TAILORING TWO-PHOTON SPONTANEOUS EMISSION USING ATOMICALLY THIN PLASMONIC NANOSTRUCTURES

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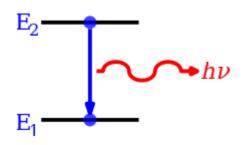




## A brief introduction about spontaneous emission

SPONTANEOUS EMISSION (SE)

• An **excited atom**, even when isolated, **decays** to its fundamental state.



- Phenomenon induced by **quantum vacuum fluctuations**.
- Quantum electrodynamics (QED): excited atom + zero photons is not a stationary state of the atom-field system.

# SEMost of the light we see is from SE.

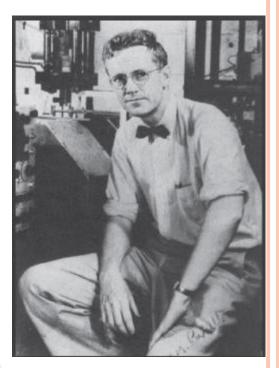






#### PURCELL EFFECT

- E.M. **Purcell** (**1946**): Bodies in the vicinities of an emitter change its SE rate.
- Reason: The presence of the bodies affects the **boundary conditions** (BC) on the electromagnetic field.

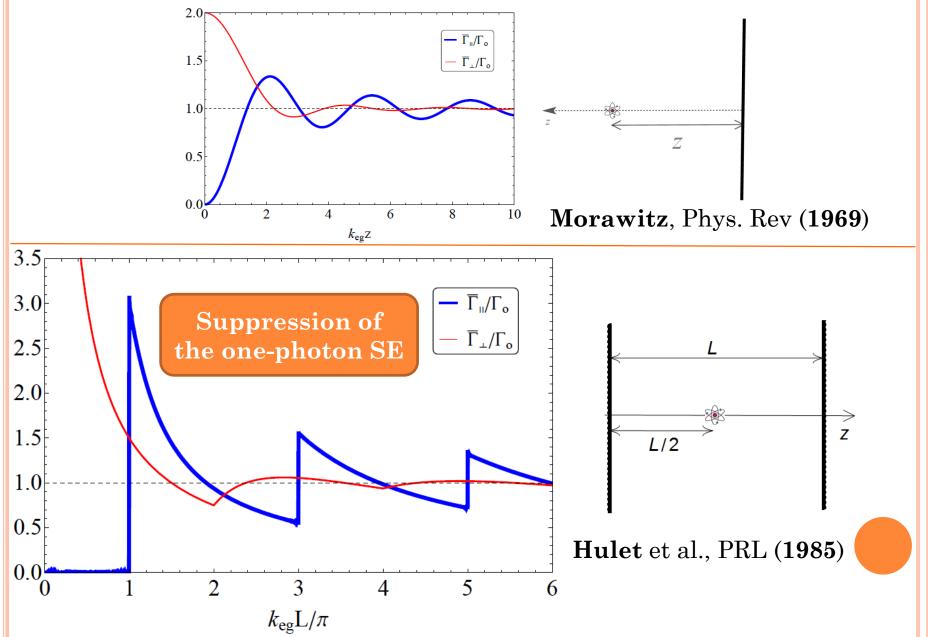


$$\Gamma(\mathbf{R}) = \frac{\pi}{\epsilon_o \hbar} \sum_{\mathbf{k}p} \omega_k |\mathbf{d}_{eg} \cdot \mathbf{A}_{\mathbf{k}p}(\mathbf{R})|^2 \delta(\omega_k - \omega_{eg})$$

• It can be shown that the SE rate is proportional to the local density of states (LDOS) of the electromagnetic field.

L. Novotny and B. Hecht, Principles of nano-optics. Cambridge university press, 2012.

## PURCELL EFFECT ON THE ONE-PHOTON SE



## TWO-PHOTON SPONTANEOUS EMISSION (TPSE)

- Second order process in perturbation theory (Göppert-Mayer, 1931).
- Relevant process when the one-photon SE is forbidden, for instance, due to **selection rules**.
- Ex: 2s 1s transition in H(Breit, Teller, 1940).  $\tau \approx 1/7s$
- Broadband spectrum of emission.
- Explains the emission spectrum of planetary nebulae. L. Spitzer and J. L. Greenstein, The Astrophysical Journal, vol. 114, p. 407 (1951).

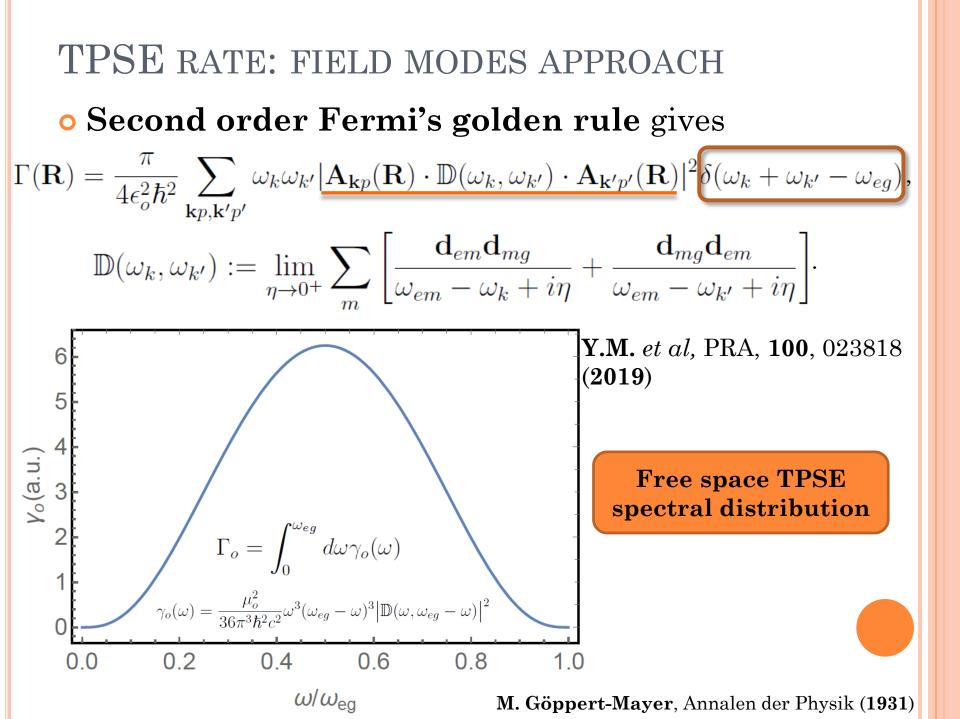
## PURCELL EFFECT ON THE TPSE

• Not widely discussed in the literature.

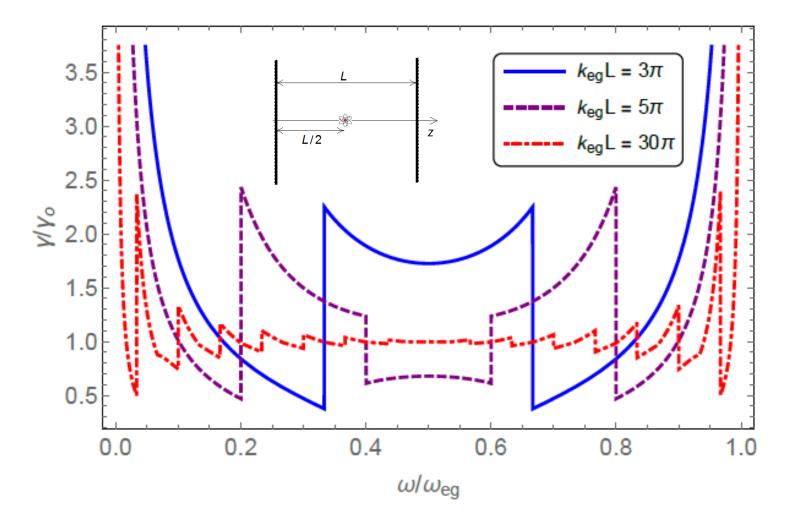
- The progress in **near-field optics, plasmonics,** and **materials science** in general has improved our **control** over **radiation-matter interactions**.
- In some situations the TPSE can even dominate conventionally fast transitions!

**N. Rivera** *et al*, "Making two-photon processes dominate one-photon processes using mid-ir phonon polaritons", PNAS, p. **201713538** (2017)

• TPSE is a rich phenomenon, with very much to be explored yet.

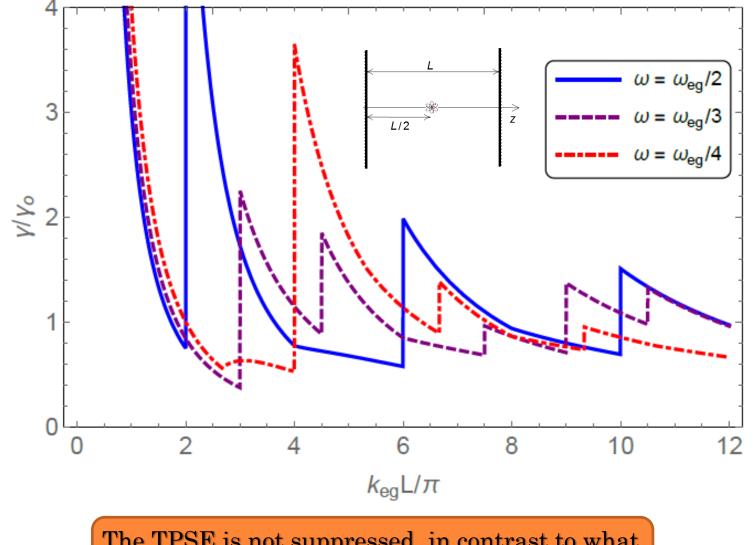


#### AN EMITTER BETWEEN TWO PERFECT MIRRORS (S $\rightarrow$ S)



Abrupt changes in the spectral density due to discontinuities in the LDOS.

#### AN EMITTER BETWEEN TWO PERFECT MIRRORS (S $\rightarrow$ S)



The TPSE is not suppressed, in contrast to what happens to the one-photon SE in this situation.

RELATION BETWEEN TPSE AND ONE-PHOTON SE• It is possible to show that

$$\Gamma(\mathbf{R}_e) = \int_0^{\omega_t} d\omega \gamma_0(\omega) \sum_{a,b} t_{ab}(\omega) P_a(\mathbf{R}_e, \omega) P_b(\mathbf{R}_e, \omega_t - \omega)$$

$$t_{ab}(\omega) = |\mathbb{D}_{ab}(\omega, \omega_t - \omega)|^2 / |\mathbb{D}(\omega, \omega_t - \omega)|^2$$

 $P_a(\mathbf{R}_e,\omega)$ 

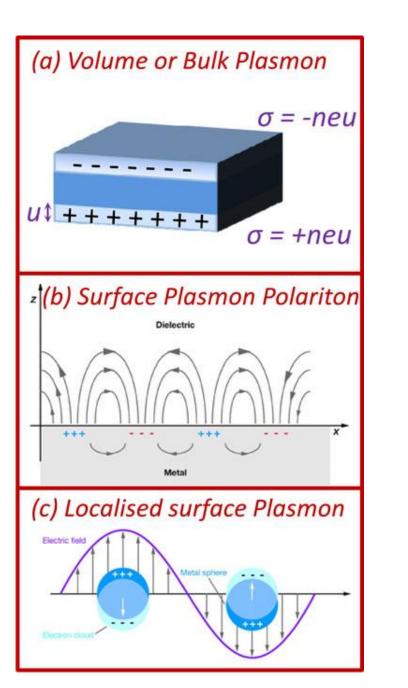
Purcell factor for an emitter at  $\mathbf{R}_e$ , with transition dipole moment oriented along  $\hat{\mathbf{e}}_a(\mathbf{R}_e, \omega)$ , and frequency  $\omega$ .

• Once we know the **one-photon SE** rate of an emitter we can obtain immediately the **TPSE spectral density**!

## TPSE near plasmonic nanostructures

## PLASMONICS

## • What is a plasmon?



## PLASMONICS

• What is plasmonics?

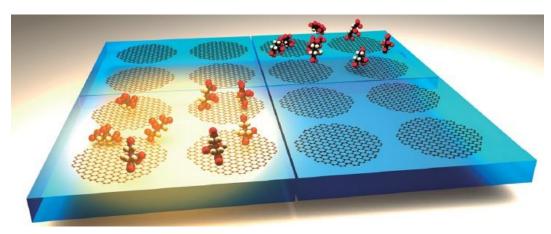
"You just have Maxwell's equations, some material properties and some boundary conditions, all classical stuff - what's new about that?"

**S. A. Maier**, *Plasmonics: fundamentals and applications*.

#### **Physics!**

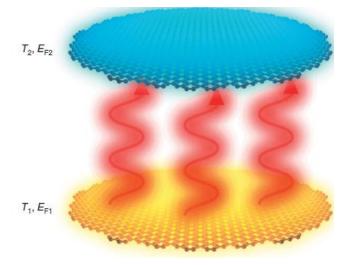
- Strong light confinement  $\rightarrow$  beyond the diffraction limit.
- Extreme enhancement of the electromagnetic field intensity → surface physics and **non-linear optics**.

# PLASMONICS IN 2D SYSTEMS - GRAPHENE • Spatially resolved optical sensing in the infrared



•ACS Photonics 2017, 4, 1831–1838 •ACS Photonics 2018, 5, 8, 3282-3290

• Ultrafast radiative heat transfer

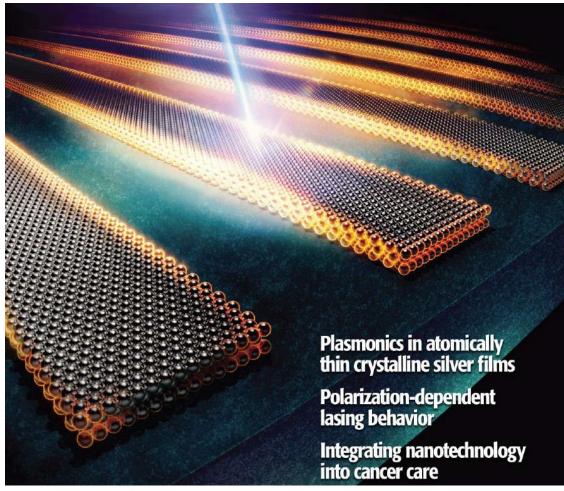


Nature Communications, 8, 2 (2017)

## (QUASI-)2D NOBLE METALS

• Wide range of frequencies (visible and near-infrared)

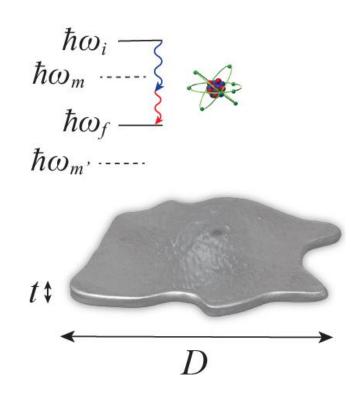
• Recent fabrication of quasi-2D metal films.



•ACS NANO,**13**, 7 (2019) •Nature Photonics, **8**, 328-333 (2019)

## System under study

• An emitter near a 2D plasmonic nanostructure of arbitrary geometry.



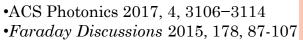
## PLASMONS IN 2D NANOSTRUCTURES

• Plasmon Wave Function (**PWF**) formalism:

$$\rho_{2D}(\mathbf{r},\omega) = \frac{4\pi\epsilon_0}{D} \sum_j \frac{c_j}{1/\eta_j - 1/\eta(\omega)} v_j(\mathbf{u}) \longrightarrow Plasmon \ Wave \ Function \ j$$

• Resonance frequencies:

 $\operatorname{Re}[1/\eta_j - 1/\eta(\omega_j)] = 0 \qquad \eta(\omega) = i\sigma(\omega)/4\pi\epsilon_0\omega D$ 



#### PLASMONS IN 2D NANOSTRUCTURES

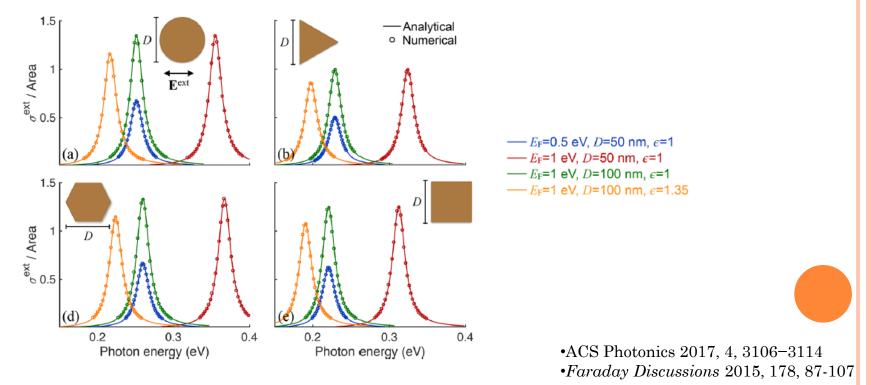
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• Resonance frequencies:

 $\operatorname{Re}[1/\eta_j - 1/\eta(\omega_j)] = 0 \qquad \eta(\omega) = i\sigma(\omega)/4\pi\epsilon_0\omega D$ 

• Excelent agreement with numerical calculations.



## PURCELL FACTORS

• The Purcell factors are numerically equal to the ratio between the power dissipated by an electric dipole near the nanostructure with respect to its free-space radiation rate.

 $P_a(\mathbf{R}_e,\omega) = W_a(\mathbf{R}_e,\omega)/W_0(\omega)$ 

## PURCELL FACTORS

• The Purcell factors are numerically equal to the ratio between the power dissipated by an electric dipole near the nanostructure with respect to its free-space radiation rate.

$$P_a(\mathbf{R}_e,\omega) = W_a(\mathbf{R}_e,\omega)/W_0(\omega)$$

$$P_a(\mathbf{R}_e, \omega) = P_{a,nr}(\mathbf{R}_e, \omega) + P_{a,r}(\mathbf{R}_e, \omega)$$

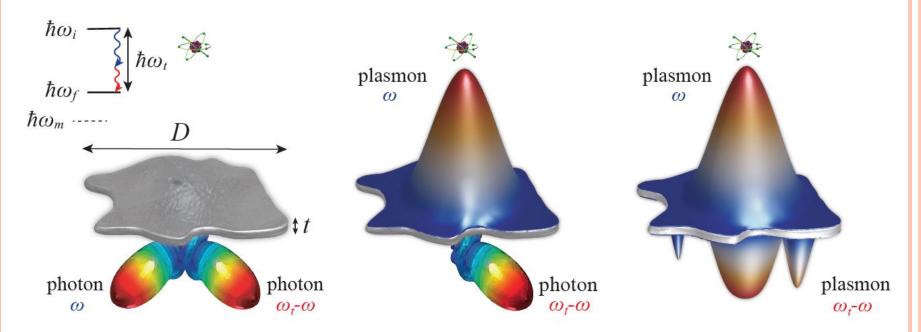
• Absorption (**plasmon emission**):

$$P_{a,nr}(\mathbf{R}_{e},\omega) = \frac{6\pi\epsilon_{0}c^{3}}{\omega^{4}|\mathbf{d}_{a}|^{2}} \int d^{3}\mathbf{R}' \operatorname{Re}\{\mathbf{J}^{*}(\mathbf{R}',\omega) \cdot \mathbf{E}(\mathbf{R}',\omega)\}$$

• Far-field radiation (**photon emission**):

$$P_{a,r}(\mathbf{R}_e,\omega) = \frac{6\pi\epsilon_0 c^3}{\omega^4 |\mathbf{d}_a|^2} \int_{R'\to\infty} d\mathbf{A}' \cdot \operatorname{Re}\{\mathbf{E}(\mathbf{R}',\omega) \times \mathbf{H}^*(\mathbf{R}',\omega)\}$$

## TPSE DECAY CHANNELS



$$\gamma(\mathbf{R}_{e},\omega) = \gamma_{0}(\omega) \sum_{a,b} t_{ab}(\omega) P_{a}(\mathbf{R}_{e},\omega) P_{b}(\mathbf{R}_{e},\omega_{t}-\omega)$$
$$P_{a}(\mathbf{R}_{e},\omega) = P_{a,nr}(\mathbf{R}_{e},\omega) + P_{a,r}(\mathbf{R}_{e},\omega)$$

• Photon-photon, photon-plasmon and plasmon-plasmon states.

#### APPROXIMATED PURCELL FACTORS: DRUDE MODEL

$$P_{a,nr}(\mathbf{R}_e,\omega) \simeq \sum_{q=1}^{N} \frac{A_{a,q}}{\omega^2} \frac{1/2\tau}{(\omega-\omega_q)^2 + (1/2\tau)^2}$$

Plasmonic contribution  $\rightarrow$  Lorentzian resonances

#### APPROXIMATED PURCELL FACTORS: DRUDE MODEL

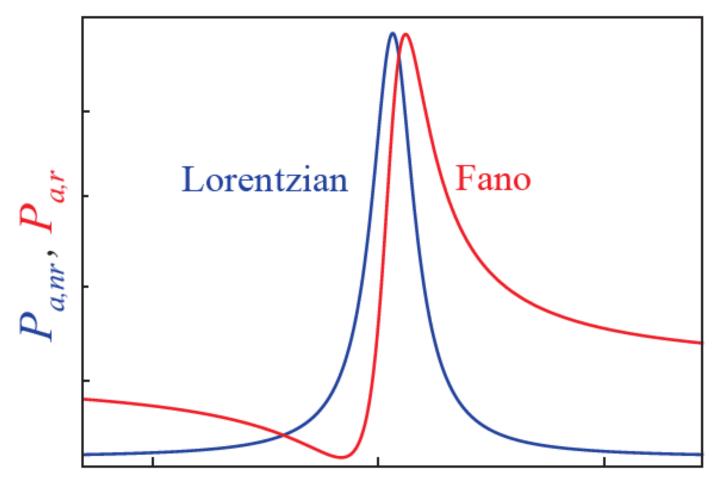
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Plasmonic contribution  $\rightarrow$  Lorentzian resonances

$$P_{a,r}(\omega) \simeq \sum_{q=1}^{N} \frac{B_{a,q}(1/2\tau)^2 + (\omega - \omega_q + f_{a,q}/2\tau)^2}{(\omega - \omega_q)^2 + (1/2\tau)^2} - (N-1)$$

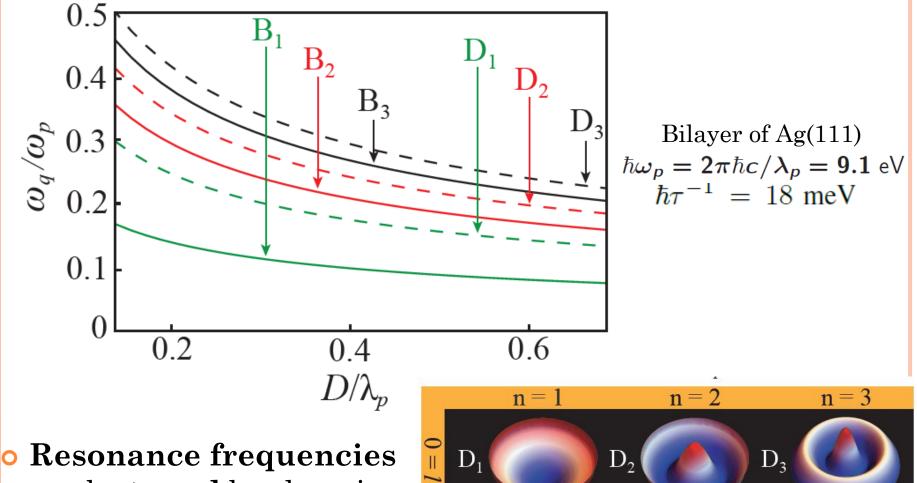
Photonic contribution  $\rightarrow$  Fano + Lorentzian resonances

## APPROXIMATED RESULTS: DRUDE MODEL



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#### A METALLIC NANODISK CLOSE TO AN ON-AXIS QUANTUM EMITTER: RESONANT MODES



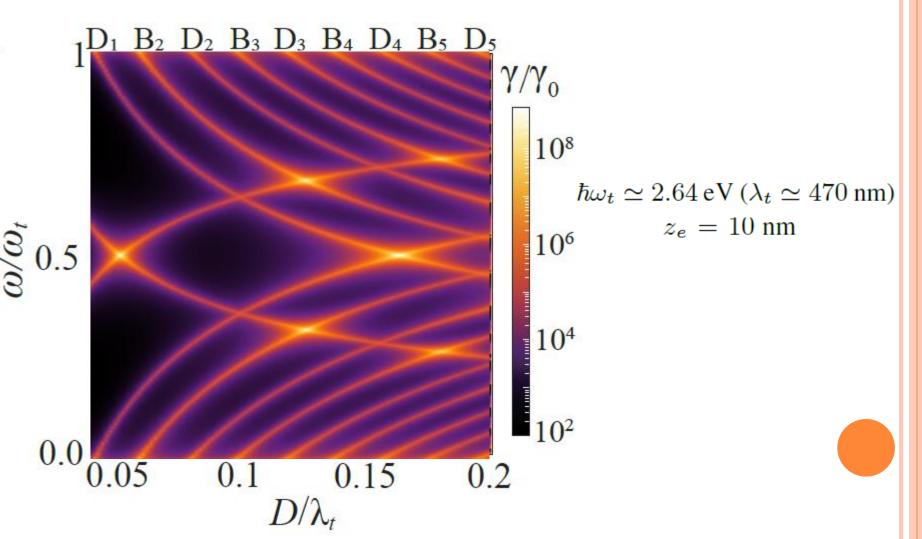
 $B_1$ 

 $B_2$ 

B,

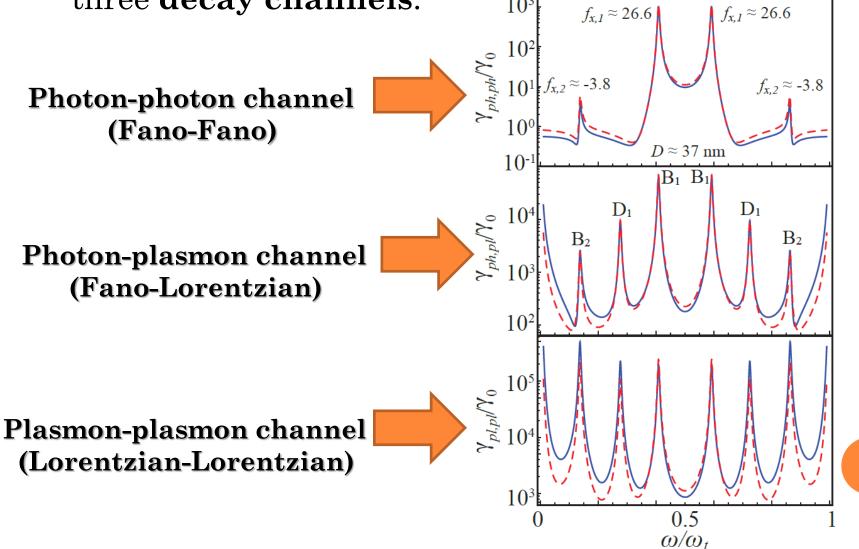
can be **tuned** by changing the **size** of the nanodisk. A METALLIC NANODISK CLOSE TO AN ON-AXIS QUANTUM EMITTER: TPSE SPECTRUM

• Crossings between Bright-Bright or Dark-Dark modes produce extreme enhancements.

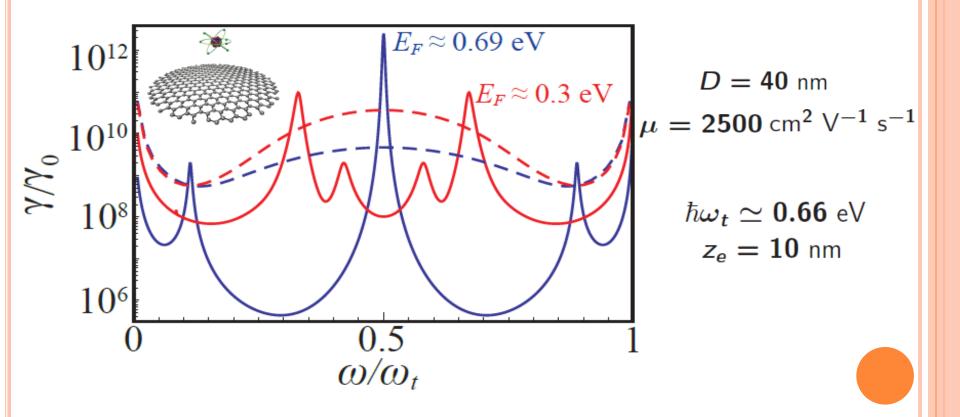


A METALLIC NANODISK CLOSE TO AN ON-AXIS QUANTUM EMITTER: DECAY CHANNELS

• Spectral line-shapes serve as **fingerprints** of the three **decay channels**.



• Enhanced selective spectral emission compared to the typical broadband spectrum of monolayers.

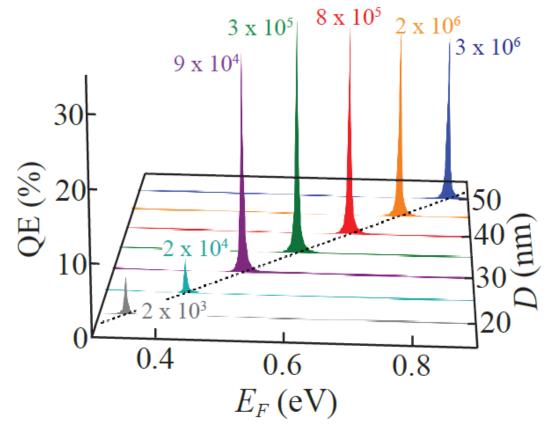


$$\begin{aligned} \mathbf{Q}\mathbf{Y}^{1q}(\omega) &= \frac{\gamma_{ph}^{1q}(\omega)}{\gamma^{1q}(\omega)} , \ \mathbf{Q}\mathbf{Y}^{\mathrm{TPSE}}(\omega) &= \frac{\gamma_{ph,ph}(\omega) + \gamma_{ph,pl}(\omega)}{\gamma(\omega)} \\ & 1 \\ & 1 \\ & 1 \\ & 0.5 \\ & 0.5 \\ & 0.5 \\ & 0.6 \\ & 0.4 \\ & 0.6 \\ & 0.8 \\ & 1.0 \\ & 1.2 \\ & E_F/\hbar\omega_t \end{aligned}$$
 The fundamental dark mode acts as an amplifier of the photon-plasmon channel, but as an attenuator of the one-photon emission pathway.

 Single photon creation via a two-quanta process can be much more efficient than standard one-photon emission.

$$QE = \Gamma_{4s \to 3s} / (\Gamma_{4s \to 3s} + \gamma_{4s \to 3p}^{1q} + \gamma_{4s \to 2p}^{1q})$$

• For any disk diameter the quantum efficiency can be optimized by tuning the Fermi energy so that  $\omega_{B_1} = \omega_t/2$ .



$$\mu = 10^4$$
 cm $^2$  V $^{-1}$  s $^{-1}$ 

Numbers near curves: **ph-ph Purcell factors** 

## CONCLUSIONS

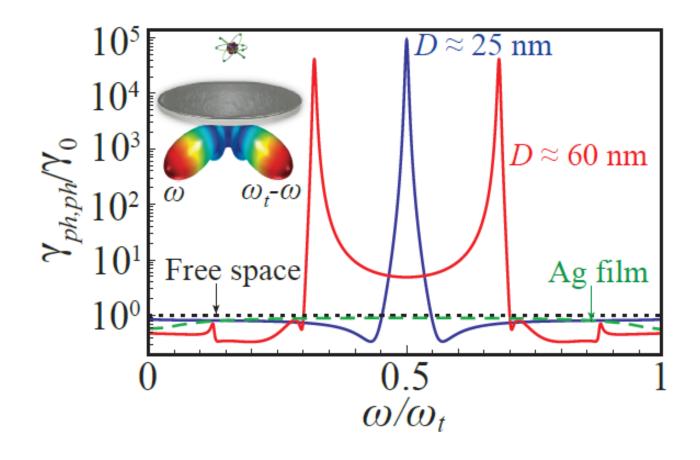
- It is possible to pre-select the frequencies of emission by tuning the size and doping of the nanostructure.
- 2D plasmonic nanostructures allow enhanced TPSE rate with **generation of photons**, not only plasmons.
- Surprisingly, **TPSE** can be a **single photon source** orders of magnitude more efficient than one-photon SE.
- Finite-sized plasmonic systems have many advantages over extended ones.

# THANK YOU



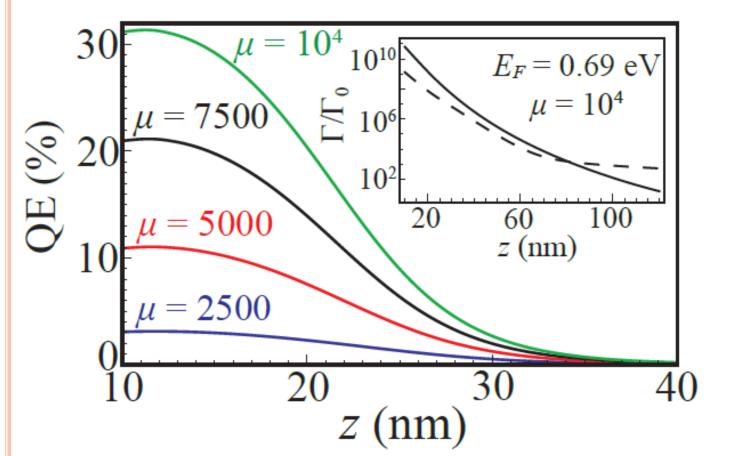
A METALLIC NANODISK CLOSE TO AN ON-AXIS QUANTUM EMITTER: PH-PH DECAY CHANNEL

• Finite size is critical to accomplish giant photonphoton production.



• Graphene nanostructures disrupt the usual unbalance between one- and two-quanta emission.

$$QE = \Gamma_{4s \to 3s} / (\Gamma_{4s \to 3s} + \gamma_{4s \to 3p}^{1q} + \gamma_{4s \to 2p}^{1q})$$



#### PURCELL EFFECT

- E.M. **Purcell** (**1946**): bodies in the vicinities of an emitter change its SE rate.
- Reason: the presence of the bodies affects the **boundary conditions** (BC) on the electromagnetic field.

$$\Gamma(\mathbf{R}) = \frac{\pi}{\epsilon_o \hbar} \sum_{\mathbf{k}p} \omega_k |\mathbf{d}_{eg} \cdot \mathbf{A}_{\mathbf{k}p}(\mathbf{R})|^2 \delta(\omega_k - \omega_{eg}).$$
$$\frac{\Gamma}{\Gamma_o} = \frac{6\pi c}{\omega_{eg}} \mathbf{\hat{n}}_{eg}^* \cdot \left[ \mathrm{Im}\mathbb{G}(\mathbf{R}, \mathbf{R}, \omega_{eg}) \right] \cdot \mathbf{\hat{n}}_{eg}$$
$$\nabla \times \nabla \times \mathbb{G}(\mathbf{r}, \mathbf{r}', \omega) - \frac{\omega^2}{c^2} \mathbb{G}(\mathbf{r}, \mathbf{r}', \omega) = \mathbb{I}\delta(\mathbf{r} - \mathbf{r}').$$

L. Novotny and B. Hecht, Principles of nano-optics. Cambridge university press, 2012.



## GREEN'S FUNCTION METHOD

• The imaginary part of the Green's function can be written in terms of the field modes as

Im 
$$\mathbb{G}(\mathbf{r}, \mathbf{r}', \omega) = \frac{\pi c^2}{2\omega} \sum_{\mathbf{k}p} \mathbf{A}^*_{\mathbf{k}p}(\mathbf{r}') \mathbf{A}_{\mathbf{k}p}(\mathbf{r}) \delta(\omega - \omega_k).$$

• Using the previous identity, we recover the well known expression for the TPSE rate, namely

$$\Gamma = \frac{\mu_0^2}{\pi\hbar^2} \int_0^{\omega_{eg}} d\omega \omega^2 (\omega_{eg} - \omega)^2 \mathrm{Im}\mathbb{G}_{il}(\omega) \mathrm{Im}\mathbb{G}_{jn}(\omega_{eg} - \omega)\mathbb{D}_{ij}(\omega, \omega_{eg} - \omega)\mathbb{D}_{ln}^*(\omega, \omega_{eg} - \omega).$$

N. Rivera et al., Science, vol. 353, no. 6296, pp. 263–269 (2016).

• This constitutes an **alternative demonstration** of this formula!

## THE PURCELL FACTORS RELATION

• Choosing the basis which diagonalizes the Green's function we have

$$\gamma(\omega) = \frac{\mu_0^2}{\pi\hbar^2} \omega^2 (\omega_{eg} - \omega)^2 \sum_{i,j} \operatorname{Im}\mathbb{G}_{ii}(\omega) \operatorname{Im}\mathbb{G}_{jj}(\omega_{eg} - \omega) |\mathbb{D}_{ij}(\omega, \omega_{eg} - \omega)|^2$$

• We define the **Purcell factors** as

$$P_i(\mathbf{R},\omega) := \frac{6\pi c}{\omega} \mathrm{Im}\mathbb{G}_{ii}(\mathbf{R},\mathbf{R},\omega).$$

• In this way, we can write

$$\frac{\gamma(\omega)}{\gamma_o(\omega)} = \sum_{i,j} \frac{|\mathbb{D}_{ij}(\omega, \omega_{eg} - \omega)|^2}{|\mathbb{D}(\omega, \omega_{eg} - \omega)|^2} P_i(\omega) P_j(\omega_{eg} - \omega).$$

• The **TPSE** rate **dependence** on the **local density of states** (LDOS) was made explicit! PLASMONS IN 2D NANOESTRUCTURES

• Plasmon Wave Function (**PWF**) formalism:

$$\rho_{2D}(\mathbf{r},\omega) = \frac{4\pi\epsilon_0}{D} \sum_j \frac{c_j}{1/\eta_j - 1/\eta(\omega)} v_j(\mathbf{u}),$$

$$\begin{split} v_j(\mathbf{u}) &= \nabla_{\mathbf{u}} \cdot \sqrt{f(\mathbf{u})} \mathbf{V}_j(\mathbf{u}) \not \rightarrow \textit{Plasmon Wave Functions} \\ \int d^2 \mathbf{u}' \, \mathbb{M}(\mathbf{u}, \mathbf{u}') \cdot \mathbf{V}_j(\mathbf{u}') &= \frac{1}{\eta_j} \mathbf{V}_j(\mathbf{u}) \, . \\ \mathbb{M}(\mathbf{u}, \mathbf{u}') &= \sqrt{f(\mathbf{u}) f(\mathbf{u}')} \nabla_{\mathbf{u}} \nabla_{\mathbf{u}'} |\mathbf{u} - \mathbf{u}'|^{-1} \end{split}$$

• Resonance frequencies:

 $\operatorname{Re}[1/\eta_j - 1/\eta(\omega_j)] = 0 \qquad \eta(\omega) = i\sigma(\omega)/4\pi\epsilon_0\omega D$ 

• External field dependence:

$$c_j = \int d^2 \mathbf{u} \, \mathbf{V}_j^*(\mathbf{u}) \cdot \boldsymbol{\mathcal{E}}^{ext}(\mathbf{u}, \omega)$$

•ACS Photonics 2017, 4, 3106–3114 •Faraday Discussions 2015, 178, 87-107

## POWER DISSIPATED BY ABSORPTION

$$\mathbf{J}(\mathbf{R}',\omega) = \mathbf{K}(\mathbf{r}',\omega)\delta(z') = \sigma(\omega)f(\mathbf{r}')\mathbf{E}_{\parallel}(\mathbf{r}',\omega)\delta(z')$$

$$+$$

$$\boldsymbol{\mathcal{E}}(\mathbf{u},\omega) = \sum_{\alpha} \frac{c_{\alpha}}{1-\eta(\omega)/\eta_{\alpha}} \mathbf{V}_{\alpha}(\mathbf{u}), c_{\alpha} = \int d^{2}\mathbf{u} \mathbf{V}_{\alpha}^{*}(\mathbf{u}) \cdot \boldsymbol{\mathcal{E}}^{ext}(\mathbf{u},\omega)$$

$$\mathbf{E}^{ext}(\mathbf{R}',\omega) = \frac{1}{4\pi\epsilon_{0}} \nabla \mathbf{d}_{a} \cdot \nabla |\mathbf{R} - \mathbf{R}'|^{-1}$$

$$+$$

$$\int d^{2}\mathbf{u} \mathbf{V}_{\alpha}^{*}(\mathbf{u}) \cdot \mathbf{V}_{\alpha'}(\mathbf{u}) = \delta_{\alpha\alpha'}.$$

$$P_{a,nr}(\mathbf{R}_{e},\omega) = \frac{3c^{3}}{2D^{3}\omega^{3}} \operatorname{Im} \sum_{\alpha} \hat{\mathbf{e}}_{a} \cdot \frac{\mathbf{F}_{\alpha}(\mathbf{R}_{e}) \otimes \mathbf{F}_{\alpha}^{*}(\mathbf{R}_{e})}{1/\eta(\omega) - 1/\eta_{\alpha}} \cdot \hat{\mathbf{e}}_{a}.$$

$$\mathbf{F}_{\alpha}(\mathbf{R}_{e}) = \int d^{2}\mathbf{u}' \frac{v_{\alpha}(\mathbf{u}')(\mathbf{R}_{e}/D - \mathbf{u}')}{|\mathbf{R}_{e}/D - \mathbf{u}'|^{3}}$$

## POWER DISSIPATED BY RADIATION

• The system is spatially localized, therefore we can make a multipole expansion. The first contribution to the power radiated by the system is

$$\begin{split} P_{a,r}(\mathbf{R}_{e},\omega) \simeq \frac{|\mathbf{d}_{a} + \mathbf{d}_{a,ind}(\mathbf{R}_{e},\omega)|^{2}}{|\mathbf{d}_{a}|^{2}} \\ \mathbf{d}_{a,ind}(\mathbf{R}_{e},\omega) &= \int d^{2}\mathbf{r} \, \mathbf{r} \rho_{2D}(\mathbf{r},\omega) \\ \rho_{2D}(\mathbf{r},\omega) &= \frac{4\pi\epsilon_{0}}{D} \sum_{\alpha} \frac{c_{\alpha}}{1/\eta_{\alpha} - 1/\eta(\omega)} v_{\alpha}(\mathbf{u}) \\ & & & & \\ P_{a,r}(\mathbf{R}_{e},\omega) &= \left| \hat{\mathbf{e}}_{a} + \sum_{\alpha} \frac{\zeta_{\alpha} \otimes \mathbf{F}_{\alpha}^{*}(\mathbf{R}_{e})}{1/\eta_{\alpha} - 1/\eta(\omega)} \cdot \hat{\mathbf{e}}_{a} \right|^{2} . \quad \zeta_{\alpha} = \int d^{2}\mathbf{u} \, \mathbf{u} v_{\alpha}(\mathbf{u}) \end{split}$$

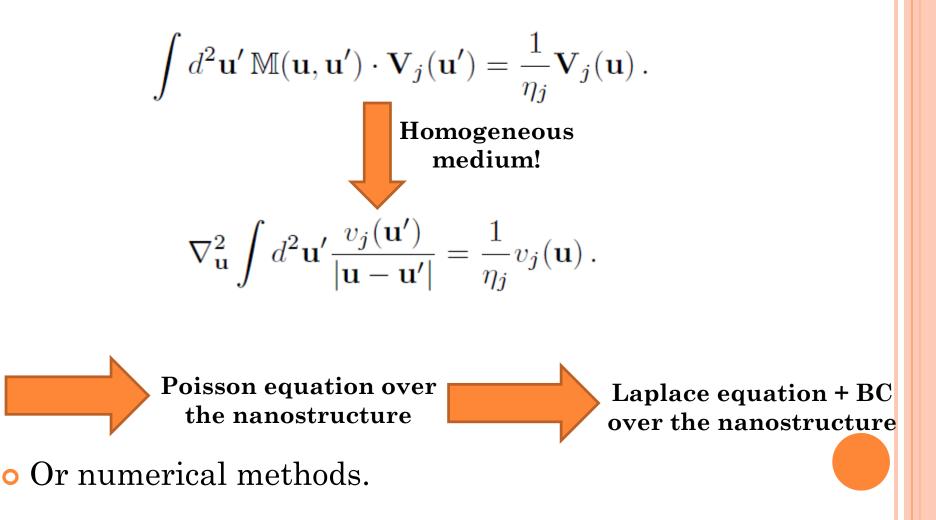
## APPROXIMATED RESULTS: DRUDE MODEL

$$P_{a,nr}(\mathbf{R}_e,\omega) \simeq \sum_{q=1}^N \frac{A_{a,q}}{\omega^2} \frac{1/2\tau}{(\omega-\omega_q)^2 + (1/2\tau)^2}$$
$$A_{a,q} = \frac{3c^3\omega_p^2 t}{16\pi D^4\omega_q^2} \sum_{j=1}^{g_q} |\hat{\mathbf{e}}_a \cdot \mathbf{F}_{q,j}(\mathbf{R}_e)|^2.$$

$$P_{a,r}(\omega) \simeq \sum_{q=1}^{N} \frac{B_{a,q}(1/2\tau)^2 + (\omega - \omega_q + f_{a,q}/2\tau)^2}{(\omega - \omega_q)^2 + (1/2\tau)^2} - (N-1)$$
$$f_{a,q} = \frac{\omega_p^2 \tau t}{4\pi D \omega_q} \sum_{j=1}^{g_q} \operatorname{Re} \left[ \hat{\mathbf{e}}_a \cdot \mathbf{F}_{q,j}^*(\mathbf{R}_e) \zeta_{a;q,j}^{\parallel} \right]$$

PLASMONS IN 2D NANOSTRUCTURES

• How do we obtain the PWFs?



## PLASMONS IN A NANODISK

#### • Analytical solution!

PWFs = 
$$R_{ln}(u)e^{il\phi}$$
.  $R_{ln}(u) = (2u)^{|l|} \sum_{m'} a_{m'}^{ln} P_{m'}^{(|l|,0)} (1 - 8u^2)$ 

$$\mathbb{G}^l \mathbf{a}^{ln} = -4\pi \eta_{ln} \mathbb{K}^l \mathbf{a}^{ln},$$

$$\mathbb{K}^{l}_{mm'} = \frac{(-1)^{m-m'+1}}{\pi[4(m-m')^2 - 1](|l| + m + m' + 1/2)(|l| + m + m' + 3/2)}, \quad m, m' = 0, 1, 2, 3...$$

$$\begin{split} \mathbb{G}^{l}_{mm'} &= \frac{\delta_{m0}\delta_{m'0}}{8|l|(|l|+1)^{2}} + \frac{\delta_{mm'}}{4(|l|+2m')(|l|+2m'+1)(|l|+2m'+2)} + \frac{\delta_{m+1,m'}}{8(|l|+2m+1)(|l|+2m+2)(|l|+2m+3)} \\ &+ \frac{\delta_{m,m'+1}}{8(|l|+2m'+1)(|l|+2m'+2)(|l|+2m'+3)}, \quad m,m'=0,1,2,3... \end{split}$$

•PRB 1986, **33**, 5221 •PRB 2016, **93**, 035426