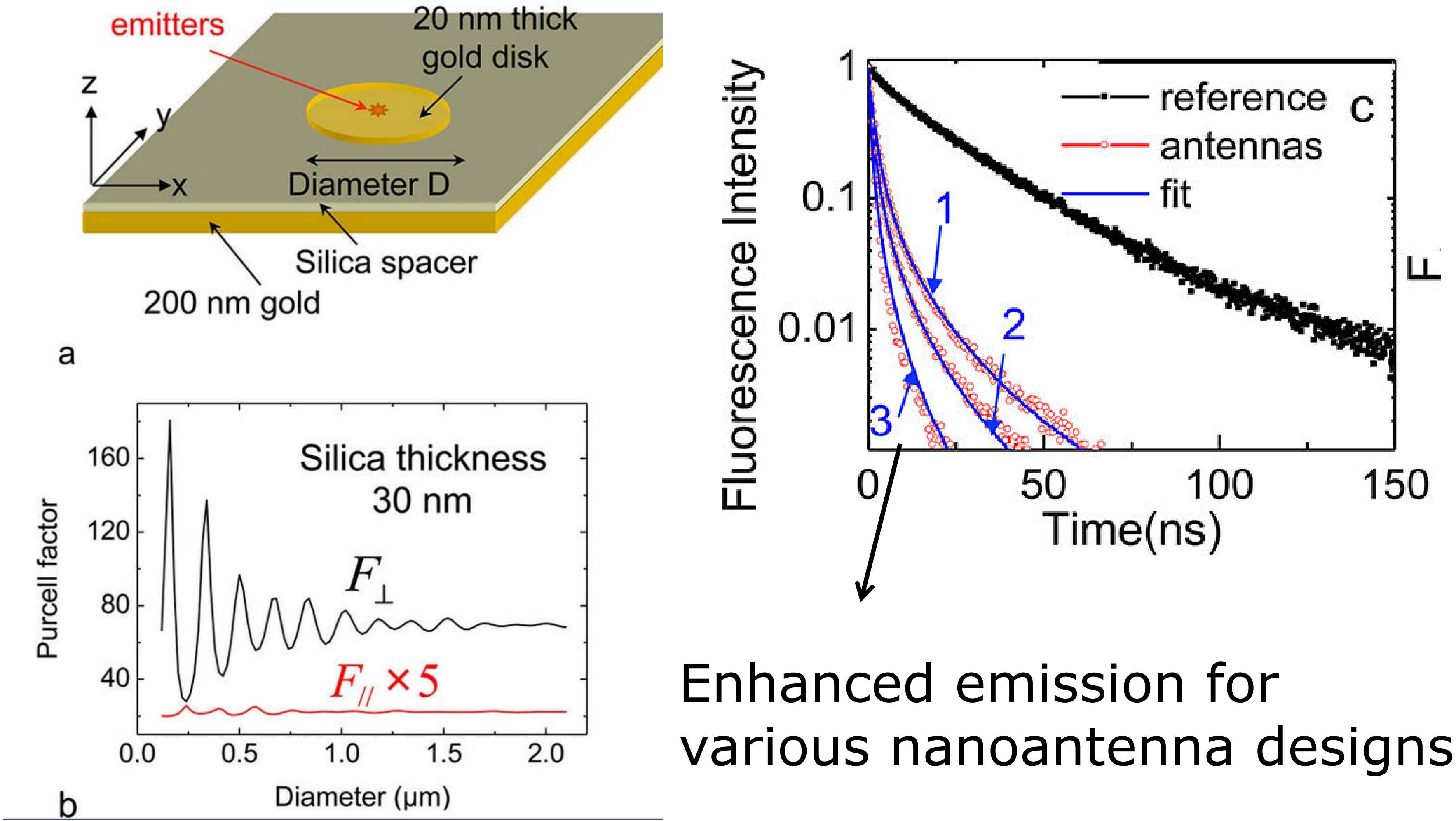
Tame et al, Nature Physics (2013)

Quantum tunneling regime



Tame et al, Nature Physics (2013)

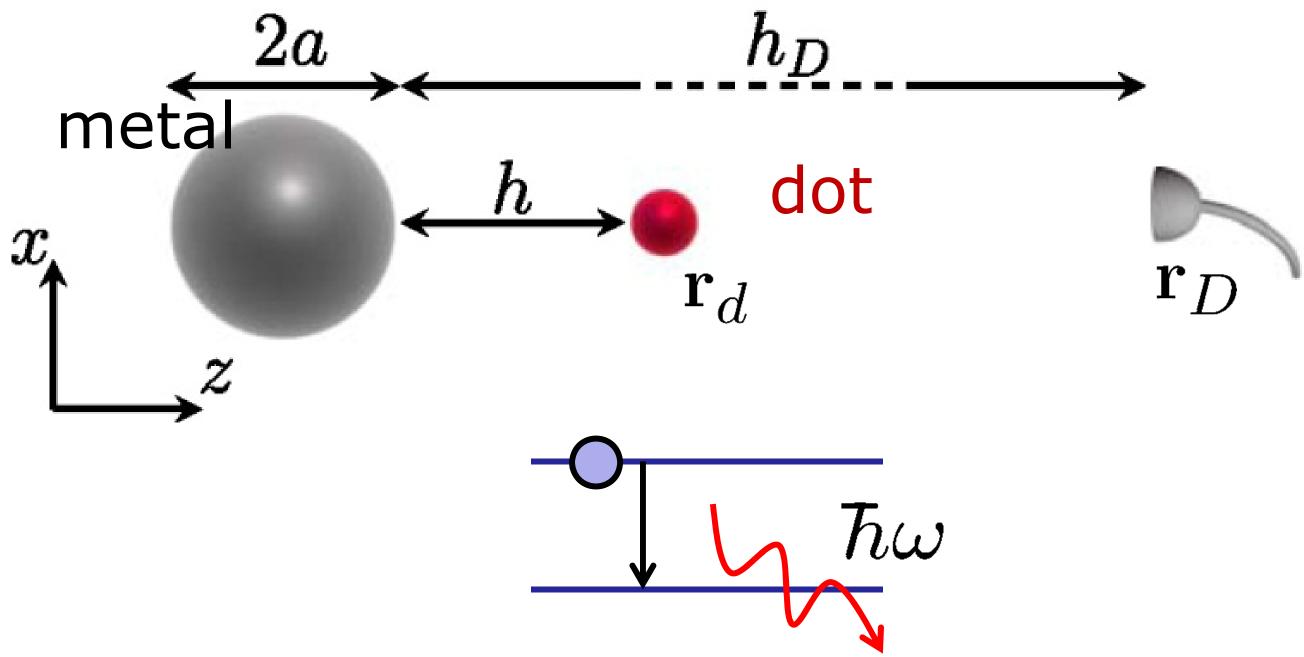
Enhanced spontaneous emission with plasmonic *nanoscale* resonators



Enhanced emission for various nanoantenna designs

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

Strong Coupling – vacuum Rabi splitting



Consider an electron-hole pair (exciton) coupled to a metal nanosphere, and we detect the light emission ar some detector outside the system

[1] Van Vlack, Kristensen, Van Vlack, Hughes, PRB (2012)

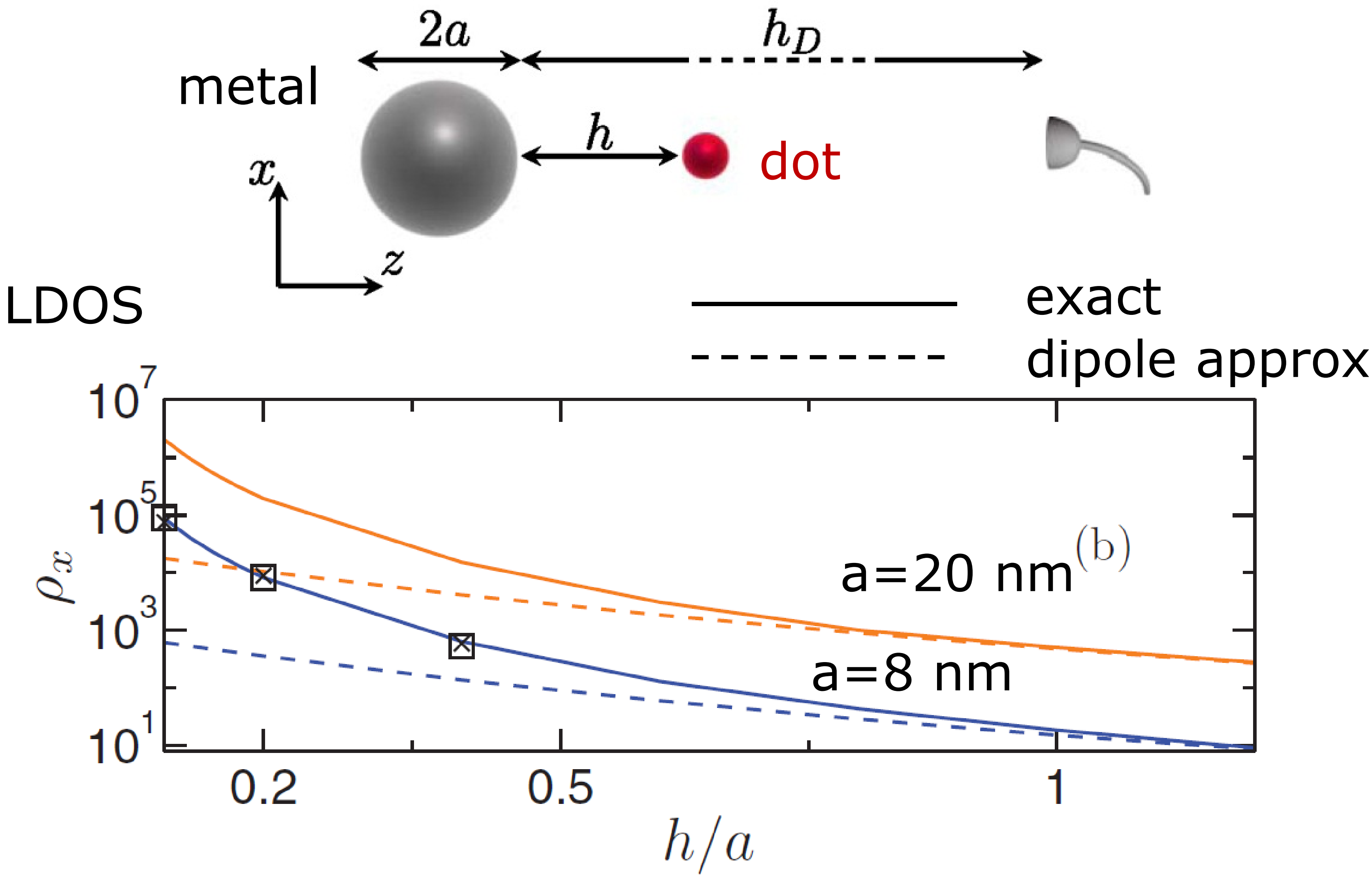
Metal nanoparticle

Spherical silver metal nanoparticle with Drude model for complex dielectric constant

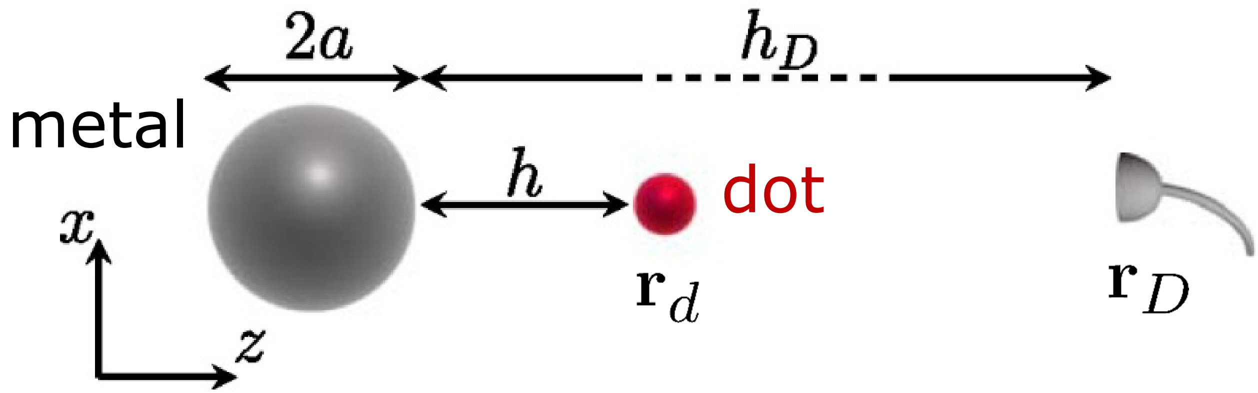
$$\epsilon(\omega) = 1 - \frac{\omega_p^2}{\omega(\omega + i\gamma)}$$

For a spherical structure, the Green function can be obtained analytically.

LDOS (near LSP) versus Height



Hamiltonian for any arbitrary bath



$$H = \int d\mathbf{r} \int_0^\infty d\omega \hbar\omega \hat{\mathbf{f}}^\dagger(\mathbf{r},\omega)\hat{\mathbf{f}}(\mathbf{r},\omega) + \hbar\omega_x \hat{\sigma}^+\hat{\sigma}^- - \left[i\sigma^+ \int_0^\infty d\omega\, \hat{\mathbf{E}}(\mathbf{r}_d,\omega) + \text{H.c.} \right]$$

[1] Van Vlack, Kristensen, Van Vlack, Hughes, PRB (2012)

Basic approach from Heisenberg EOM

$$\frac{d\hat{\sigma}^-}{dt} = -i\omega_l \hat{\sigma}^- + i\hbar^{-1}\mathbf{d}\cdot\hat{\mathbf{F}}(\mathbf{r}_d), \qquad (6)$$

$$\frac{d\hat{\mathbf{f}}_e(\mathbf{r},\omega_l)}{dt} = -i\omega_l \hat{\mathbf{f}}_e(\mathbf{r},\omega_l) - \sqrt{\frac{\varepsilon_I(\mathbf{r},\omega_l)}{\pi\hbar\varepsilon_0}}\boldsymbol{d}\cdot\mathbf{G}^*(\mathbf{r}_d,\mathbf{r};\omega_l)[\hat{\sigma}^- + \hat{\sigma}^+], \qquad (7)$$

We then make a Laplace transform and obtain an *exact* expression for the electric-field operator

$$\hat{\mathrm{E}}(\mathbf{r}_D,\omega) = \mathbf{G}(\mathbf{r}_D,\mathbf{r}_d,\omega)\cdot\mathrm{d}\,[\hat{\sigma}^-(\omega) + \hat{\sigma}^+(\omega)]$$

Spectrum from excited dot

$$S(\mathbf{r}_D,\omega) = \int dt_1 \int dt_2 \, \langle [\hat{\mathbf{E}}(\mathbf{r}_D,t_1)]^\dagger \hat{\mathbf{E}}(\mathbf{r}_D,t_2)\rangle$$

$$S(\mathbf{r}_D,\omega) \propto \left| \frac{\mathbf{G}(\mathbf{r}_D,\mathbf{r}_d,\omega)\cdot\mathbf{d}}{\omega_d^2-\omega^2-2\omega_d\mathbf{d}\cdot\mathbf{G}(\mathbf{r}_d,\mathbf{r}_d,\omega)\cdot\mathbf{d}/\hbar\varepsilon_0 - i\omega\Gamma_{\rm nr}} \right|^2$$

LDOS and non-local DOS or propagator:

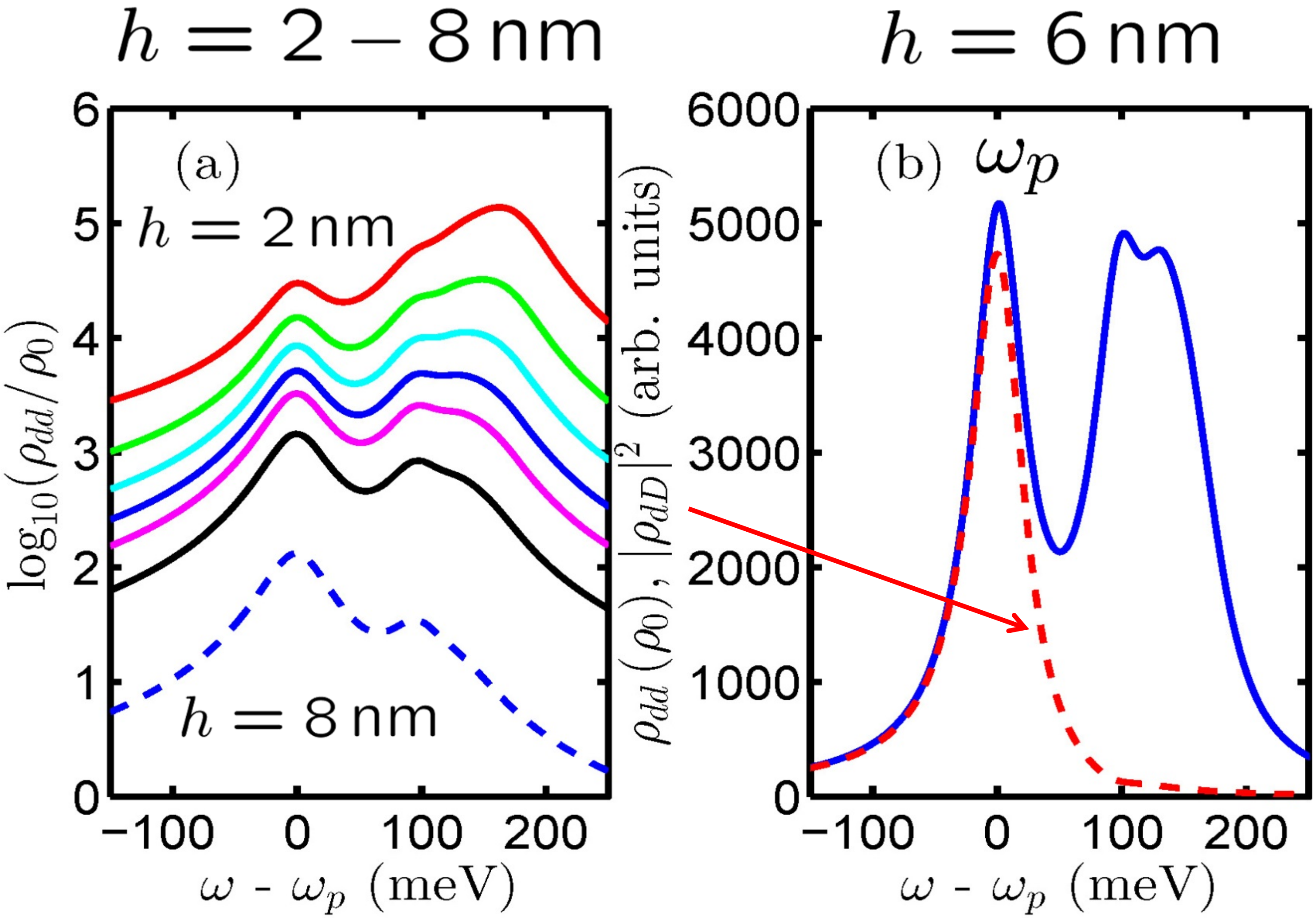
Also Exact!

$$\rho_{dd} \propto \mathbf{n}_d \cdot \mathrm{Im}\mathbf{G}(\mathbf{r}_d,\mathbf{r}_d;\omega)\cdot\mathbf{n}_d$$

$$\rho_{dD} \propto \mathbf{n}_d \cdot \mathbf{G}(\mathbf{r}_D,\mathbf{r}_d;\omega)\cdot\mathbf{n}_d$$

Ge, Van Vlack, Yao, Young, Hughes, PRB (2013)

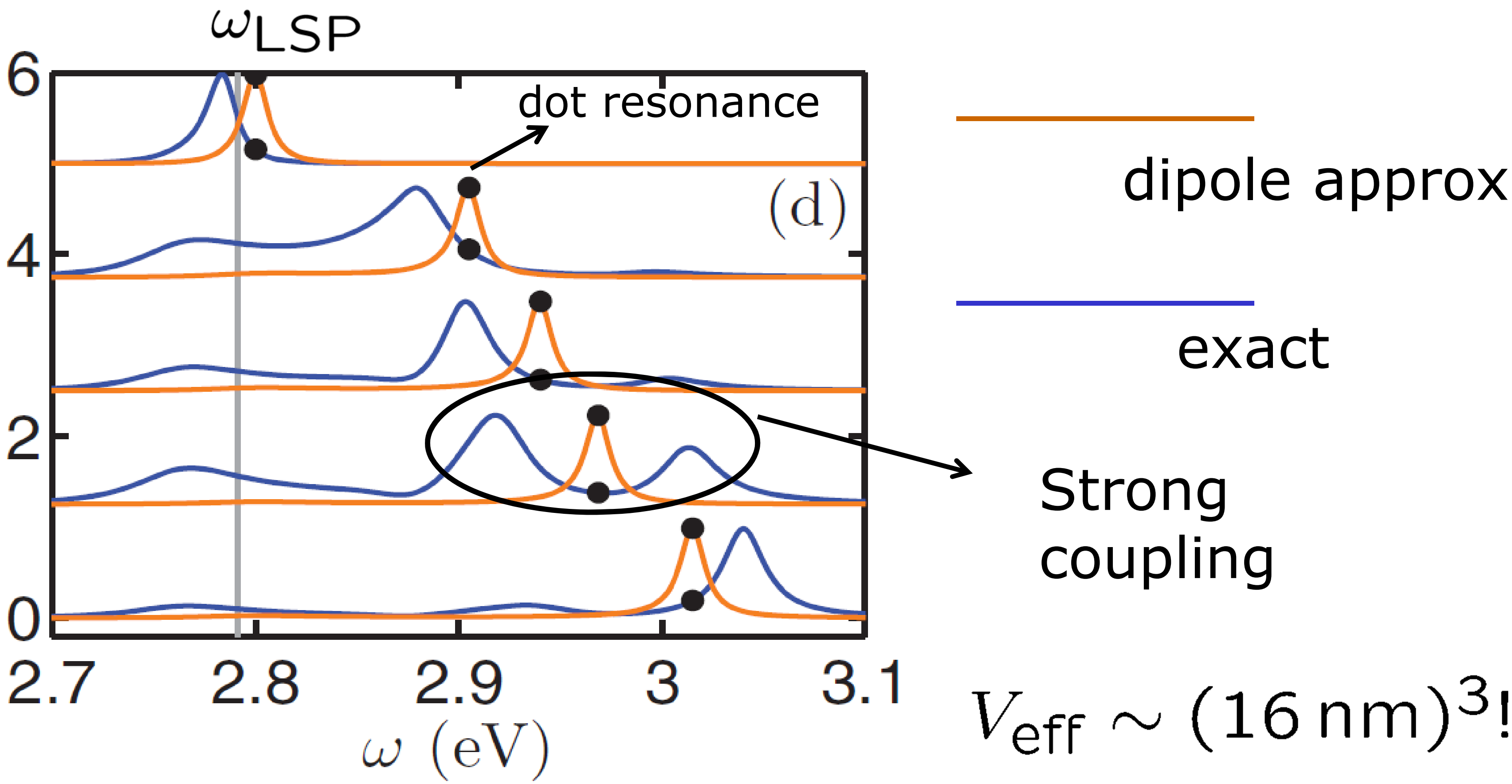
Green functions for 8nm-radius metal particle



Ge, Van Vlack, Yao, Young, Hughes, PRB (2013)

Far Field Spectrum, *h*=3 nm

$$S(\mathbf{r}_D,\omega) \propto \left| \frac{\mathbf{G}(\mathbf{r}_D,\mathbf{r}_d,\omega)\cdot\mathbf{d}}{\omega_d^2 - \omega^2 - 2\omega_d \mathbf{d}\cdot\mathbf{G}(\mathbf{r}_d,\mathbf{r}_d,\omega)\cdot\mathbf{d}/\hbar\varepsilon_0 - i\omega\Gamma_{\rm nr}} \right|^2$$



$V_{\rm eff} \sim (16\,{\rm nm})^3$!

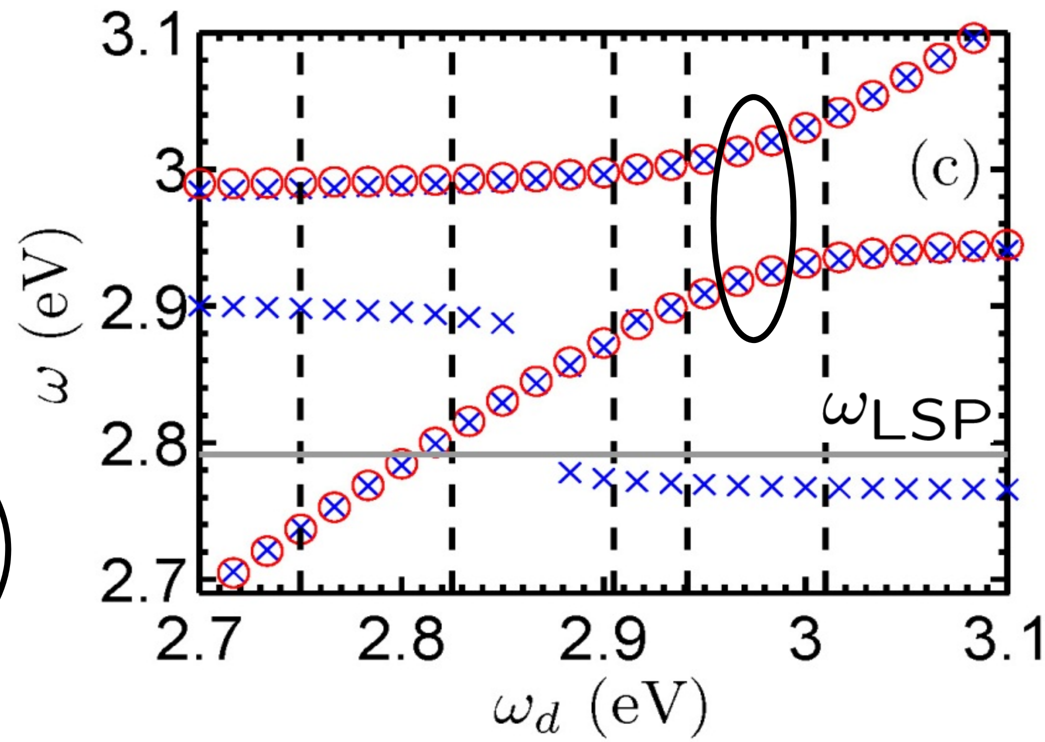
[1] Van Vlack, Kristensen, Hughes, PRB (2012)

Vacuum Rabi Splitting is huge!

20 nm metal particle
2 nm QD
4 nm separation

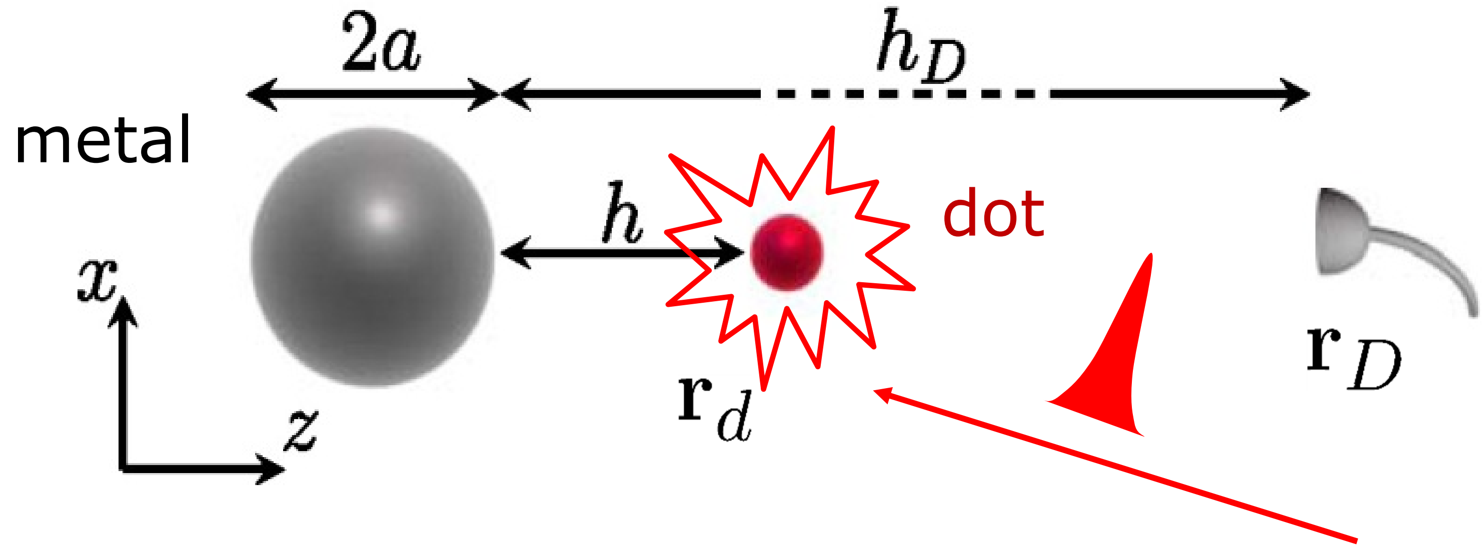
$$V \sim (16\,\mathrm{nm})^3 ! \ll \left(\frac{\lambda}{n}\right)^3$$

Dressed resonances



[1] Van Vlack, Kristensen, Hughes, PRB (2012)

Plasmon-modified Mollow triplets

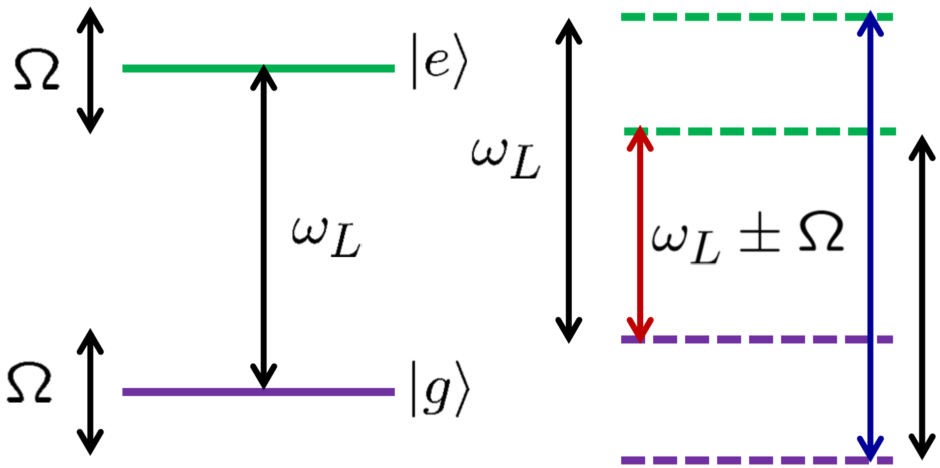


Ge, Van Vlack, Yao, Young, Hughes, PRB (2013)

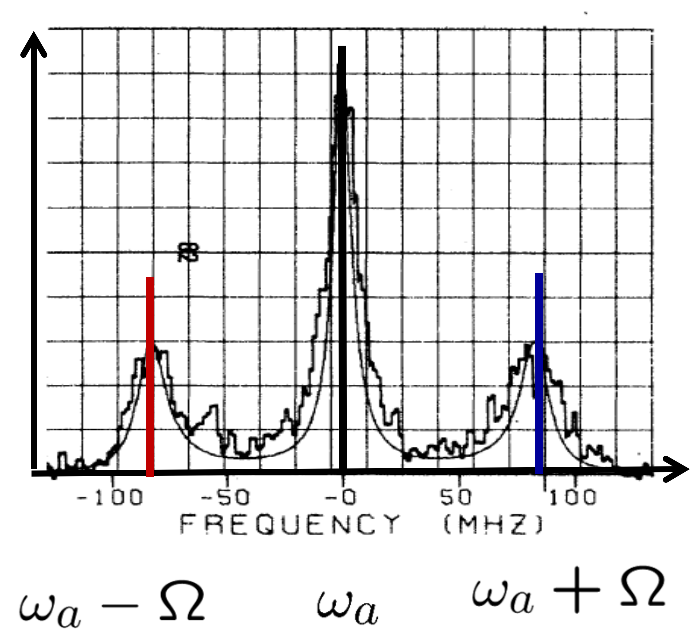
Simple Lorentz model: see also Ridolfo et al, PRL (2010)

Simple Mollow Triplet

Two level atom
+ CW Rabi field: $\Omega = dE$



* e.g., sodium atom:
Grove et al, PRA 1997



B.R.Mollow. "Power spectrum of light scattered by two-level systems," Phys. Rev. 188, 1969 (1969).

* also: Schuda et al, J. Phys. B 7, L198 (1974)

Light-Matter Hamiltonian

$$H = \int d\mathbf{r} \int_0^\infty d\omega\, \omega\, \hat{\mathbf{f}}^\dagger(\mathbf{r},\omega)\hat{\mathbf{f}}(\mathbf{r},\omega) + \omega_x \hat{\sigma}^+\hat{\sigma}^- - \left[\hat{\sigma}^+ \int_0^\infty d\omega\, \mathbf{d}\cdot\hat{\mathbf{E}}(\mathbf{r}_d,\omega) + \text{H.c.}\right] + H_{\text{drive}}$$

With: $$\hat{\mathbf{E}}(\mathbf{r},\omega) \propto \int d\mathbf{r}'\mathbf{G}(\mathbf{r},\mathbf{r}';\omega)\sqrt{\text{Im}[\varepsilon(\mathbf{r}',\omega)]}\hat{\mathbf{f}}(\mathbf{r}',\omega)$$

$$H_{\text{drive}} = \eta_x(\hat{\sigma}^+ e^{-i\omega_L} + \hat{\sigma}^- e^{i\omega_L})$$

This *H* applies to any inhomogeneous and lossy structure, including metal nanoparticles.

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e.g., S. Scheel, L. Knoll, D.-G. Welsch, PRA 60 4094 (1999)

Master equation with plasmonic bath

$$\frac{\partial \rho}{\partial t} = -i[H_S, \rho] + \mathcal{L}(\rho)$$

$$\int_0^t d\tau J_{\mathrm{ph}}(\tau)\left[-\hat{\sigma}^+\hat{\sigma}^-(-\tau)\rho + \hat{\sigma}^-(-\tau)\rho\hat{\sigma}^+\right] + H.c.$$

*Internal Coupling:

$$\hat{\sigma}^{\pm}(-\tau) = e^{-iH_S\tau}\hat{\sigma}^{\pm}e^{iH_S\tau}$$

$$H_s = \delta_{xL}\hat{\sigma}^+\hat{\sigma}^- + \eta_x(\hat{\sigma}^+ + \hat{\sigma}^-)$$

Photon bath correlation function:

$$J_{\mathrm{ph}}(\tau) = \int_0^\infty d\omega \frac{\mathbf{d}\cdot \mathrm{Im}[\mathbf{G}(\mathbf{r}_d, \mathbf{r}_d; \omega)]\cdot \mathbf{d}}{4\pi\hbar\varepsilon_0} e^{i(\omega_L - \omega)\tau}$$

*see H. Carmichael and D. Walls, J. Phys. A (1973)

Additional Dissipation and Spectrum

Background radiative decay and pure dephasing:

$$\mathcal{L}(\rho) = \frac{\gamma}{2} L(\hat{\sigma}^-, \rho) + \frac{\gamma'}{2} L(\hat{\sigma}^+ \hat{\sigma}^-, \rho)$$

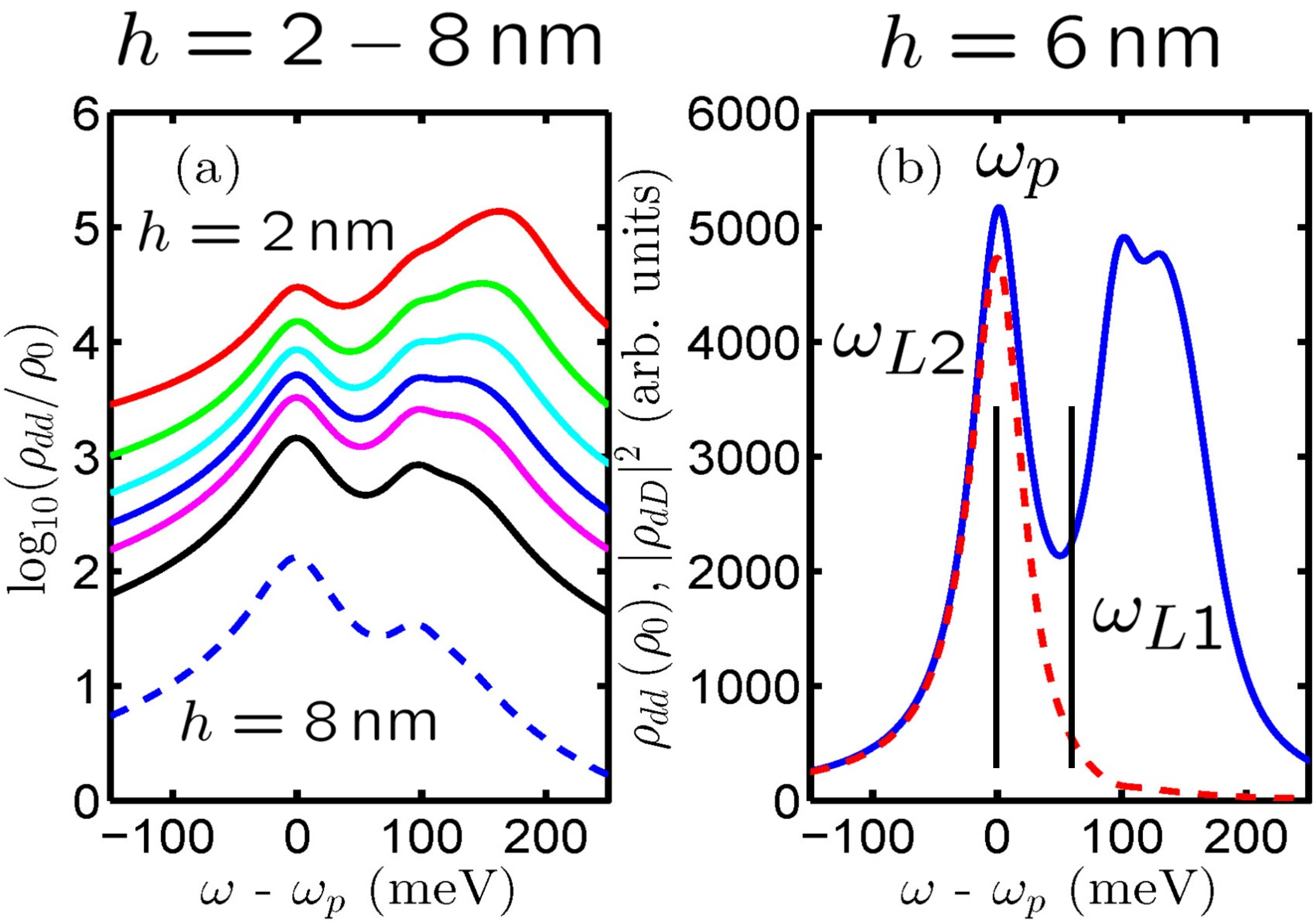
$$L(\hat{D}, \rho) = 2D\rho\hat{D}^\dagger - \hat{D}^\dagger\hat{D}\rho - \rho\hat{D}^\dagger\hat{D} \quad \text{(Lindblad)}$$

Fluorescence spectrum (exact):

$$S_p(\mathbf{R}, \omega) \propto |\mathbf{G}(\mathbf{R}, \mathbf{r}_d; \omega) \cdot \mathbf{d}|^2 S_0(\omega)$$

$$S_0(\omega) = \lim_{t \to \infty} \mathrm{Re}\left[\int_0^\infty d\tau \, \langle \hat{\sigma}^+(t+\tau)\hat{\sigma}^-(t) \rangle \, e^{i(\omega_L - \omega)\tau} \right]$$

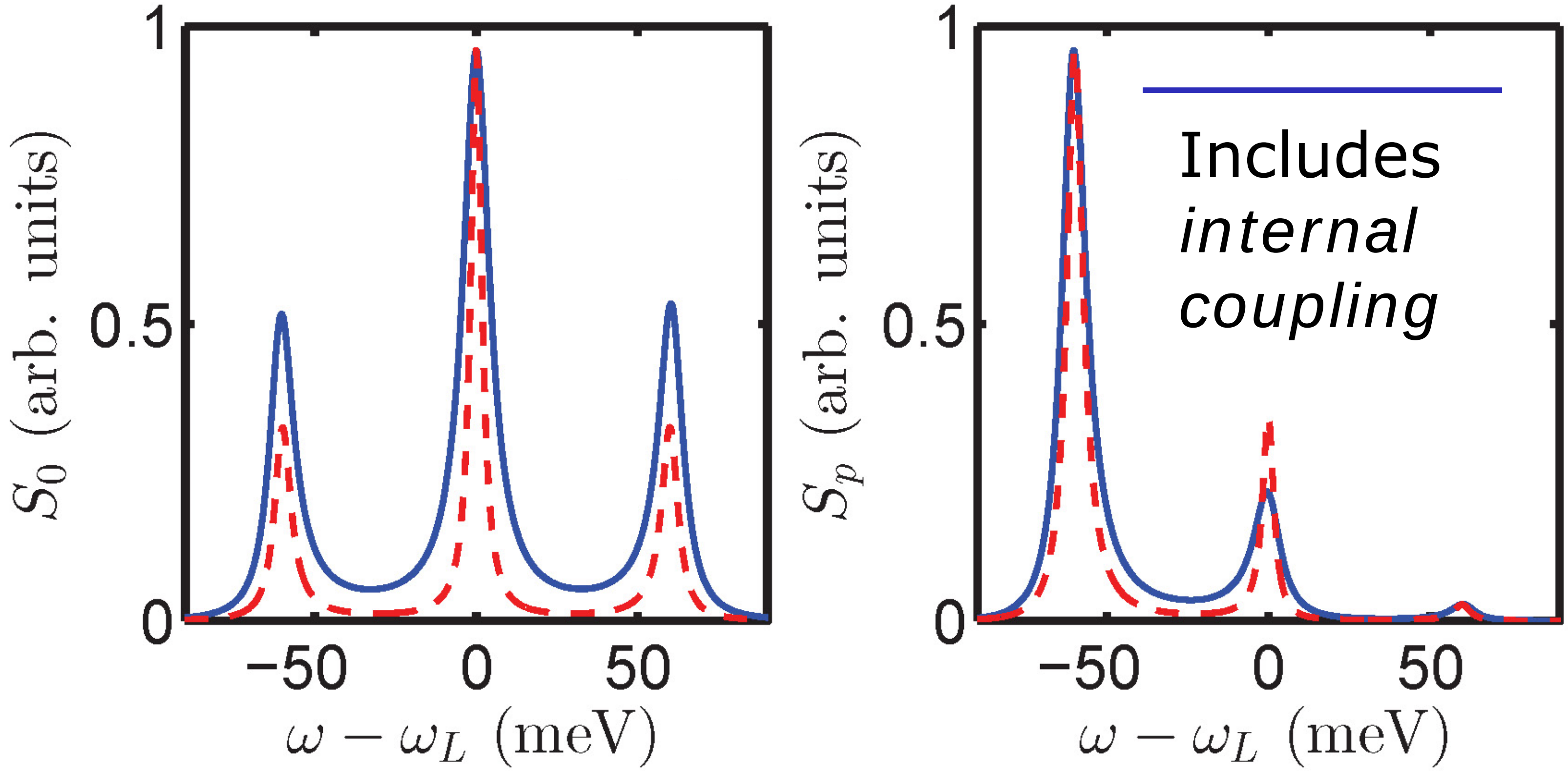
Green functions for 8nm-radius metal particle



Ge, Van Vlack, Yao, Young, Hughes, PRB (2013)

Driving field 60 meV blueshifted from LSP

$$\omega_{L1} = \omega_x = \omega_p + 60\,\text{meV}$$



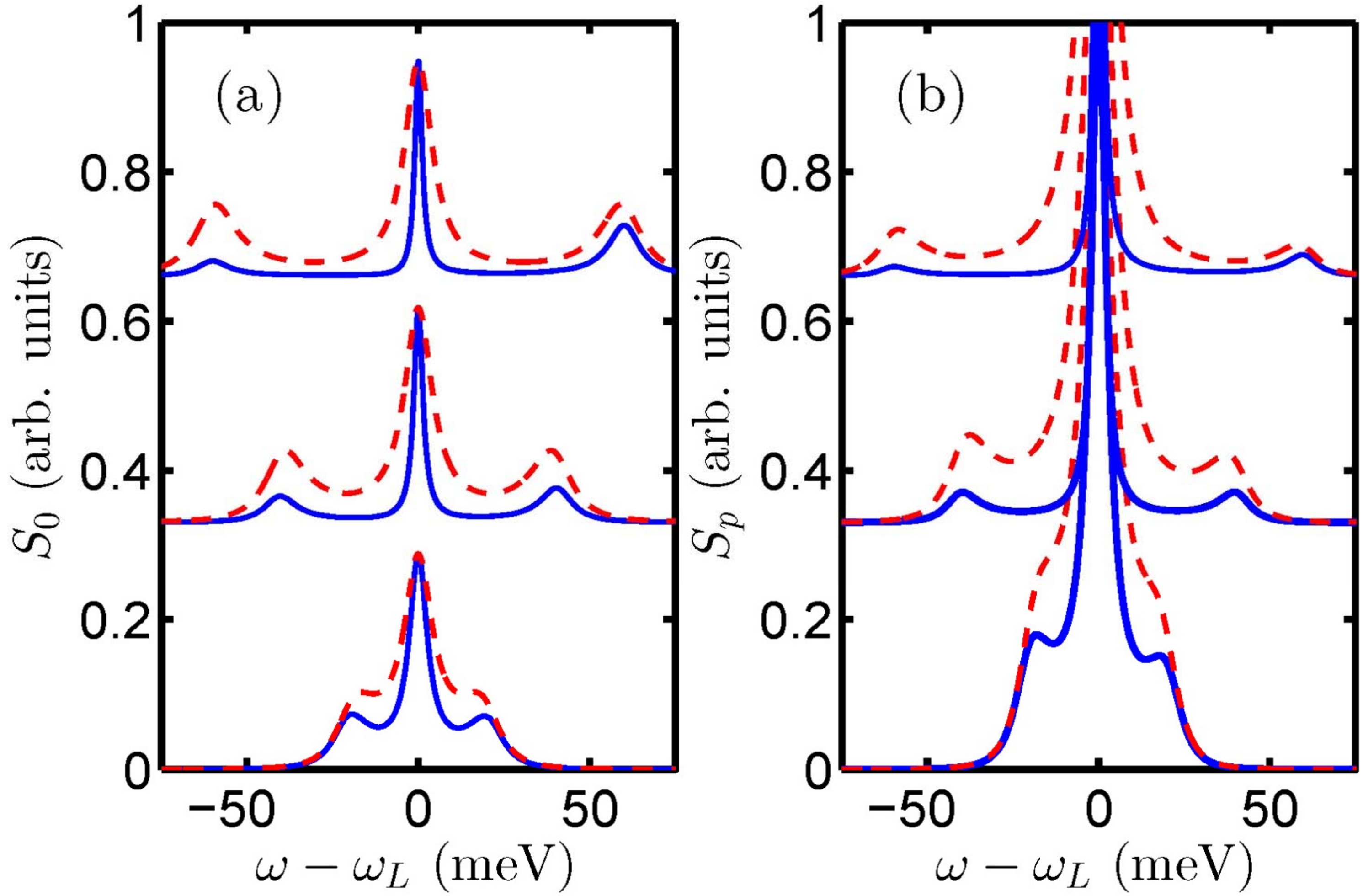
$$\Gamma_{\text{center}} \propto (LDOS(\omega_l - \Omega) + LDOS(\omega_l + \Omega))/2$$

$$\Gamma_{\text{side}} \propto (LDOS(\omega_l - \Omega) + LDOS(\omega_l + \Omega) + 4\,LDOS(\omega_l))/4$$

Ge, Van Vlack, Yao, Young, Hughes, PRB (2013)

Driving field on resonance with LSP

$\omega_{L2} = \omega_x = \omega_p \quad a = 8\,\text{nm},\ L = 6\,\text{nm}$



Ge, Van Vlack, Yao, Young, Hughes, PRB (2013)