Surface plasmon polaritons (SPP): an alternative to cavity QED

Strong coupling to excitons & Intermediary for quantum entanglement

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SPP

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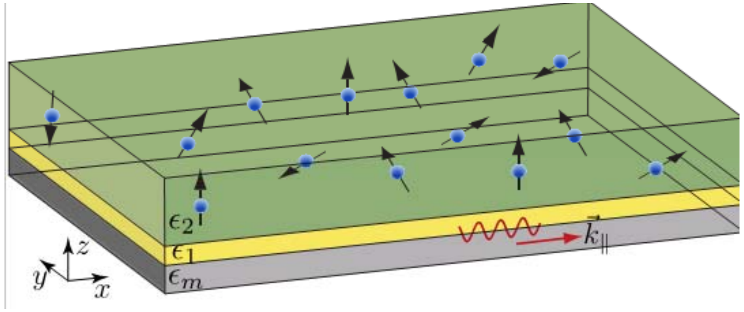

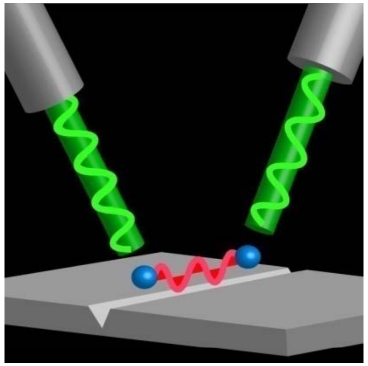

Outline

- Introduction to surface plasmon polaritons (SPP)
- Coupling of 1 quantum emitter (QE) to SPP
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SPP

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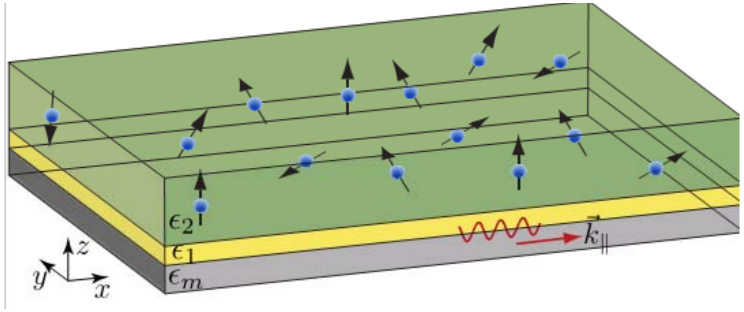

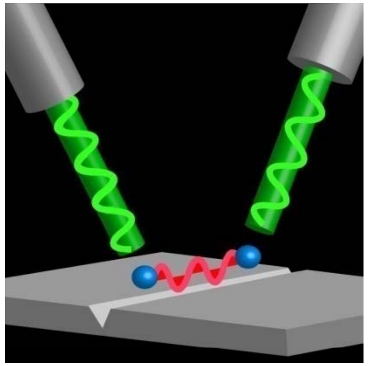

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Intro: Surface plasmon polaritons

- **Dielectric response of a metal is governed by free electron plasma:**

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

ω_p **: plasma frequency**

γ **: damping factor**

Below its plasma frequency ε(ω) is negative...

⇓

wavevector : $k = \frac{\omega\sqrt{\varepsilon}}{c}$ ⟶ **purely imaginary** ⟶ **photonic insulator**

What is a surface plasmon polariton ?

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What is a surface plasmon polariton ?

Bulk plasmon

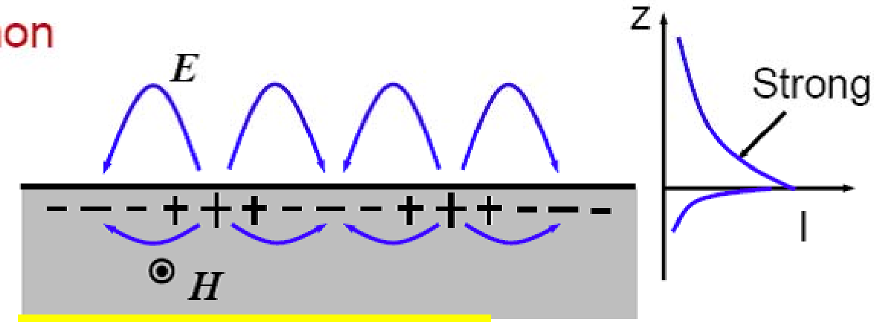
- Metals allow for EM wave propagation above the plasma frequency

They become transparent!

Surface plasmon

Dielectric

Metal



Note: SP is a TM wave!

- Sometimes called a surface plasmon-polariton (strong coupling to EM field)

Intro: Surface plasmon polaritons

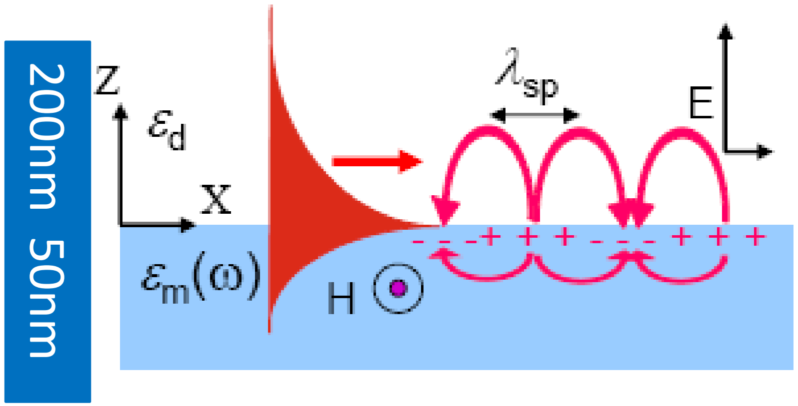
Electromagnetic radiation in dielectric ⊕ Localized Plasmons in a metal surface

⇓

SURFACE PLASMON POLARITONS

1. SPPs are primarily transverse in dielectrics but longitudinal in metals!

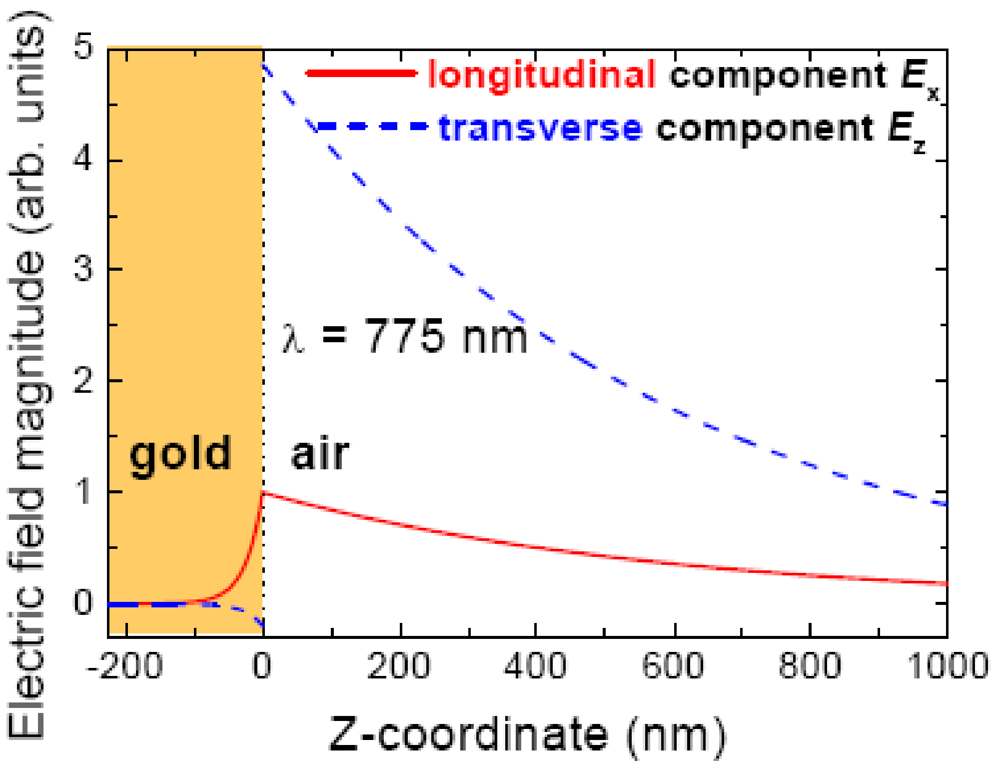
2. SPP properties are dictated by the boundary conditions for $E_{\|}$ and $E_{\perp}$!



$$E_z^d = i\sqrt{-\varepsilon_m / \varepsilon_d}\, E_x^0$$

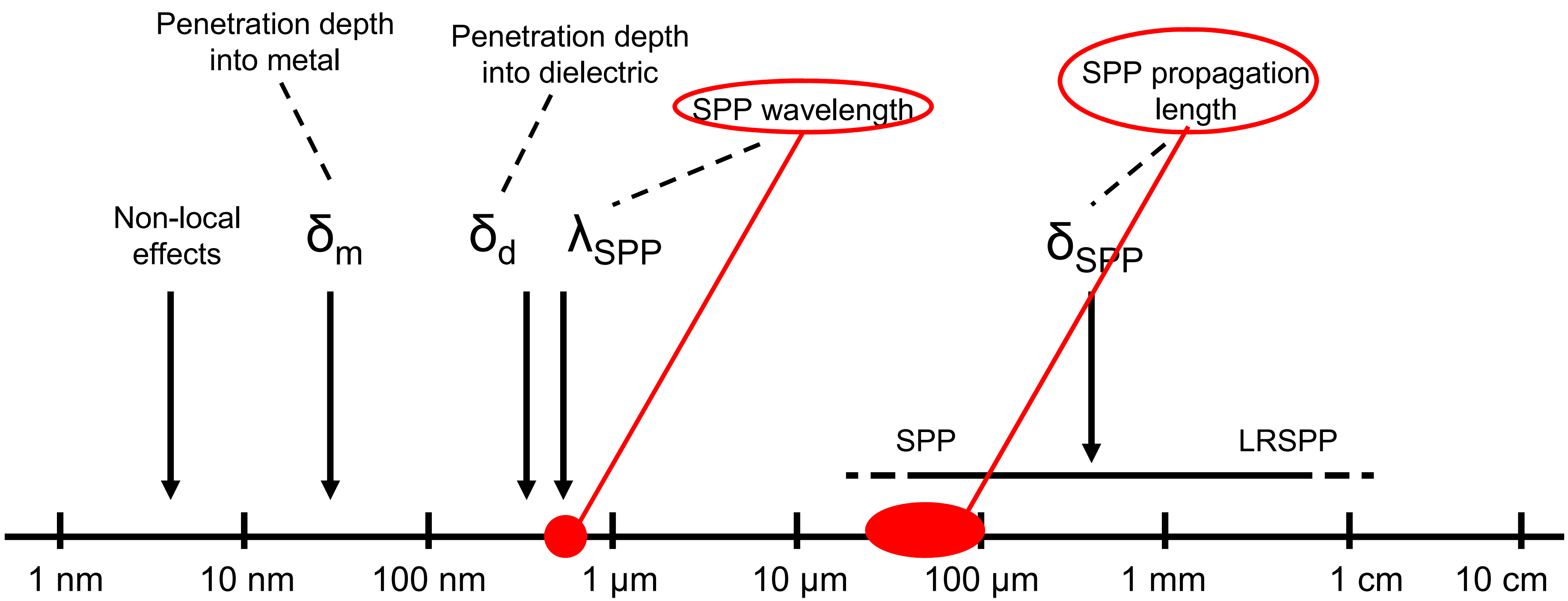
and

$$E_z^m = -i\sqrt{-\varepsilon_d / \varepsilon_m}\, E_x^0$$



Intro: Surface plasmon polaritons

SPP Length Scales span photonics and nano



Length scales span 7 orders of magnitude!

Intro: Surface plasmon polaritons

Plasmonics: Merging Photonics and Electronics at Nanoscale Dimensions

Ekmel Ozbay, Science, vol.311, pp.189-193 (13 Jan. 2006).

Roadmap for plasmonics

Some of the challenges that face plasmonics research in the coming years are

(i) demonstrate optical frequency subwavelength metallic wired circuits with a propagation loss that is comparable to conventional optical waveguides;

(ii) develop highly efficient plasmonic organic and inorganic LEDs with tunable radiation properties;

(iii) achieve active control of plasmonic signals by implementing electro-optic, all-optical, and piezoelectric modulation and gain mechanisms to plasmonic structures;

(iv) demonstrate 2D plasmonic optical components, including lenses and grating couplers, that can couple single mode fiber directly to plasmonic circuits;

(v) develop deep subwavelength plasmonic nanolithography over large surfaces;

(vi) develop highly sensitive plasmonic sensors that can couple to conventional waveguides;

(vii) demonstrate quantum information processing by mesoscopic plasmonics.

Intro: Surface plasmon polaritons

- **Interesting features of SPPs for photonic circuits**:

•Propagation length: 50-100 μm (Ag or Au) ⇔ lifetime ≤ 1 ps

•Two-dimensional character of EM-fields

•Optical and electrical signals carried without interference

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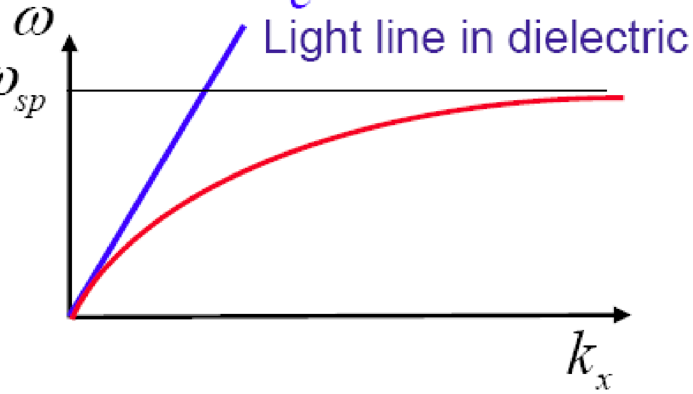
$k_{SPP}(\approx 10-100\,\mu m^{-1}) > \omega\sqrt{\varepsilon_d}\,/\,c \Rightarrow$ **Beyond the diffraction limit**

$$k_x = \frac{\omega}{c}\sqrt{\varepsilon_d}$$

$$\frac{\omega_p}{\sqrt{1+\varepsilon_d}} = \omega_{sp}$$



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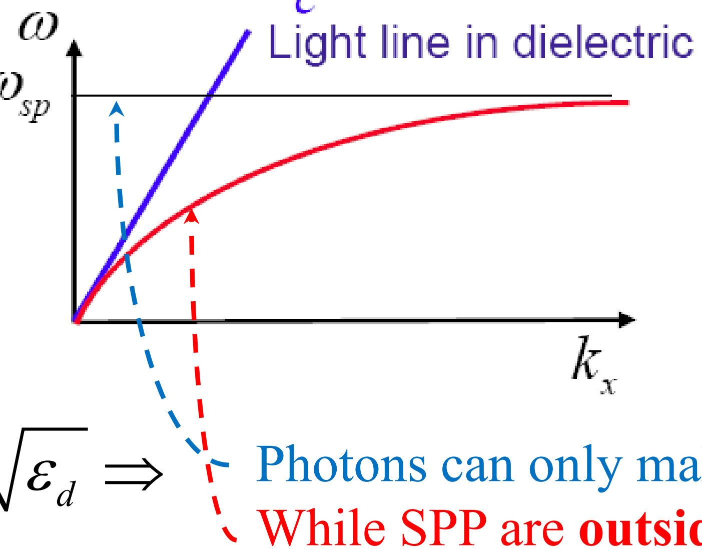
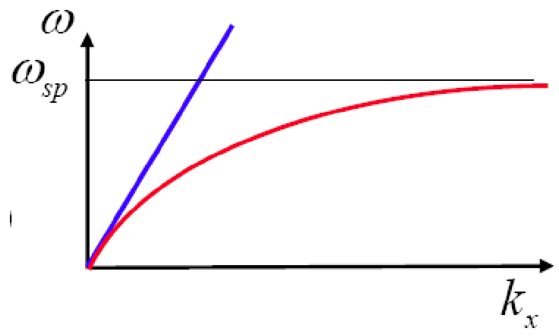


Main problem:
coupling in and out to SPPs

$$k_{\parallel} < k_{3D} = \frac{\omega}{c}\sqrt{\varepsilon_d} \Rightarrow$$

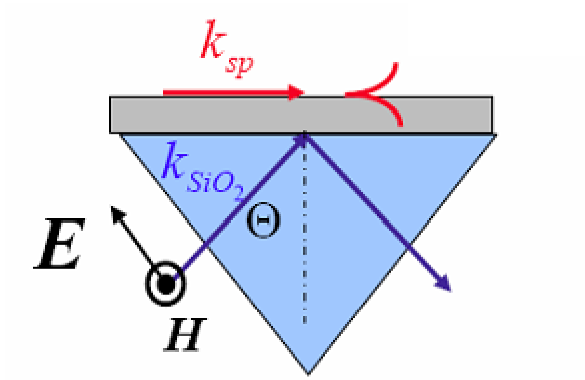
Photons can only make excitations **inside** the light cone
While SPP are **outside** the light cone

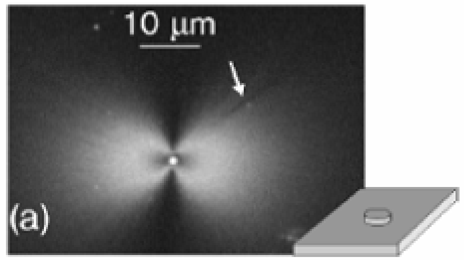
One problem: coupling light to SPPs

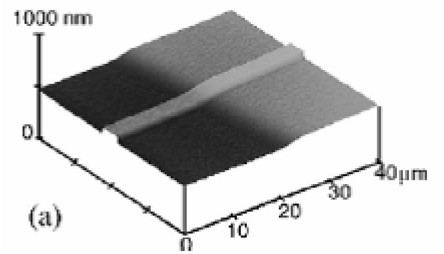


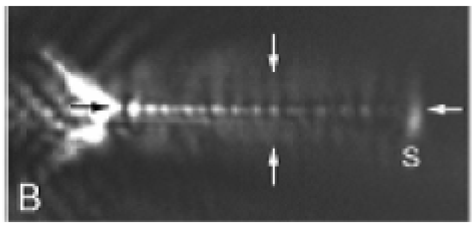
Coupling light to surface plasmon-polaritons

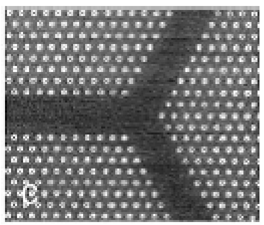
- Using high energy electrons (EELS)
- Kretchman geometry $$k_{//,SiO_2} = \sqrt{\varepsilon_d}\,\frac{\omega}{c}\sin\theta = k_{sp}$$
- Grating coupling
- Coupling using subwavelength features
- A diversity of guiding geometries











SPP

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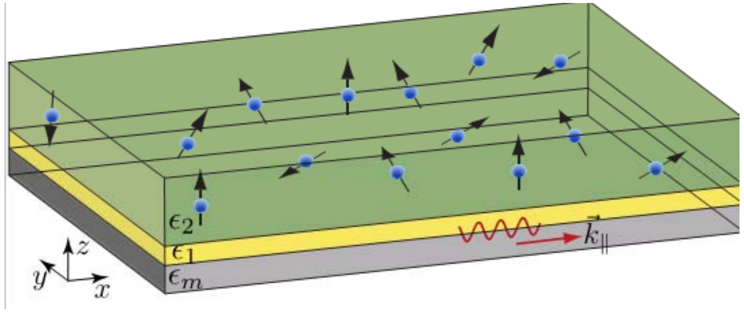

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Quantization of plasmons (without losses)

Quantization of an electric field

$$\vec{E}(\vec{r}) = \sum_{\vec{k}} \sqrt{\frac{\hbar\omega(k)}{2\epsilon_0 A}}\vec{u}_{\vec{k}}(z)e^{i(\vec{k}\cdot\vec{\rho}+k_z z)}a_{\vec{k}}$$

$$\vec{r} = (\vec{\rho}, z)$$

$$\vec{u}_{\vec{k}}(z) = \frac{1}{\sqrt{L(\vec{k})}}e^{-k_z z}\left(\hat{u}_{\vec{k}} - \frac{|\vec{k}|}{ik_z}\hat{u}_z\right)$$

with

$$L(\omega) = \frac{\pi}{2}\frac{\epsilon_m(\omega) - \epsilon_d}{\sqrt{\epsilon_d\epsilon_m(\omega)}\,|\vec{k}(\omega)|}\left[\epsilon_m(\omega) + \epsilon_d\left(1 + \omega\frac{d\epsilon_m(\omega)}{d\omega}\right)\right]$$

effective length to normalize the energy of each mode,

$$H_{EM} = \sum_{\vec{k}} \omega(k)a^{\dagger}_{\vec{k}}a_{\vec{k}}$$

Quantization of modes in a medium with dissipation

"Quantum Optics " Vogel & Welsch (Wiley 2006)

$\vec{P}_N\left(\vec{r},t\right)$ **Noise Polarization** associated with absorption

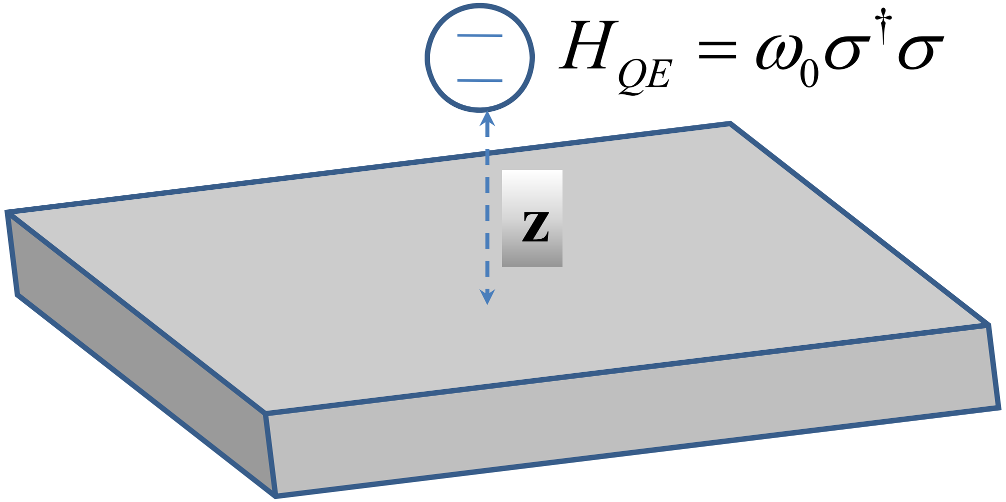
$$\vec{P}_N(\vec{r},\omega) = i\sqrt{\frac{\hbar\epsilon_0}{\pi}\operatorname{Im}\varepsilon\left(\vec{r},\omega\right)}\,\vec{f}(\vec{r},\omega)$$

$$\vec{E}(\vec{r},\omega) = i\sqrt{\frac{\hbar}{\pi\epsilon_0}}\frac{\omega^2}{c^2}\int d\vec{r}\,'\sqrt{\operatorname{Im}\varepsilon\left(\vec{r},\omega\right)}\,\widehat{G}\left(\vec{r},\vec{r}\,',\omega\right)\vec{f}(\vec{r},\omega)$$

$$\widehat{H} = \int d\vec{r}\int_0^\infty d\omega\,\hbar\omega\,\vec{f}^{\dagger}(\vec{r},\omega)\vec{f}(\vec{r},\omega)$$

Hereafter, everything is similar to the non-dissipation case with $\vec{f}(\vec{r},\omega)$ are **bosonic fields** playing the role of $a_{\vec{k}}$

Interaction of 1 quantum emitter (QE) with SPP



$$H_{QE} = \omega_0 \sigma^\dagger \sigma$$

Interaction with a dipole

$$U = \int d\vec{r}\, \vec{\mu}(\vec{r}) \cdot \vec{E}(\vec{r})$$

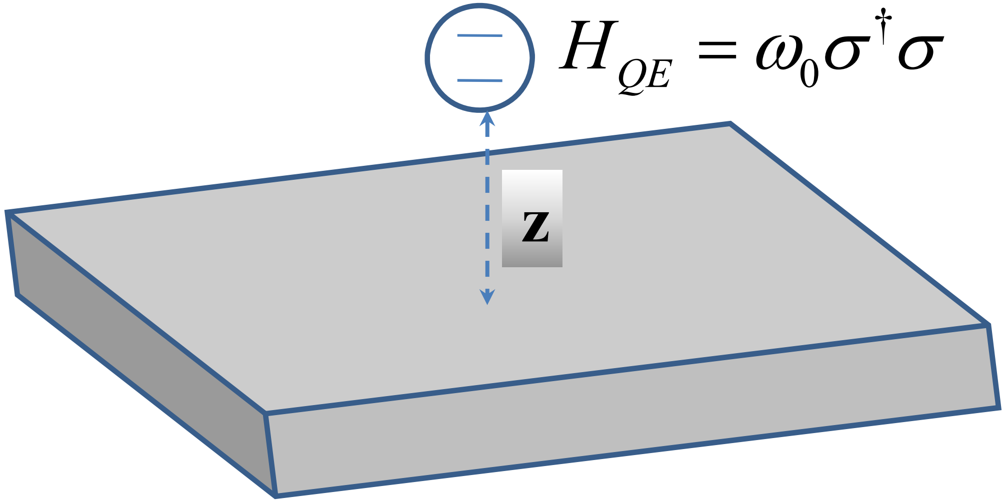
$$H_{int}(t) = \sum_{\vec{k}} \frac{g_{\vec{\mu}}(\vec{k};z)}{\sqrt{A}} \left(a_{\vec{k}} e^{i(\vec{k}\cdot\vec{r}-\omega(k)t)} + a_{\vec{k}}^\dagger e^{-i(\vec{k}\cdot\vec{r}-\omega(k)t)} \right) \left(\sigma^\dagger e^{i\omega_0 t} + \sigma e^{-i\omega_0 t} \right)$$

$$g_{\vec{\mu}}(\vec{k};z) = E_{\vec{k}} \vec{\mu} \cdot \vec{u}_{\vec{k}}(z) = \sqrt{\frac{\omega(\vec{k})}{2\epsilon_0 L(\vec{k})}} e^{-k_z z} \vec{\mu} \cdot \left(\hat{u}_{\vec{k}} - \frac{|\vec{k}|}{k_z} \hat{u}_z \right)$$

$$\mu^2 = 3\pi\varepsilon_0 c^3 \gamma_0 / \omega_0^3$$

decay rate of bare QE

Interaction of 1 quantum emitter (QE) with SPP



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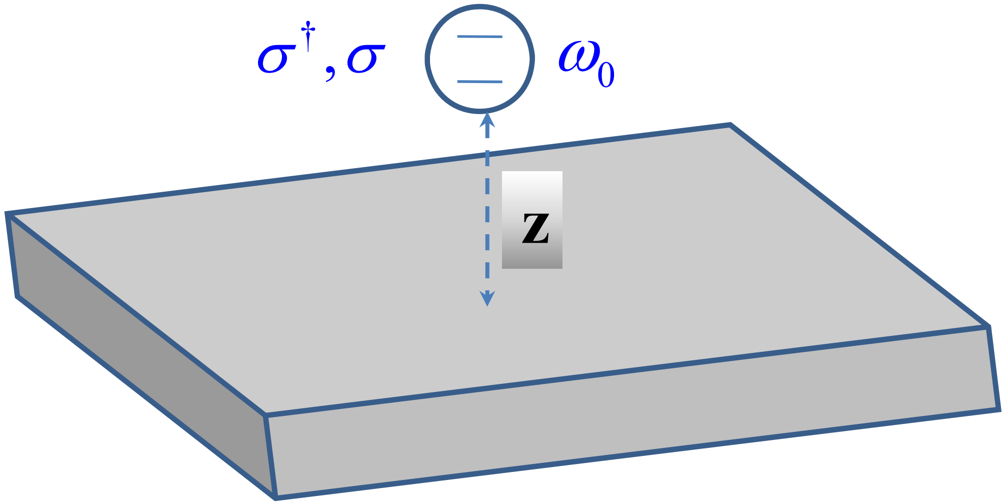
decay rate of bare QE

In RWA

$$H_{int} \approx \sum_{\vec{k}} \frac{g_{\vec{\mu}}(\vec{k};z)}{\sqrt{A}} \left(a_{\vec{k}} \sigma^\dagger e^{i\vec{k}\cdot\vec{r}} + a_{\vec{k}}^\dagger \sigma e^{-i\vec{k}\cdot\vec{r}} \right)$$

Interaction of 1 quantum emitter (QE) with SPP

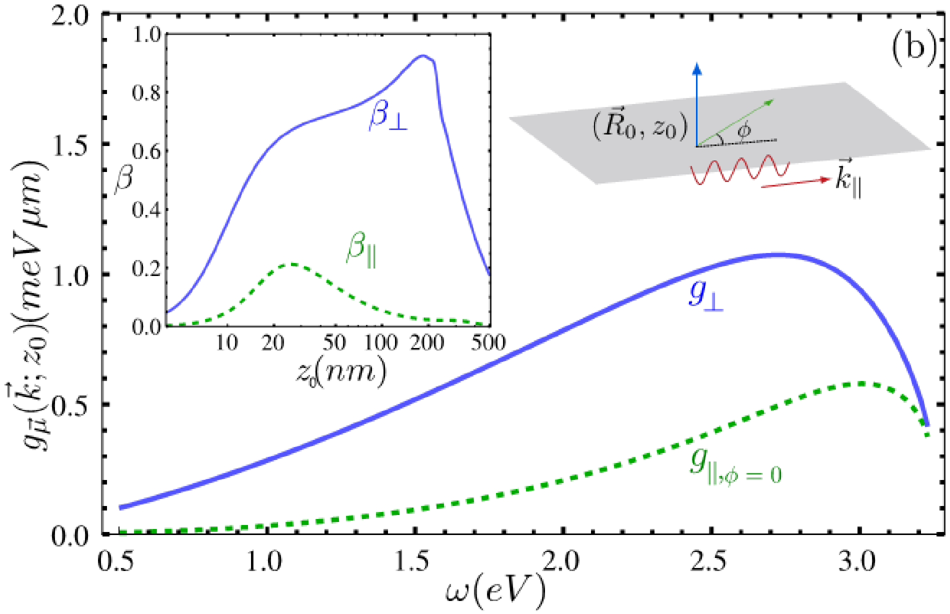


One QE with ω_0 only couples to a bright SPP ≡ symmetric linear comb. (J_0 Bessel funct.) of all the modes with

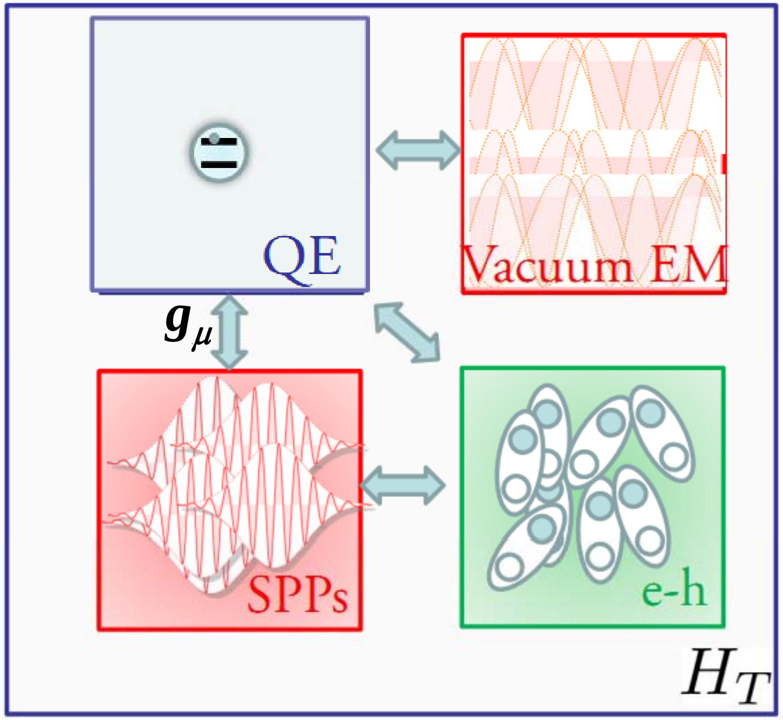
$$\left|\vec{k}\right|; \omega_0 = \omega_{SPP}\left(\left|\vec{k}\right|\right)$$

The higher coupling does not coincide with the higher β-factor

$$\beta = \frac{radiation\ to\ plasmons}{total\ radiation} = \frac{\gamma_{pl}}{\gamma}$$



Scheme of the quantum dynamics of an open system



Solving $i\frac{\partial|\Phi_{TOT}\rangle}{\partial t} = H_{TOT}|\Phi_{TOT}\rangle$

is complicated and unnecessary

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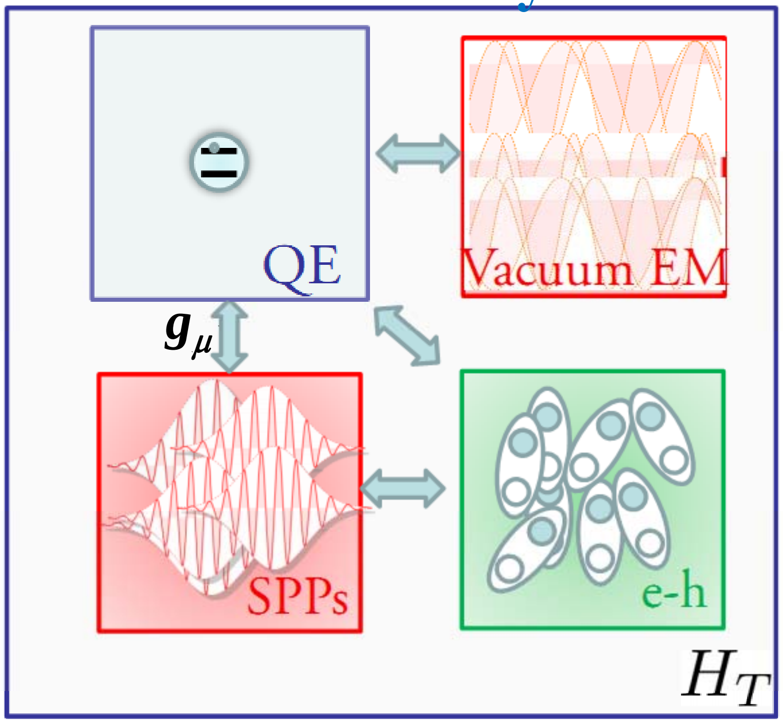
Isolated system



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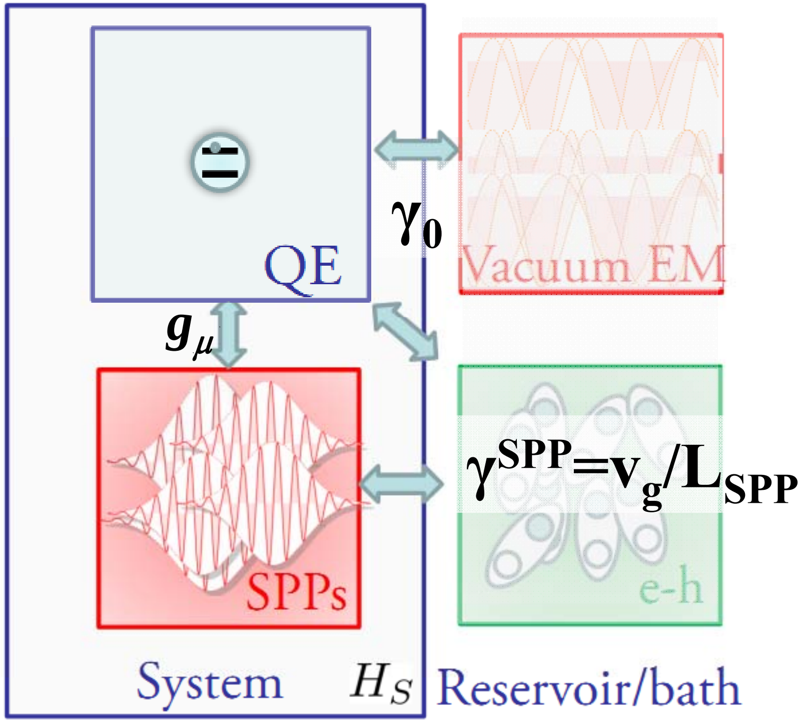

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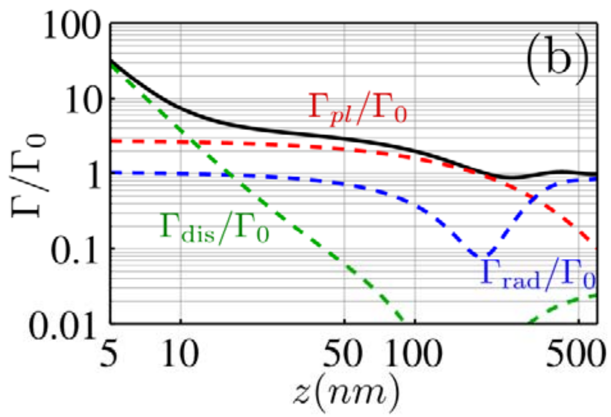
Tracing out the reservoir's degrees of freedom



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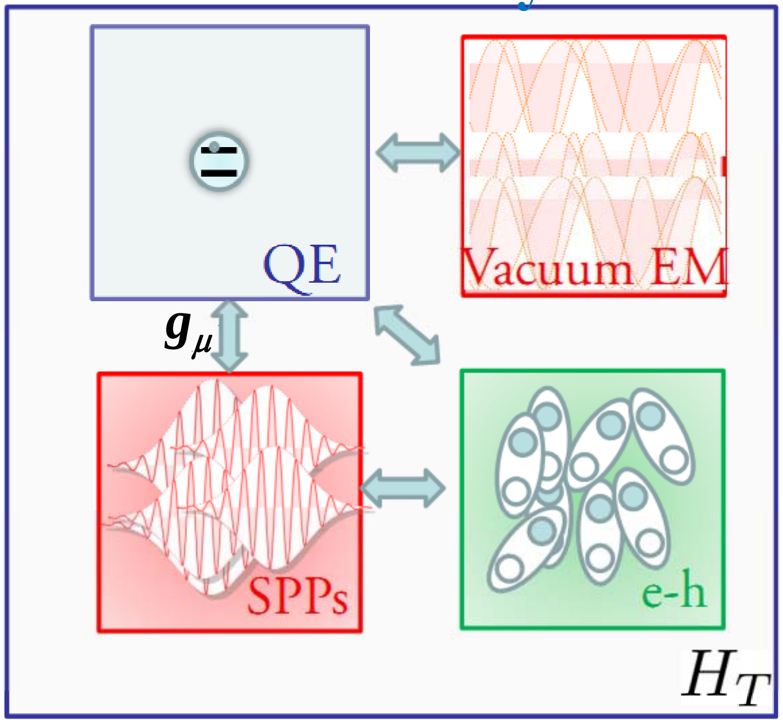

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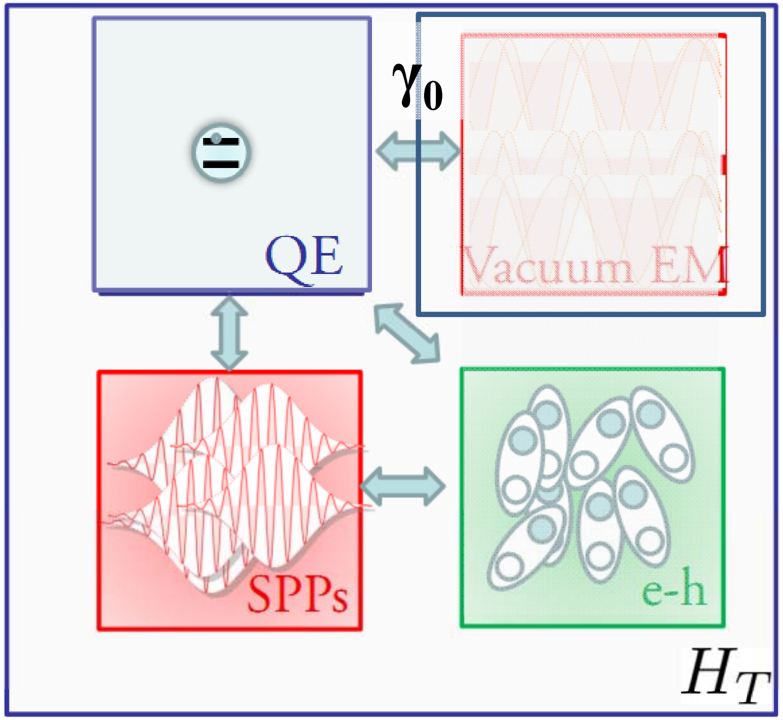
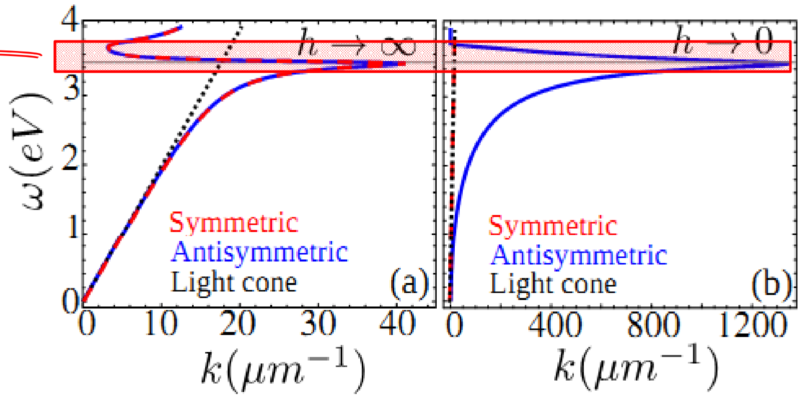


Solving $i\frac{\partial\left|\Phi_{TOT}\right\rangle}{\partial t} = H_{TOT}\left|\Phi_{TOT}\right\rangle$

is complicated and unnecessary

When dissipation at the metal is very high, tracing out **ONLY** the vacuum's degrees of freedom

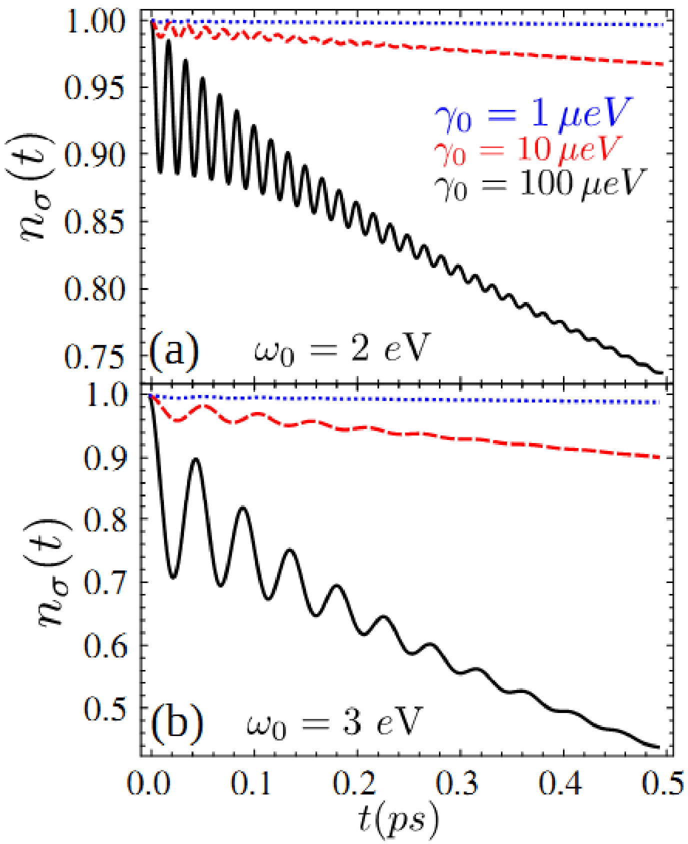


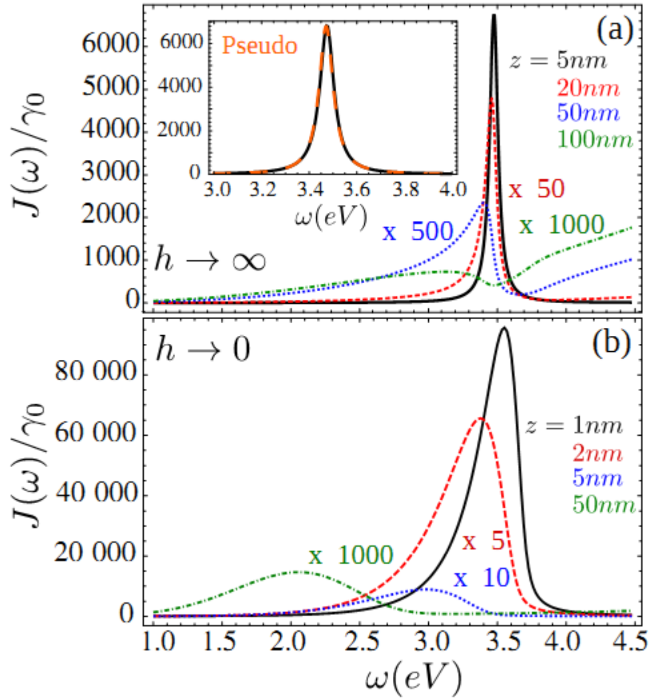
Dynamics of the QE population: Weisskopf-Wigner

$$\dot{c}_\sigma(t) = -\int_0^t K_{\vec{\mu}}(t-\tau;z)c_\sigma(\tau)d\tau - \gamma_0 c_\sigma(t)/2 \quad ; \quad c_\sigma(0)=1$$

$$K_{\vec{\mu}}(\tau;z) = \sum_{\vec{k}} |g_{\vec{\mu}}(\vec{k};z)|^2 \, e^{i[\omega_0-\omega(\vec{k})]\tau} = \int_0^{\omega_c} d\omega J(\omega;z)e^{i(\omega_0-\omega)\tau}$$

$$J(\omega;z) = \frac{1}{\pi\epsilon_0}\vec{\mu}\left[\frac{\omega^2}{c^2}\mathrm{Im}[\hat{G}(\vec{r}_0,\vec{r}_0,\omega)]\right]\vec{\mu} \;= g^2(\omega)\rho(\omega)$$

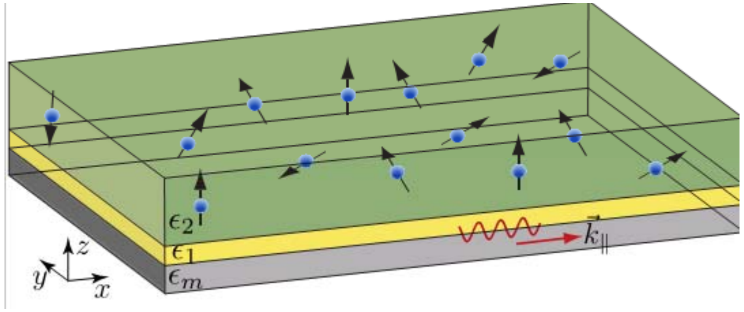




- **For a single QE with ω_0 around the cut-off, spectral density (J) has a non lorentzian shape ⇒ different dynamics that a pseudomode (cavity QED) !**
- **Non-markovian noise**
- **Height/width ratio of J determines/allows some (fast & local) reversibility !**

SPP

Strong coupling to excitons &



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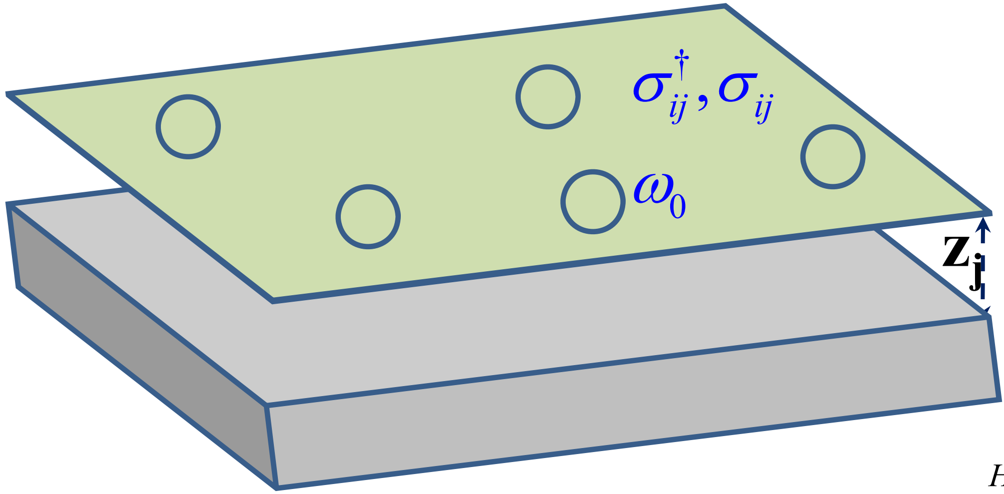
Exciton collective mode of emitters in a plane



More complicated system: Dynamics described by master eq. for density matrix & quantum regression th.

$$H_0^N + H_{pl} = \sum_{i=1}^{N_s} \omega_0 \ \sigma^{\dagger}_{i,j}\sigma_{i,j} + \sum_{\vec{k}} \ \omega(\mathrm{k}) a^{\dagger}_{\vec{k}} a_{\vec{k}}$$

$$H_{int}^N = \sum_{\vec{k}} \sum_{i=1}^{N_S} \frac{g_{\vec{\mu}}(\vec{k}; z_j)}{\sqrt{A}} (a^{\dagger}_{\vec{k}} \sigma_{i,j} e^{i\vec{k}\cdot\vec{r}_i} + a^{\dagger}_{\vec{k}} \sigma_{i,j} e^{-i\vec{k}\cdot\vec{r}_i})$$

$$g_{\vec{\mu}}(\vec{k}; z_j) = \sqrt{\frac{\omega(\vec{k})}{2\epsilon_0 L(\vec{k})}} e^{-k_z z_j} \vec{\mu} \cdot \left(\hat{u}_{\vec{k}} - \frac{|\vec{k}|}{k_z} \widehat{u_z} \right)$$

No fitting !!!

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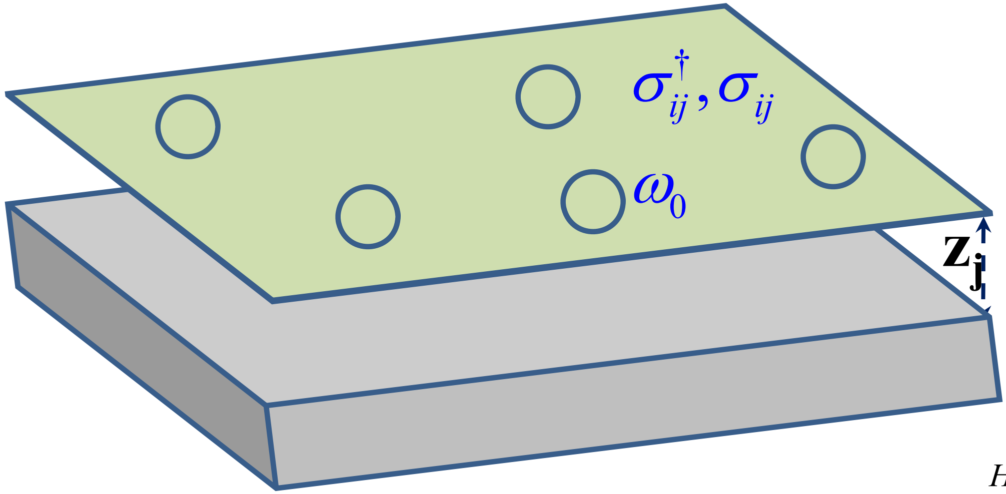

Holstein-Primakoff transf.
(low excitation $\Rightarrow$ no saturation)
& Collective bosonic mode

$\Downarrow$

$$\sigma^{\dagger}_{i,j} = \sqrt{1 - b^{\dagger}_{i,j} b_{i,j}} \cdot b_{i,j} \simeq b^{\dagger}_{i,j}$$

$$D^{\dagger}_{j}(\vec{q}) = \frac{1}{\sqrt{N_s}} \sum_{i=1}^{N_s} b^{\dagger}_{i,j} e^{i\vec{q}\cdot\vec{R}_i}$$

$$b^{\dagger}_{i,j} = \frac{1}{\sqrt{N_s}} \sum_{\vec{q}} D^{\dagger}_{j,\vec{q}} e^{-i\vec{q}\cdot\vec{R}_i}$$

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$$H_{int}^N = \sum_{\vec{k}} \sum_{i=1}^{N_S} \frac{g_{\vec{\mu}}(\vec{k}; z_j)}{\sqrt{A}} (a^{\dagger}_{\vec{k}} \sigma_{i,j} e^{i\vec{k}\cdot\vec{r}_i} + a^{\dagger}_{\vec{k}} \sigma_{i,j} e^{-i\vec{k}\cdot\vec{r}_i})$$

$$g_{\vec{\mu}}(\vec{k}; z_j) = \sqrt{\frac{\omega(\vec{k})}{2\epsilon_0 L(\vec{k})}} e^{-k_z z_j} \vec{\mu} \cdot \left(\hat{u}_{\vec{k}} - \frac{|\vec{k}|}{k_z} \hat{u}_z \right)$$

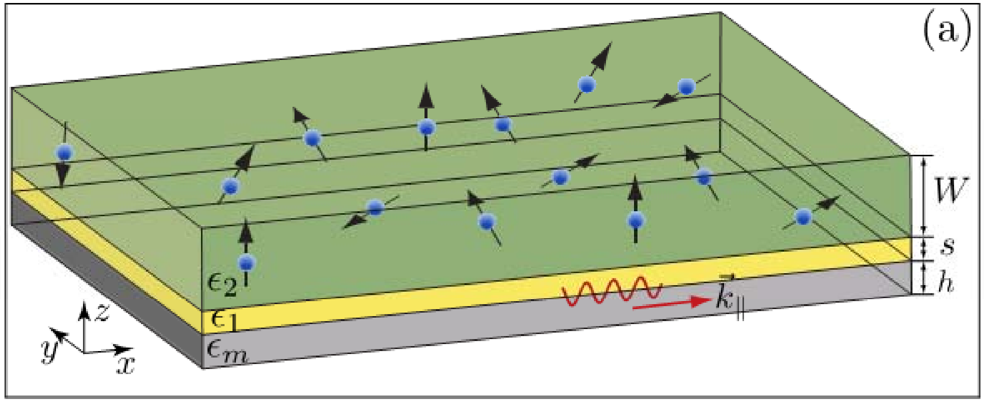
No fitting !!!

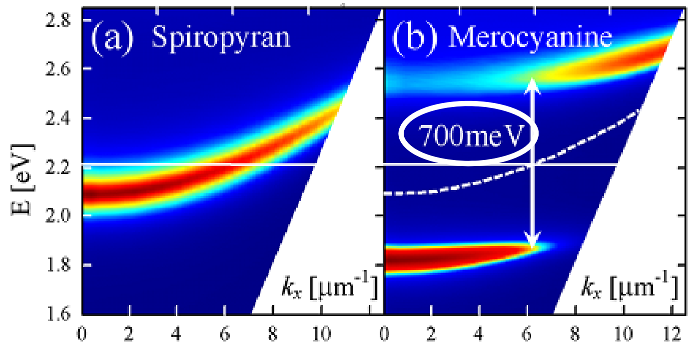
$$H_{int} = \sum_{\vec{k},\vec{q}} \sum_{i=1}^{N_s} \frac{g_{\vec{\mu}}(\vec{k}; z_j) \sqrt{n_s}}{N_s} \left(S(\vec{k} - \vec{q}) \cdot \vec{r}_i a_{\vec{k}} D^{\dagger}_j(\vec{q}) + S^{*}(\vec{k} - \vec{q}) \cdot \vec{r}_i a^{\dagger}_{\vec{k}} D_j(\vec{q}) \right)$$

$$S(\vec{k}) = \frac{1}{N_s} \sum_{i=1}^{N_s} e^{i\vec{k}\cdot\vec{r}_i}$$ Structure factor

Experimental evidence of strong coupling of SPP & excitons

QE are not just in a plane





Ensembles of organic molecules

- J. Bellessa, et al, Phys. Rev. Lett. 93, 036404 (2004).
- J. Dintinger, et al, Phys. Rev. B 71, 035424 (2005).
- T. K. Hakala, et al, Phys. Rev. Lett. 103, 053602 (2009).
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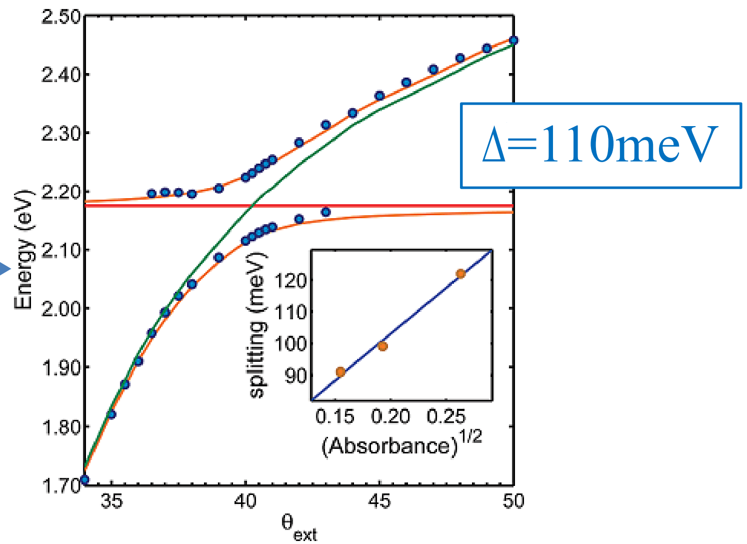

Semicond. nanocrystals & Quantum wells

- P. Vasa, et al., Phys. Rev. Lett. 101, 116801 (2008).
- J. Bellessa, et al, Phys. Rev. B 78 (2008).
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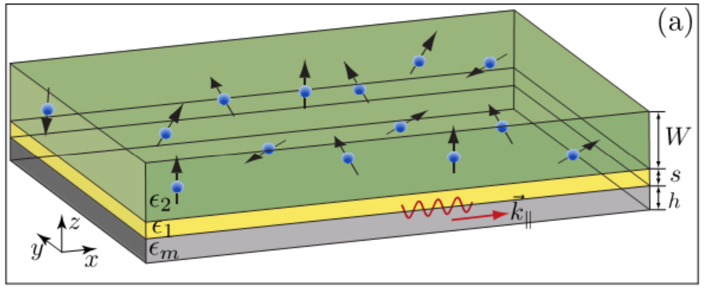
- M. Geiser, et al, Phys. Rev. Lett. 108, 106402 (2012).

Excitonic collective mode in the volume of width W



Coupling depends on distance z_j

Average of random orientations

$g_{\vec{\mu}}(\vec{k};z_j) \Rightarrow$ **More complicated collective mode**

For many QE with disorder $S(\vec{k}-\vec{q}) \approx \delta_{\vec{k},0}$ momentum is conserved

$$H_{\text{int}} = \sum_{\vec{k}} \sum_{j=1}^{N_L} g_{\vec{\mu}}(\vec{k};z_j)\sqrt{n_s}\left(a_{\vec{k}} D_j^{\dagger}(\vec{k}) + a^{\dagger}_{\vec{k}} D_j(\vec{k})\right)$$

$$D^{\dagger}(\vec{k}) = \frac{1}{g_{\vec{\mu}}^{N}(\vec{k})} \sum_{j=1}^{N_L} g_{\vec{\mu}}(\vec{k};z_j) D_j^{\dagger}(\vec{k}) \quad ; \quad [D_i(\vec{k}), D_j^{\dagger}(\vec{k})] = \delta_{ij}$$

$$g_{\vec{\mu}}^{N}(\vec{k}) = \sqrt{\sum_{j=1}^{N_L} |g_{\vec{\mu}}(\vec{k},z_j)|^2} \rightarrow \sqrt{n\int_s^{s+W} \mathrm{d}z |g_{\vec{\mu}}(\vec{k},z)|^2}$$

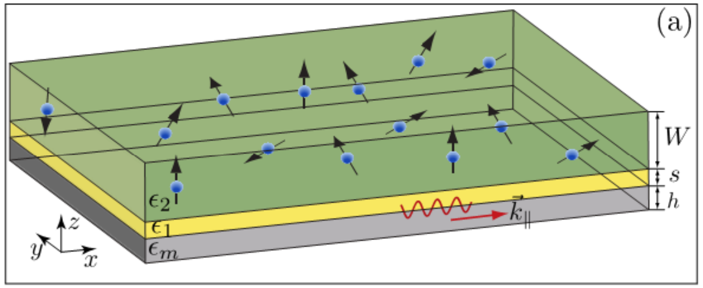
$$H_{\vec{k}}^{N} = \omega_0 D^{\dagger}(\vec{k})D(\vec{k}) + \omega_{\vec{k}} a^{\dagger}(\vec{k})a(\vec{k}) + g_{\vec{\mu}}^{N}(\vec{k})(a(\vec{k})D^{\dagger}(\vec{k}) + a^{\dagger}(\vec{k})D(\vec{k}))$$

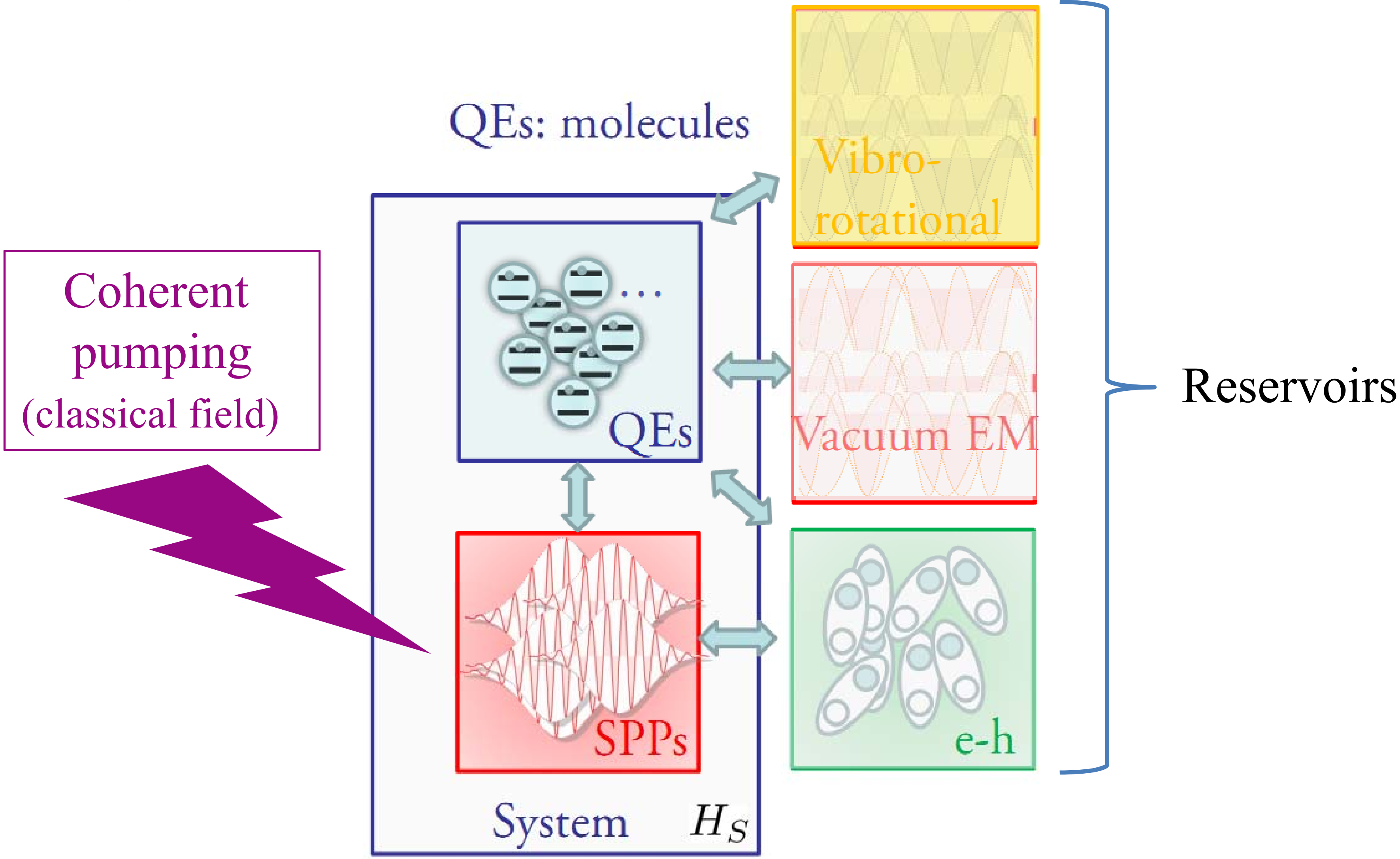
No fitting !

Decay of the collective mode $\gamma_{D_{\vec{k}}} = \dfrac{n}{|g_{\vec{\mu}}^{N}(\vec{k})|^2} \displaystyle\int_s^{s+W} \mathrm{d}z\, \gamma_{\sigma}(z) |g_{\vec{\mu}}^{N}(\vec{k},z)|^2$

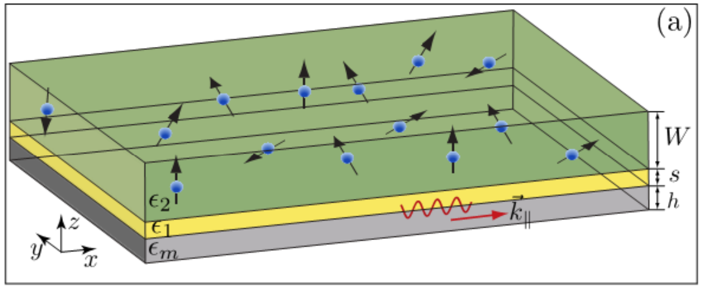
Dynamics under coherent pumping of a SPP with k-vector

Average of random orientations





Dynamics under coherent pumping of a SPP with k-vector



$$H_{\vec{k}}^{N} = \omega_0 D^\dagger(\vec{k})D(\vec{k}) + \omega_{\vec{k}} a^\dagger(\vec{k})a(\vec{k}) + g_{\vec{\mu}}^{N}(\vec{k})(a(\vec{k})D^\dagger(\vec{k}) + a^\dagger(\vec{k})D(\vec{k}))$$

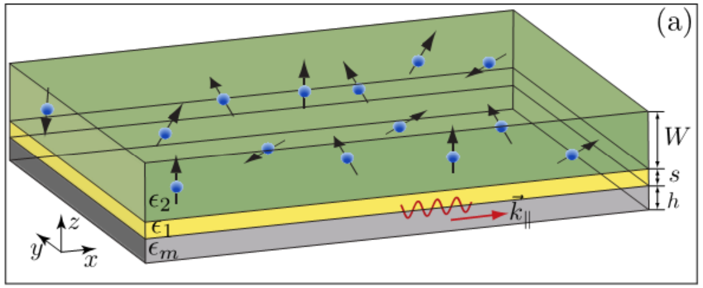
$$H_{\vec{k}}^{L}(t) = \Omega_{\vec{k}}(a_{\vec{k}} e^{i\omega_L t} + a_{\vec{k}}^\dagger e^{-i\omega_L t})$$

$$\dot{\rho}_{\vec{k}} = i[\rho_{\vec{k}}, H_{\vec{k}}^{N} + H_{\vec{k}}^{L}] + \frac{\gamma_{D_{\vec{k}}}}{2}\mathcal{L}_{D_{\vec{k}}} + \frac{\gamma_{a_{\vec{k}}}}{2}\mathcal{L}_{a_{\vec{k}}} + \frac{\gamma_\phi}{2}\mathcal{L}_{D_{\vec{k}}^\dagger D_{\vec{k}}}$$

Exciton decay | Plasmon decay | Pure dephasing (vibro-rotation)

$$\mathcal{L}_c = (2c\rho c^\dagger - c^\dagger c\rho - \rho c^\dagger c)$$

Dynamics under coherent pumping of a SPP with k-vector



$$H_{\vec{k}}^{N}=\omega_0 D^{\dagger}(\vec{k})D(\vec{k})+\omega_{\vec{k}}a^{\dagger}(\vec{k})a(\vec{k})+g_{\vec{\mu}}^{N}(\vec{k})(a(\vec{k})D^{\dagger}(\vec{k})+a^{\dagger}(\vec{k})D(\vec{k}))$$

$$H_{\vec{k}}^{L}(t)=\Omega_{\vec{k}}(a_{\vec{k}}e^{i\omega_L t}+a_{\vec{k}}^{\dagger}e^{-i\omega_L t})$$

$$\dot{\rho}_{\vec{k}}=i[\rho_{\vec{k}},H_{\vec{k}}^{N}+H_{\vec{k}}^{L}]+\frac{\gamma_{D_{\vec{k}}}}{2}\mathcal{L}_{D_{\vec{k}}}+\frac{\gamma_{a_{\vec{k}}}}{2}\mathcal{L}_{a_{\vec{k}}}+\frac{\gamma_{\phi}}{2}\mathcal{L}_{D_{\vec{k}}^{\dagger}D_{\vec{k}}}$$

Exciton decay — Plasmon decay — Pure dephasing (vibro-rotation)

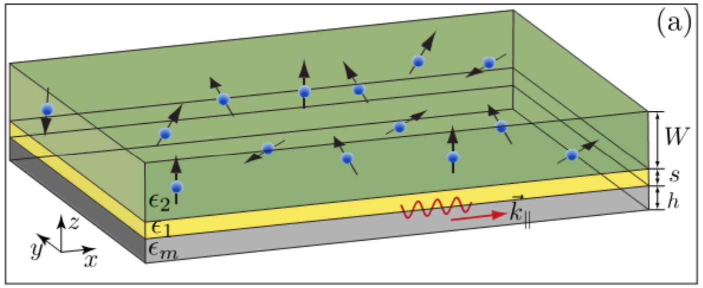
$$\mathcal{L}_c=(2c\rho c^{\dagger}-c^{\dagger}c\rho-\rho c^{\dagger}c)$$

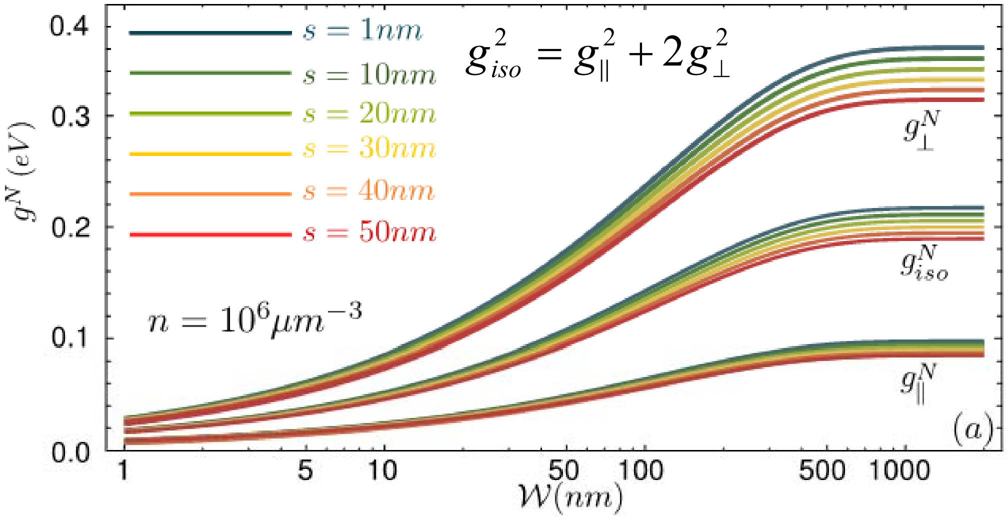
At the crossing (k_0) between exciton and SPP,
Rabi splitting is analytical

$$R=\sqrt{[g_{\vec{\mu}}^{N}(\vec{k}_0)]^2-(\gamma_{D_{\vec{k}_0}}+\gamma_{\phi}-\gamma_{a_{\vec{k}_0}})^2/4}\quad with\ [g_{\vec{\mu}}^{N}(\vec{k}_0)]^2\propto n$$

Strong coupling between SPP & excitons

$$\gamma_0 = 0.1 meV$$



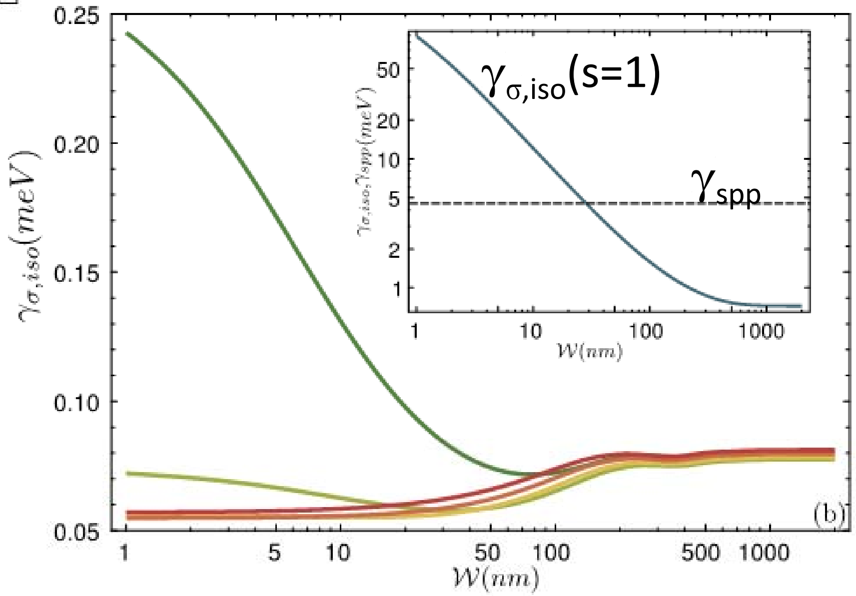


- g^N **depends on W** with saturation due to SPP z-decay
- g^N **practically independ. on s**

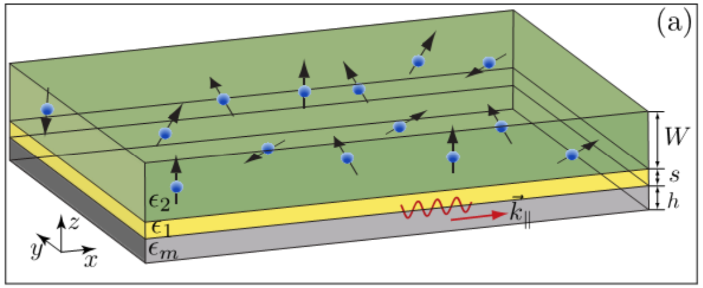
$$\Omega_{\vec{k}} = 0.1 g^N$$

$$n = 10^6 \ \mu m^{-3}$$

$$\omega_0 = 2eV$$



Strong coupling between SPP & excitons

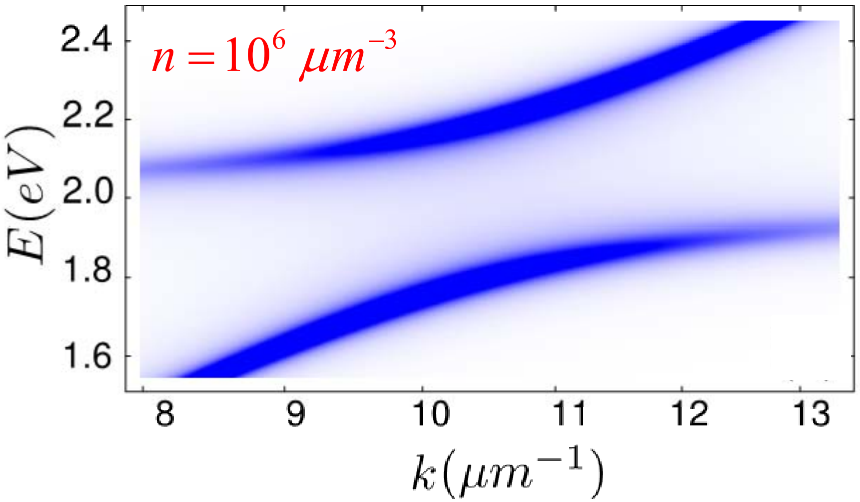


$$s = 1\,nm\,;\, W = 500\,nm$$

$$\omega_0 = 2eV\,;\;\; \Omega_{\vec{k}} = 0.1g^N\; (40meV)$$

$$\gamma_0 = 0.1meV\,;\, \gamma_\phi = 0.1g^N\, (RT)$$

Polariton populations $\propto$ absorption spect.



Strong coupling between SPP & excitons

$s = 1\,nm\,;\, W = 500\,nm$

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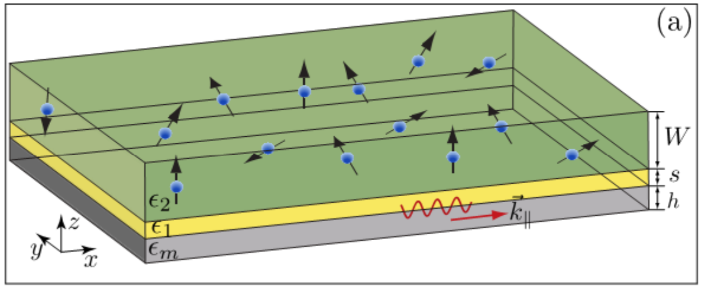
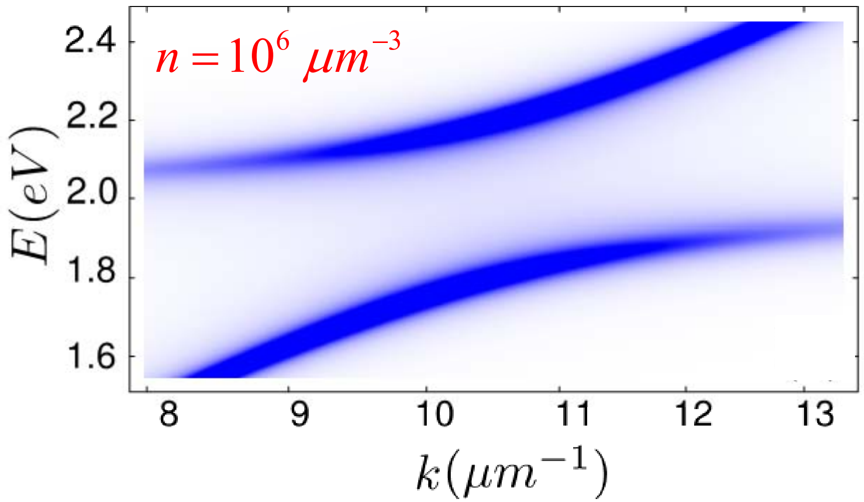
$\gamma_0 = 0.1meV\,;\, \gamma_\phi = 0.1g^N\,(RT)$

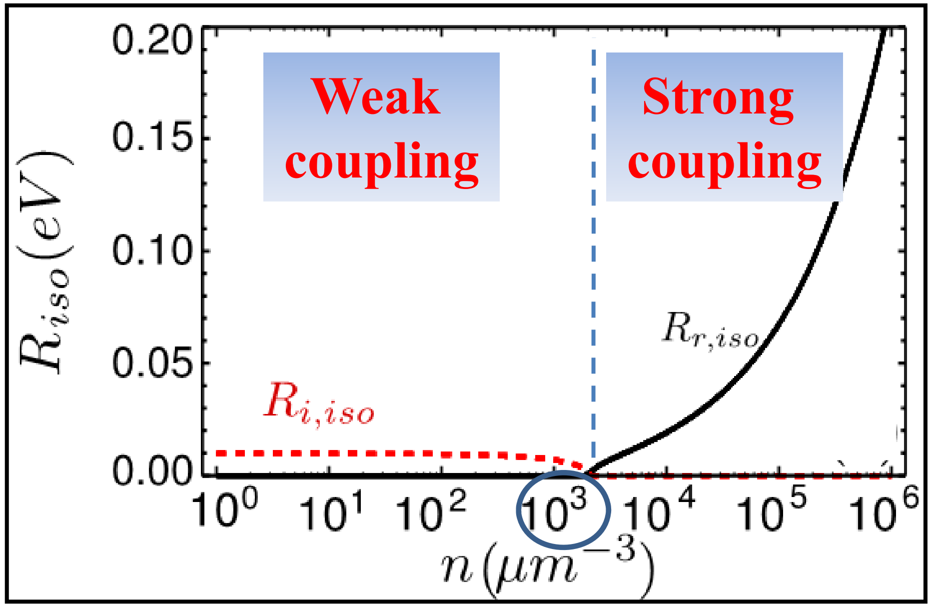


Rabi splitting (at k_0)

$R = \sqrt{[g^N_{\vec{\mu}}(\vec{k}_0)]^2 - (\gamma_{D_{\vec{k}_0}} + \gamma_\phi - \gamma_{a_{\vec{k}_0}})^2/4}\,;[g^N_{\vec{\mu}}(\vec{k}_0)]^2 \propto n$

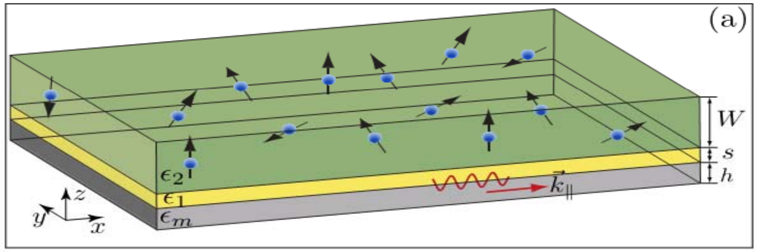
Polariton populations $\propto$ absorption spect.





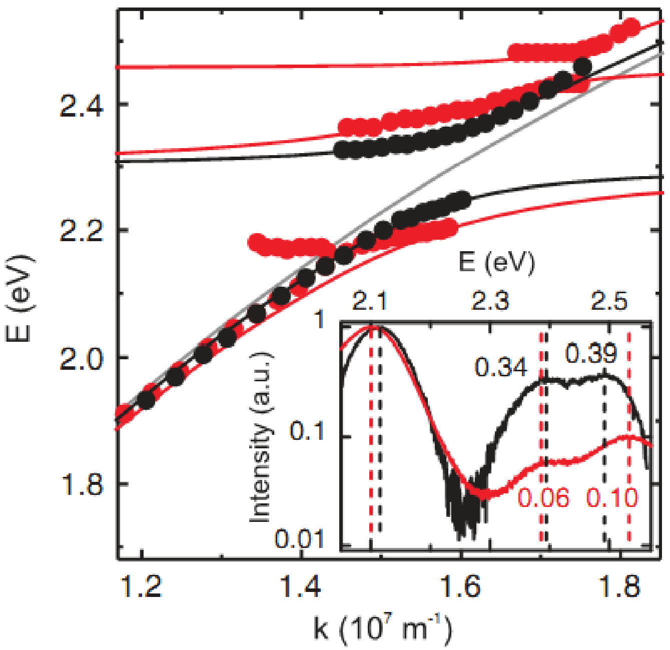
At RT, the incoherent processes (γ_ϕ) determine a critical density for observing strong coupling

Strong coupling between SPP & excitons: comparison with experiments



Experiment

Hakala et al. PRL, 103, 053602 (09)



Our theory

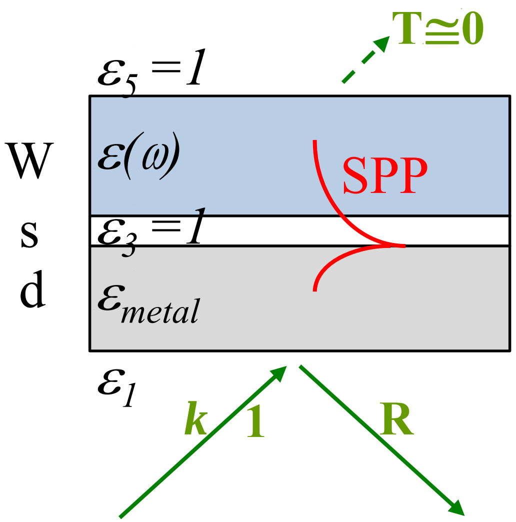
$$W = 50nm\,; n = 1.2\times10^{8}\,\mu m^{-3}\,; \gamma_0 = 1\mu eV$$

$$R_{th}^{\parallel} = 40meV, \quad R_{th}^{\perp} = 180meV, \quad R_{th}^{iso} = 100meV$$

$$R_{\exp} = 115meV$$

Quantum effects? : (Semi)-classical description

Polarizability of 1 emitter $\alpha(\omega) = \dfrac{f_0 e^2 / m}{\omega_0^2 - \omega^2 i\omega\gamma}$; $\varepsilon(\omega) = \dfrac{1 + (2/3) N\alpha(\omega)}{1 - (1/3) N\alpha(\omega)}$ Eff. dielectric funct. of emitters



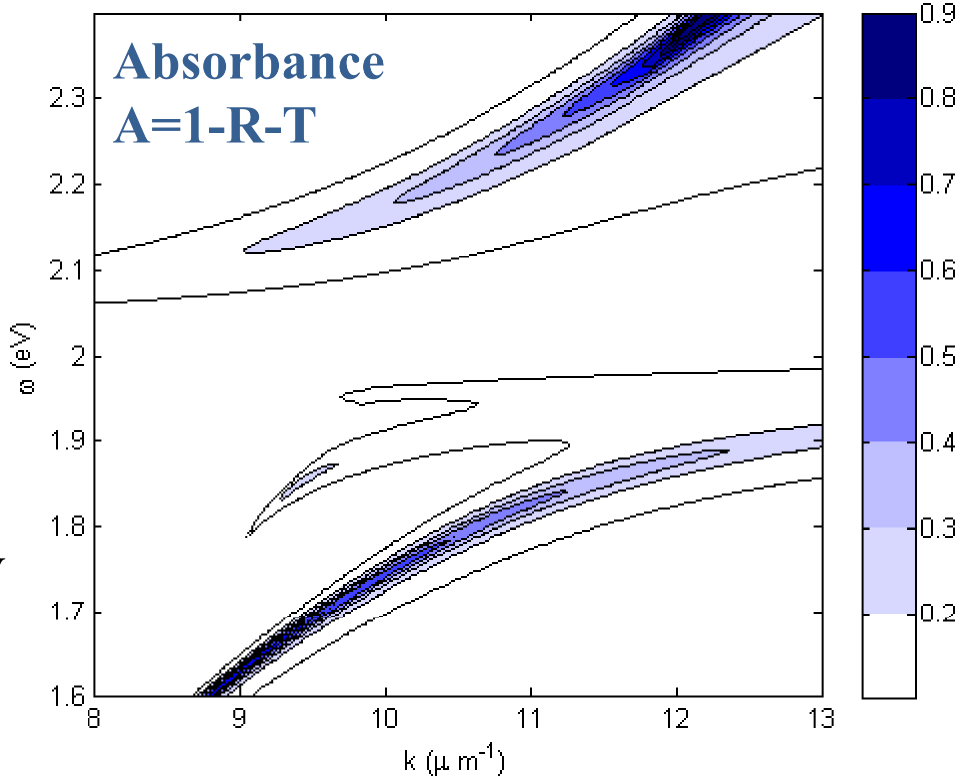
Oscillator strength from microsocpic info.

$$f_0 = \frac{2m\omega_0}{3e^2\hbar}\left|\vec{\mu}\right|^2 = \frac{2\pi m}{e^2}\frac{\varepsilon_0 c^3}{\omega_0^2}\gamma_0$$

ε_1=3; d=50nm; s=1nm; W=500nm

ω_0=2eV; N=10^6 μm^{-3}

$\gamma = \gamma_0 + \gamma_{deph}$; γ_0=0.1meV; γ_{deph}=40meV

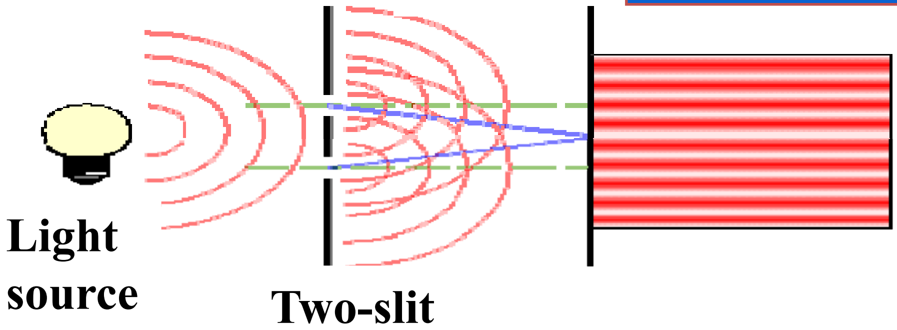


Quantum effects: 1st & 2nd order coherences

1 detector

Young's interfer. exp.

$$g^{(1)}(t,0) \propto \langle E^{(-)}(t) E^{(+)}(t) \rangle \propto \langle I(t) \rangle$$



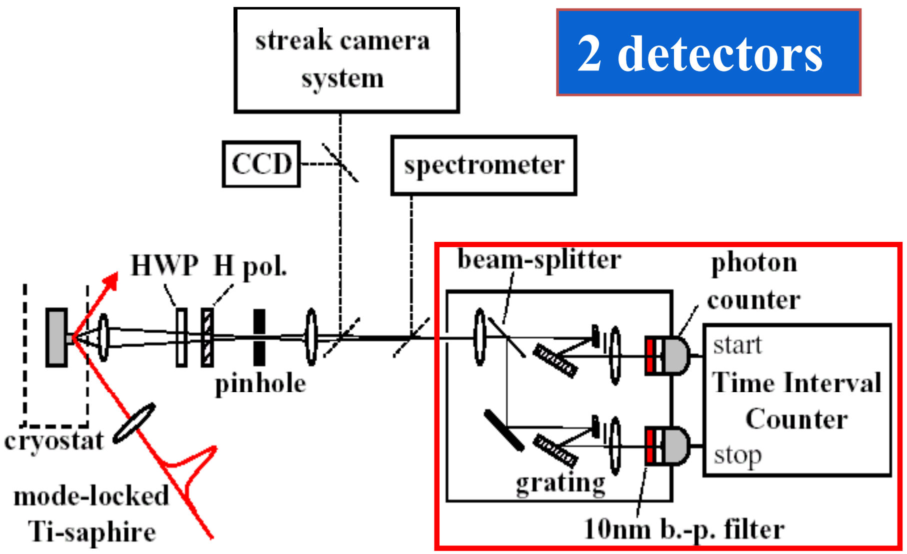
Amplitude interference pattern: the Fouriertransform is the spectrum

$g^{(1)}$ does not distinguish between classical & quantum light

Hanbury-Brown Twiss exp.

2 detectors

$$g^{(2)}(t,\tau) \propto \langle E^{(-)}(t) E^{(-)}(t+\tau) E^{(+)}(t+\tau) E^{(+)}(t) \rangle$$
$$\propto \langle I(t) I(t+\tau) \rangle$$



Intensity-intensity correlations

Quantum effects: 1st & 2nd order coherences

$$g^{(1)}(t,\tau) \propto \langle E^{(-)}(t) E^{(+)}(t+\tau) \rangle \qquad S(r,\omega) = \frac{1}{\pi} \Re \int_0^\infty d\tau \left\langle E^{(-)}(r,t) E^{(+)}(r,t+\tau) \right\rangle e^{i\omega\tau}$$

Spectrum ($g^{(1)}$) $\Leftrightarrow$ Amplitude interf. $\oplus$ 1 measurement

It does not distinguish between classical & quantum waves

$$g^{(2)}(t,\tau) \propto \langle E^{(-)}(t) E^{(-)}(t+\tau) E^{(+)}(t+\tau) E^{(+)}(t) \rangle \propto \langle I(t) I(t+\tau) \rangle$$

Intensity-int. correlations ($g^{(2)}$) $\Leftrightarrow$ 2 measurements

Classical: First detection does not affect second detection

Quantum: First detection affects second detection

$g^{(2)}$ can distinguish between

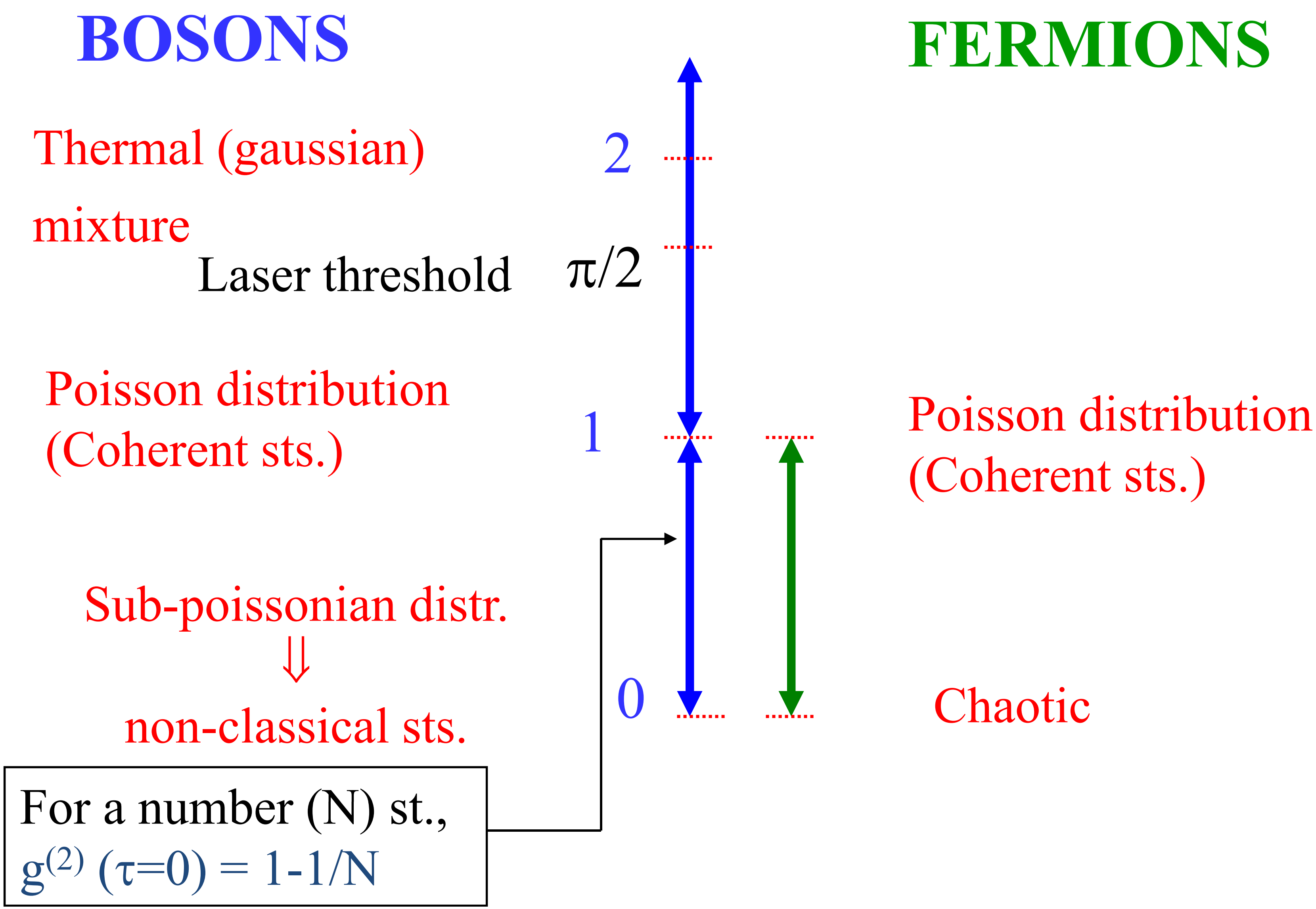
classical fields ($1 < g^{(2)} < \infty$)

0 — 1 — ∞

quantum fields ($0 < g^{(2)} < \infty$)

(Better to measure Bell's inequalities)

Quantum effects: $g^{(2)}$ (0) in different regimes



Quantum effects in the coupling between SPP & excitons

Quantum effects appear when non-linear effects are important: Holstein-Primakoff up to 2nd order

$$\sigma_{i,j} = \sqrt{1 - b_{i,j}^\dagger b_{i,j}} \cdot b_{i,j} \approx (1 - b_{i,j}^\dagger b_{i,j} / 2) b_{i,j}$$

Free energy part of the Hamilt.

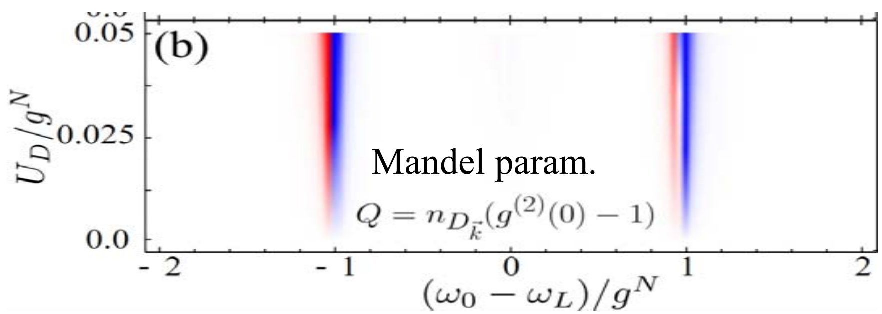
$$H_0^N = \sum_{j=1}^{N_L} \sum_{i=1}^{N_s} \omega_0 \ b_{i,j}^\dagger b_{i,j} - \sum_{j=1}^{N_L} \sum_{i=1}^{N_s} \omega_0 \ b_{i,j}^\dagger b_{i,j}^\dagger b_{i,j} b_{i,j}$$

In the quasi-2D limit & using The collective operators:

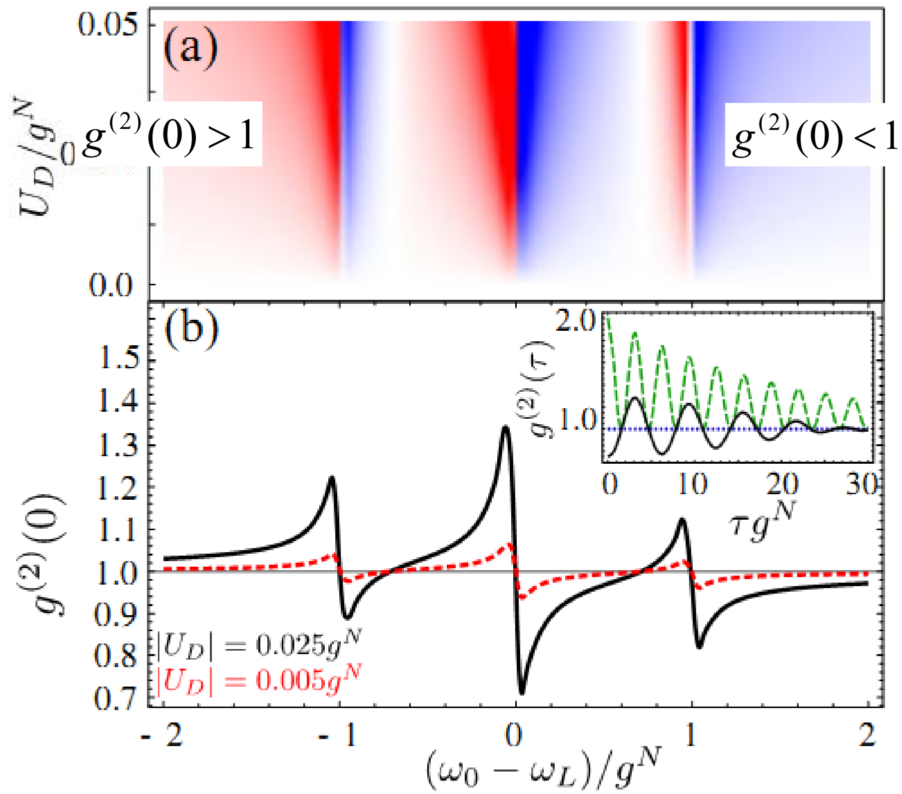
$$H_{nl} = U_D \sum_{\vec{k},\vec{k}',\vec{q}} D_{\vec{k}+\vec{q}}^\dagger D_{\vec{k}'-\vec{q}}^\dagger D_{\vec{k}} D_{\vec{k}'}$$

$$U_D = -\frac{\omega_0}{N}$$

$$g^{(2)}(\tau) = \lim_{t\to\infty} \frac{\left\langle D_{\vec{k}}^\dagger(t)(D_{\vec{k}}^\dagger D_{\vec{k}})(t+\tau) D_{\vec{k}}(t)\right\rangle}{\left\langle D_{\vec{k}}^\dagger D_{\vec{k}}(t)^2\right\rangle}$$

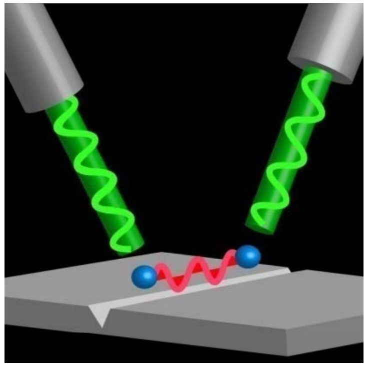


Coherent pumping of plasmons & $\gamma_\phi = 0$



SPP

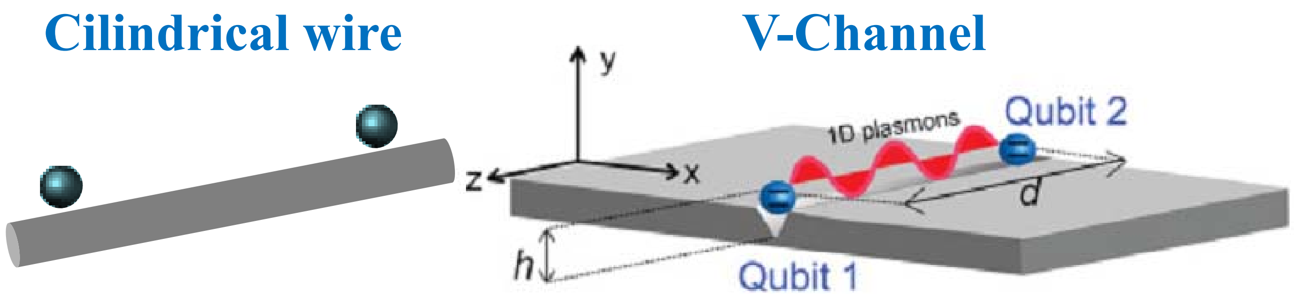
Intermediary for quantum entanglement



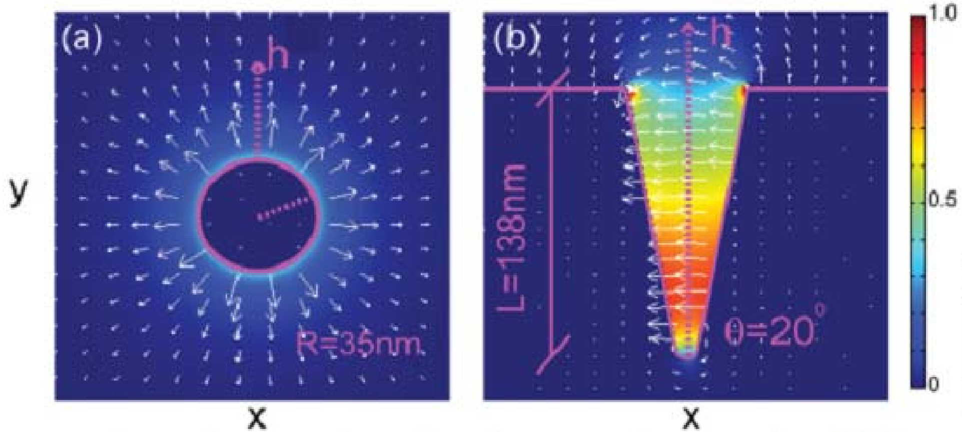
Outline

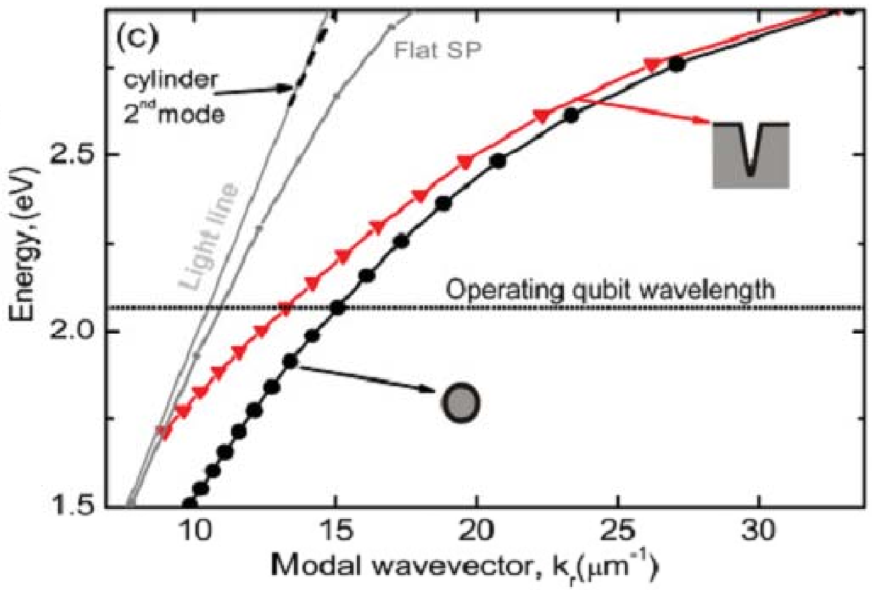
- Introduction to surface plasmon polaritons (SPP)
- Coupling of 1 quantum emitter (QE) to SPP
- Collective mode (excitons) strongly coupled to SPP
- **Coupling & entanglement of 2 QE mediated by SPP**
- Conclusion

QE-QE coupling mediated by plasmonic waveguides



Waveguide to reinforce QE-QE effects



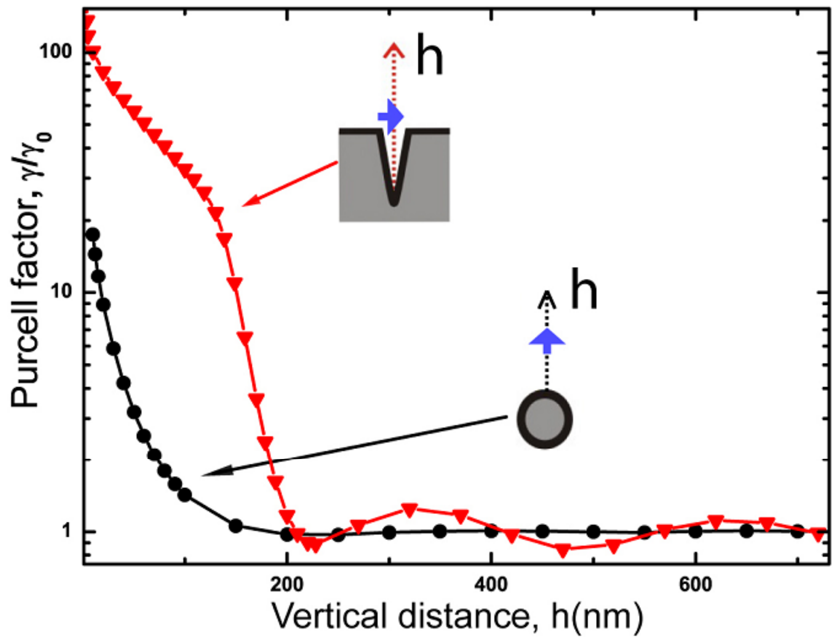


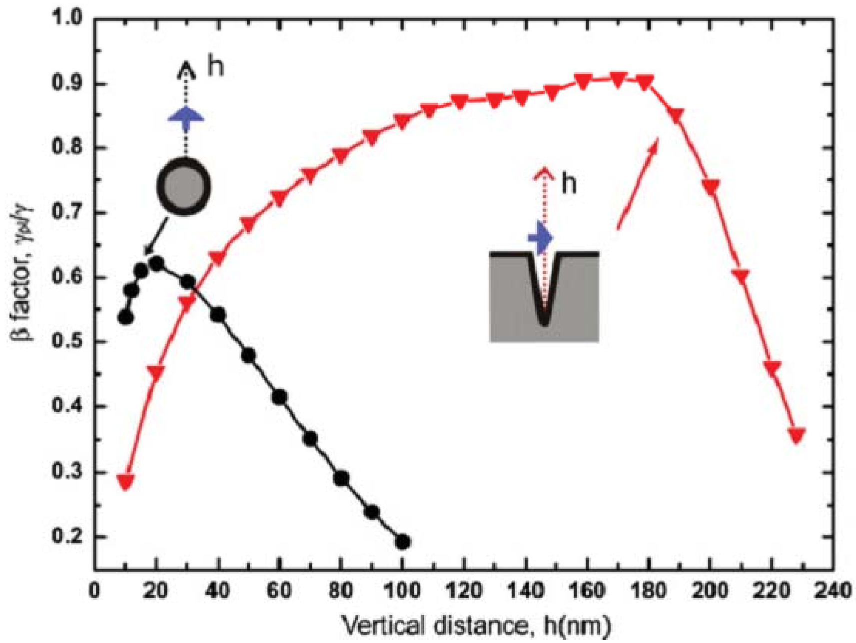
Fields are stronger in the channel than in the cilinder

1QE: β and Purcell factors

Metallic nanostructures increase the emission from a QE (Purcell)
but,
Is it always a coherent emission of SPP's**???** **(β)**

$$Purcell\ \ factor = \frac{total\ radiation}{QE\ radiation\ to\ vacuum} = \frac{\gamma}{\gamma_0}\ ;\ \beta\ factor = \frac{radiation\ to\ plasmons}{total\ radiation} = \frac{\gamma_{pl}}{\gamma}$$

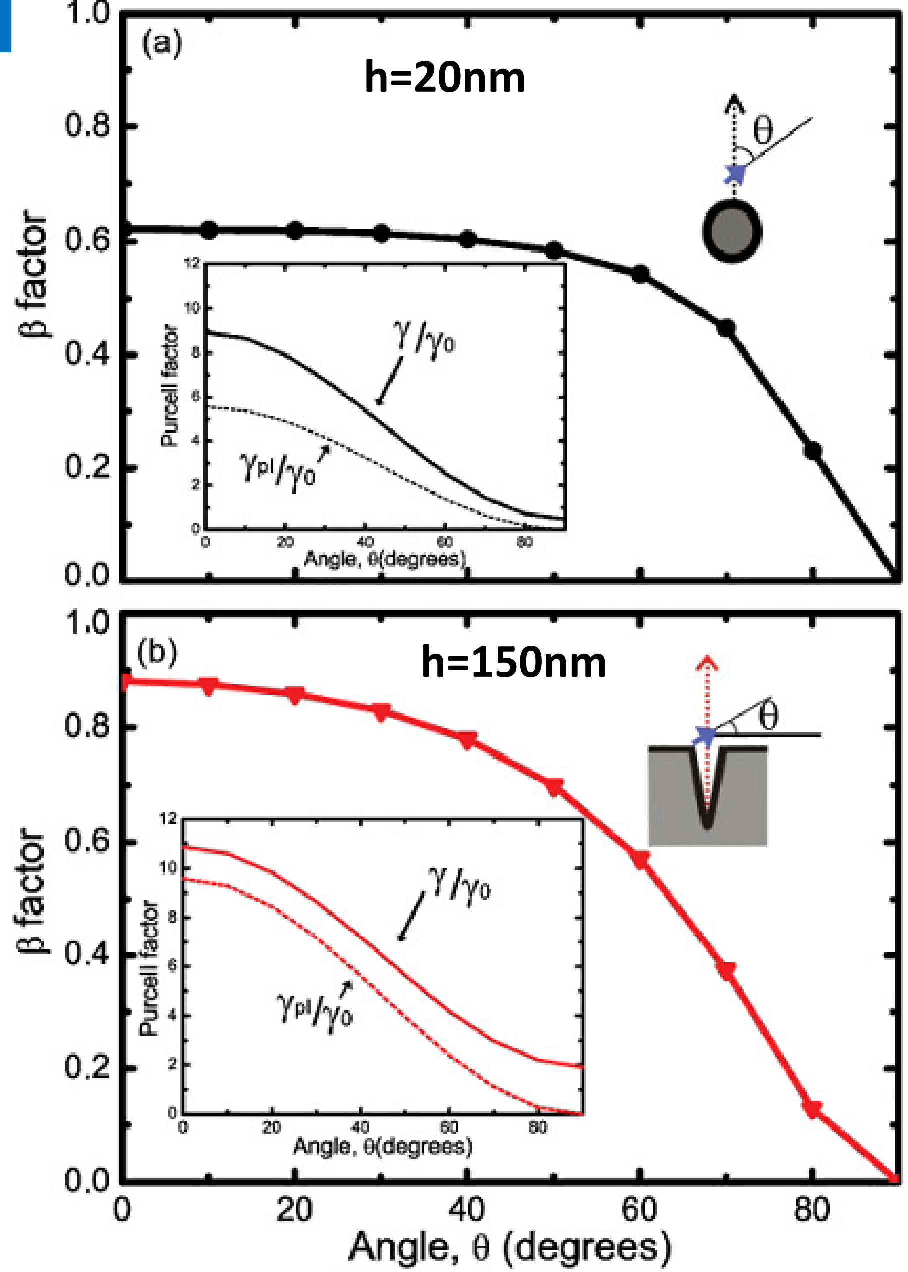




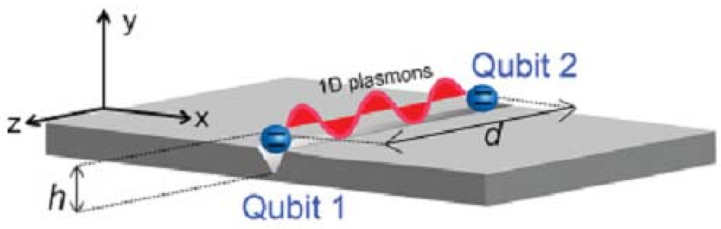
The channel is more convenient than the cilinder

β and Purcell factors

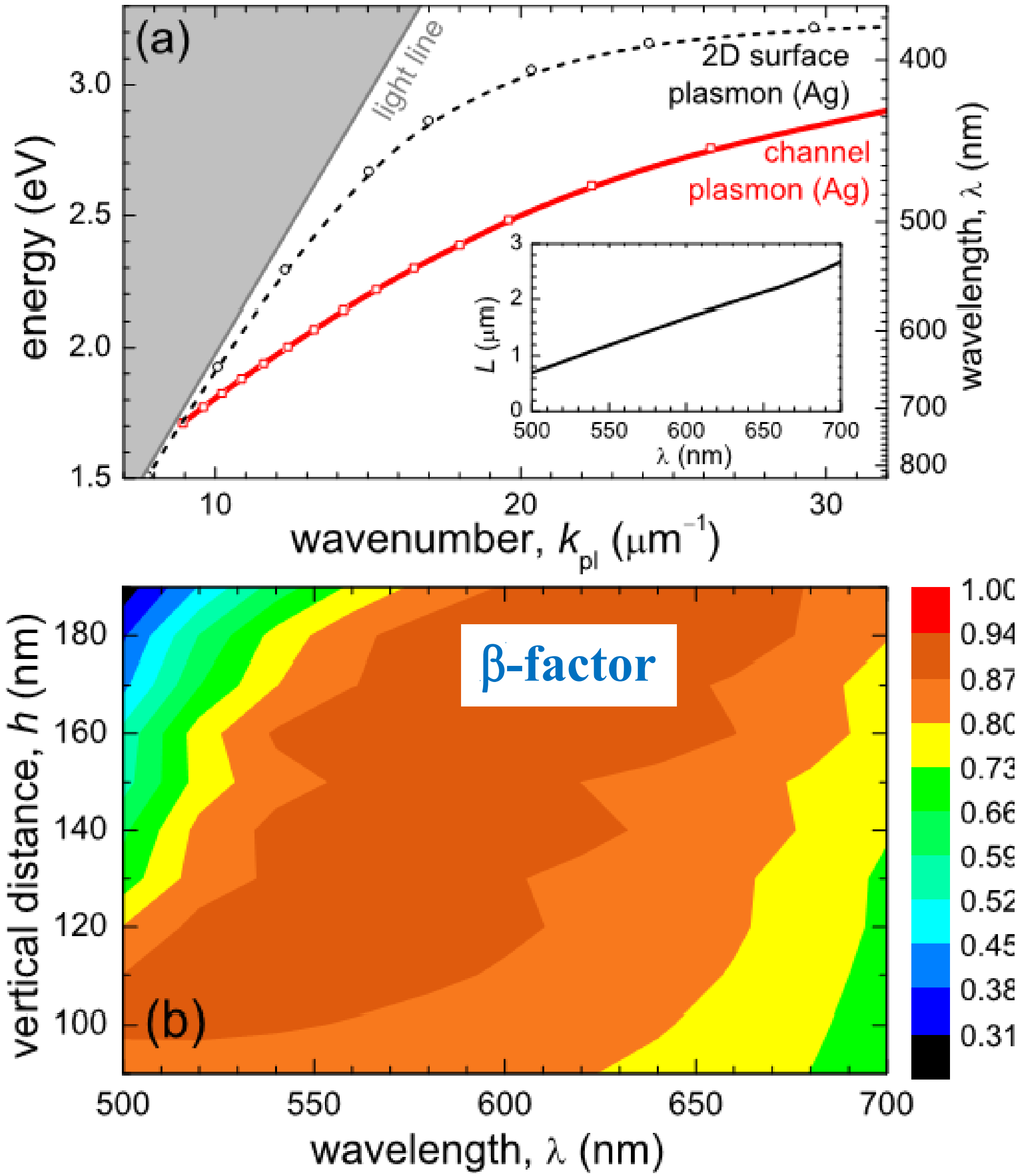
β- factor is very stable in a broad range of dipole orientations while Purcell factor decreases significantly when the dipole is not properly oriented



Dispersion & β factor for V-channel



h=140nm
V-angle= 20 degrees



Two QE's dynamics

All the degrees of freedom (SPP, dissipation, radiation) can be traced out producing effective coherent & incoherent interactions between the two QE's that can be computed from the classical Green's function:

$$\hat{H} = \int d^3\mathbf{r} \int_0^\infty d\omega \hbar\omega a^\dagger(\mathbf{r},\omega) a(\mathbf{r},\boldsymbol{\omega}) + \sum_{i=1,2} \hbar\omega_0 \hat{\sigma}_i^\dagger \hat{\sigma}_i - \sum_{i=1,2} \int_0^\infty d\omega [\hat{\mathbf{d}}_i \hat{\mathbf{E}}(\mathbf{r}_i,\omega) + h.c.]$$

$$\hat{\mathbf{E}}(\mathbf{r},\omega) = i\sqrt{\frac{\hbar}{\pi\epsilon_0}\frac{\omega^2}{c^2}} \int d^3\mathbf{r}' \sqrt{\varepsilon_i(\mathbf{r}',\omega)} \mathbf{G}(\mathbf{r},\mathbf{r}',\boldsymbol{\omega}) a(\mathbf{r}',\omega)$$

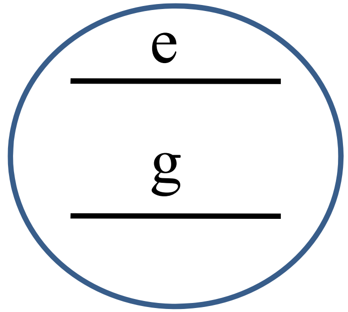
$$\frac{\partial\hat{\rho}}{\partial t} = -\frac{i}{\hbar}[\hat{H}_s + \hat{H}_L, \hat{\rho}] - \frac{1}{2}\sum_{i,j} \gamma_{ij} (\hat{\rho}\hat{\boldsymbol{\sigma}}_i^\dagger \hat{\boldsymbol{\sigma}}_j + \hat{\boldsymbol{\sigma}}_i^\dagger \hat{\boldsymbol{\sigma}}_j \hat{\rho} - 2\hat{\boldsymbol{\sigma}}_j \hat{\rho} \hat{\boldsymbol{\sigma}}_i^\dagger)$$

$$\hat{H}_s = \sum_i \hbar(\omega_0 + \delta_i) \hat{\sigma}_i^\dagger \hat{\sigma}_i + \sum_{i \neq j} \hbar g_{ij} \hat{\sigma}_i^\dagger \hat{\sigma}_j \qquad \hat{H}_L = -\frac{1}{2}\sum_i \hbar\Omega_i \hat{\sigma}_i^\dagger e^{i\omega_L t} + h.c.$$

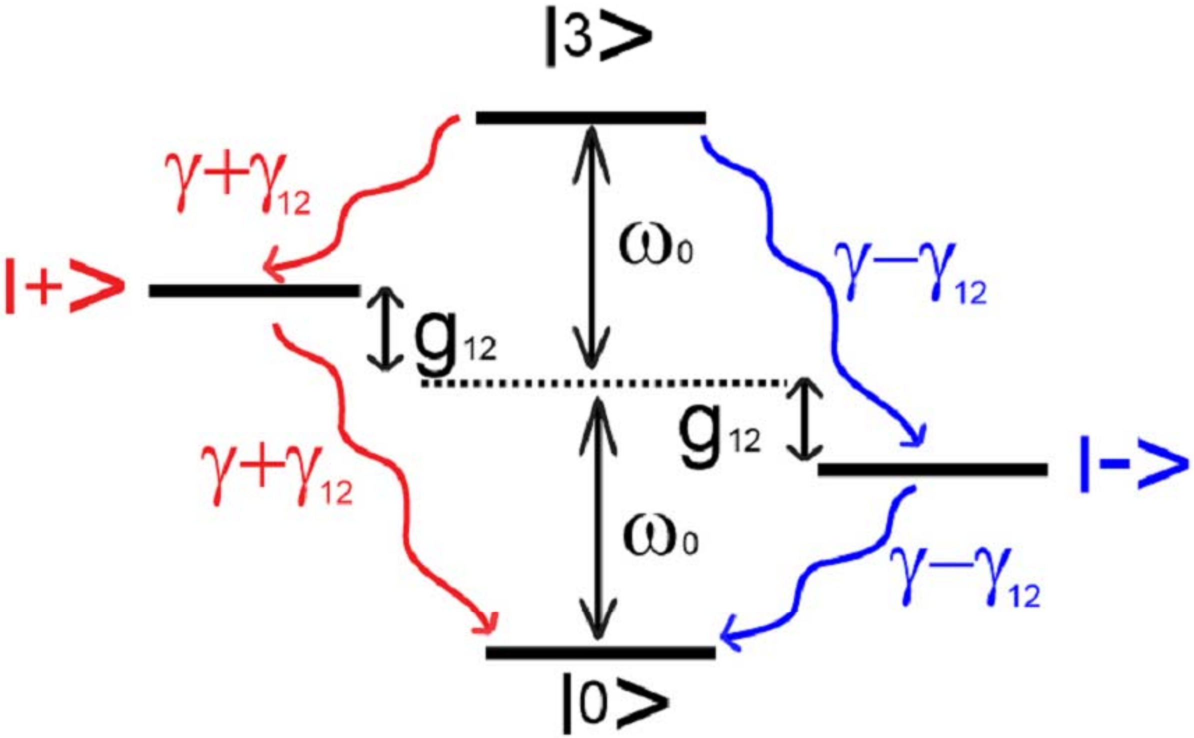
$$g_{ij} = \frac{\omega_0^2}{\hbar\varepsilon_0 c^2} \mathbf{d}_i^* Re\mathbf{G}(\mathbf{r}_i, \mathbf{r}_j, \omega_0) \mathbf{d}_j \qquad \gamma_{ij} = \frac{2\omega_0^2}{\hbar\varepsilon_0 c^2} \mathbf{d}_i^* Im\mathbf{G}(\mathbf{r}_i, \mathbf{r}_j, \omega_0) \mathbf{d}_j$$

Scheme of levels

QE



$$|3\rangle = |e_1 e_2\rangle \quad |0\rangle = |g_1 g_2\rangle \quad |\pm\rangle = \frac{1}{\sqrt{2}}\left(|g_1 e_2\rangle \pm |e_1 g_2\rangle\right)$$



Modulation of γ_{12} would allow to swicth on/off red and blue paths

Two QE's dynamics

It is possible to identify the effects of SPP & dissipation

SPP Green's function

$$\mathbf{G}_{SPP}(\mathbf{r},\mathbf{r}') = \frac{i\,\mathbf{E}^{t}(\mathbf{r}^{t}) \otimes \mathbf{E}^{t}(\mathbf{r}'^{t})}{2\omega\mu_0 \int_{S_\infty} dS\,\mathbf{u}_z(\mathbf{E}^{t} \times \mathbf{H}^{*t})} e^{ik(z-z')}$$

⇓

Effective interaction

$$\mathcal{L}(\rho) = J_{1,2}(\sigma_1 \rho \sigma_2^{\dagger} - \rho \sigma_2^{\dagger} \sigma_1) + h.c.$$

$$J_{1,2} = \frac{J(\omega_0)}{2} e^{iq(\omega_0)|x_1 - x_2|}$$

Coherent

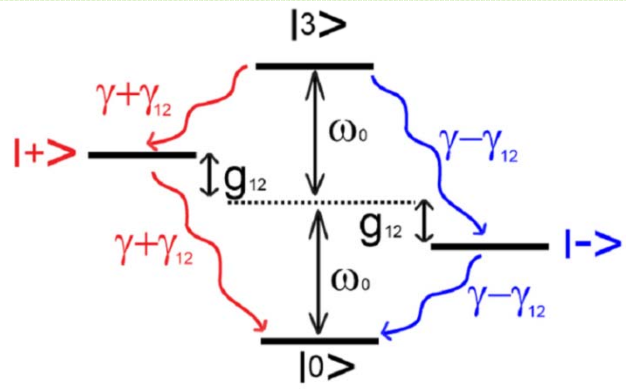
$$g_{ij} \simeq g_{ij,\mathrm{pl}} = \frac{\gamma}{2} \beta e^{-d/2\ell} \sin(k_r d)$$

Incoherent

$$\gamma_{ij} \simeq \gamma_{ij,\mathrm{pl}} = \gamma \beta e^{-d/2\ell} \cos(k_r d)$$

π/2 shift allows switching on/off

- **Coherent versus incoherent interactions**
- **Control of different decay paths**



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Incoherent

$$\gamma_{ij} \simeq \gamma_{ij,\mathrm{pl}} = \gamma\beta e^{-d/2\ell}\cos(k_r d)$$

$$\frac{\partial\hat{\rho}}{\partial t} = -\frac{i}{\hbar}[\hat{H}_s + \hat{H}_L, \hat{\rho}]$$

$$-\frac{1}{2}\sum_{i,j}\gamma_{ij}(\hat{\rho}\hat{\boldsymbol{\sigma}}_i^\dagger\hat{\boldsymbol{\sigma}}_j + \hat{\boldsymbol{\sigma}}_i^\dagger\hat{\boldsymbol{\sigma}}_j\hat{\rho} - 2\hat{\boldsymbol{\sigma}}_j\hat{\rho}\hat{\boldsymbol{\sigma}}_i^\dagger)$$

$$\hat{H}_s = \sum_i \hbar(\omega_0 + \delta_i)\hat{\sigma}_i^\dagger\hat{\sigma}_i + \sum_{i\neq j}\hbar g_{ij}\hat{\sigma}_i^\dagger\hat{\sigma}_j$$

$$\hat{H}_L = -\frac{1}{2}\sum_i \hbar\Omega_i\hat{\sigma}_i^\dagger e^{i\omega_L t} + h.c.$$

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- **Coherent versus incoherent interactions**
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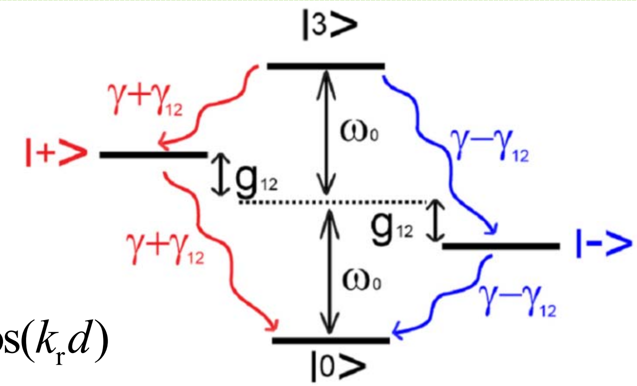
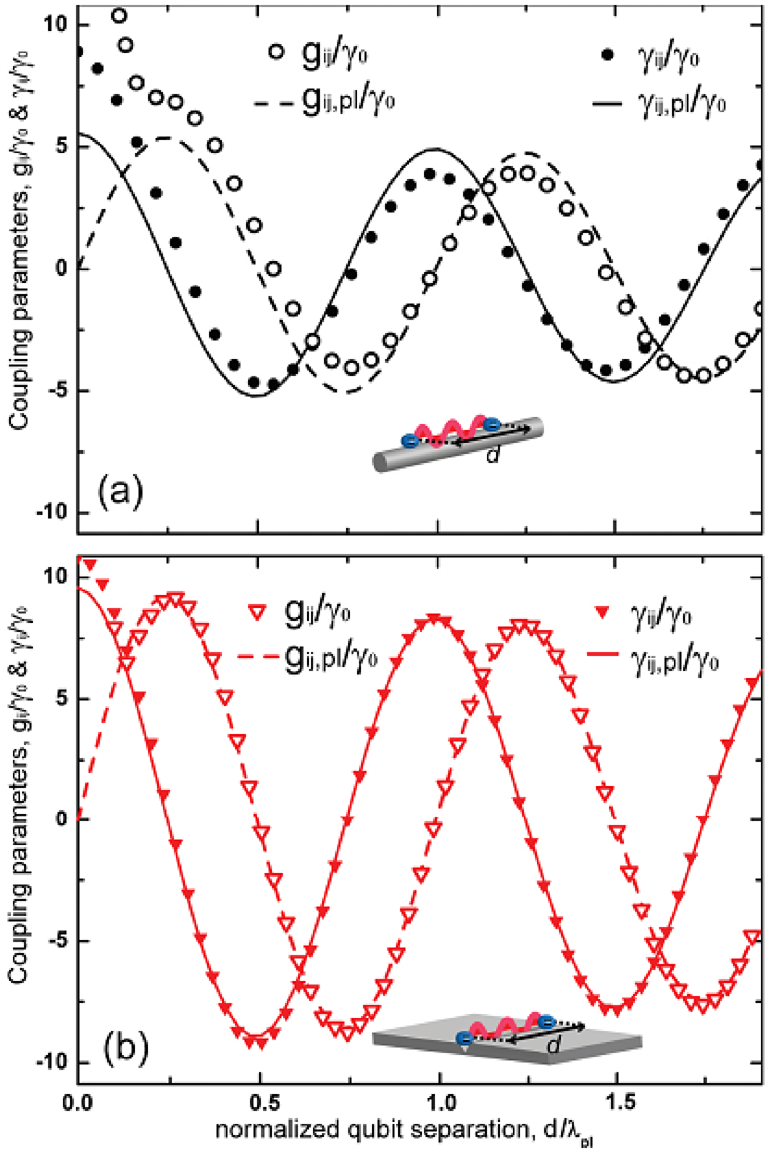


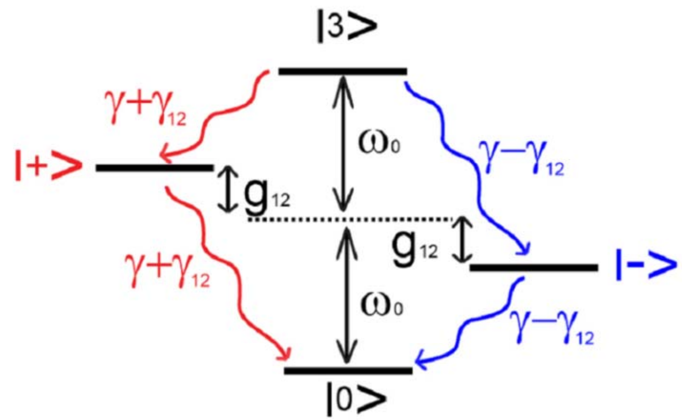
For inequivalent dipoles $\gamma_{ij,\mathrm{pl}} = \sqrt{\gamma_{ii}\gamma_{jj}}\sqrt{\beta_i\beta_j}e^{-d/2\ell}\cos(k_r d)$

Coherent (g_{ij}) & incoherent (γ_{ij}) effective couplings between QE's



Incoherent coupling much more important than the coherent one because it switches on/off each decay path with respect to the other

$$|\gamma_{12}| \leq \gamma$$



Entanglement measure

Concurrence

Complex definition

Wooters, PRL 80, (1988)

$$C(\rho) = \max(0, \lambda_1 - \lambda_2 - \lambda_3 - \lambda_4)$$
$$R = \sqrt{\sqrt{\rho}\tilde{\rho}\sqrt{\rho}}$$
$$\tilde{\rho} = (\sigma_y \otimes \sigma_y)\rho^*(\sigma_y \otimes \sigma_y)$$

What we need to know: for **pure states**

Separable states (e.g. $|0\rangle$) => $C = 0$
Entangled states (e.g. $|-\rangle$) => $C = 1$

Entanglement measure

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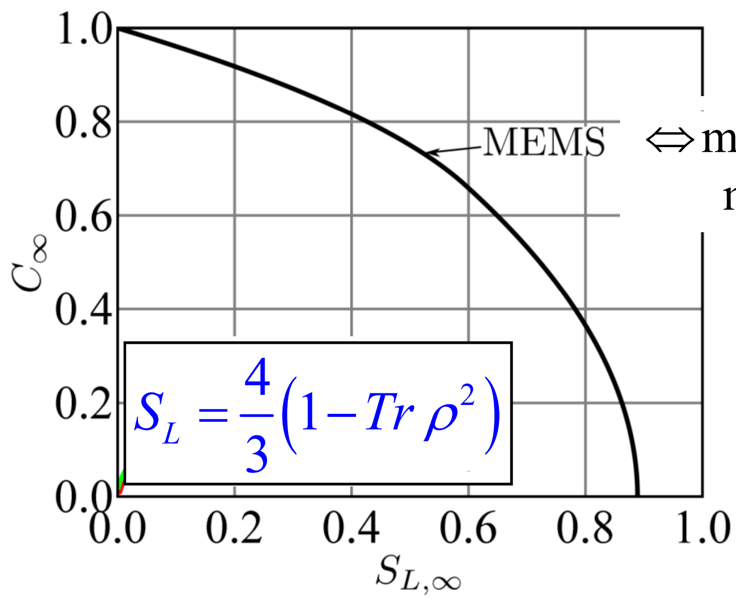
For mixed states:
Concurrence -
Linear entropy
diagram

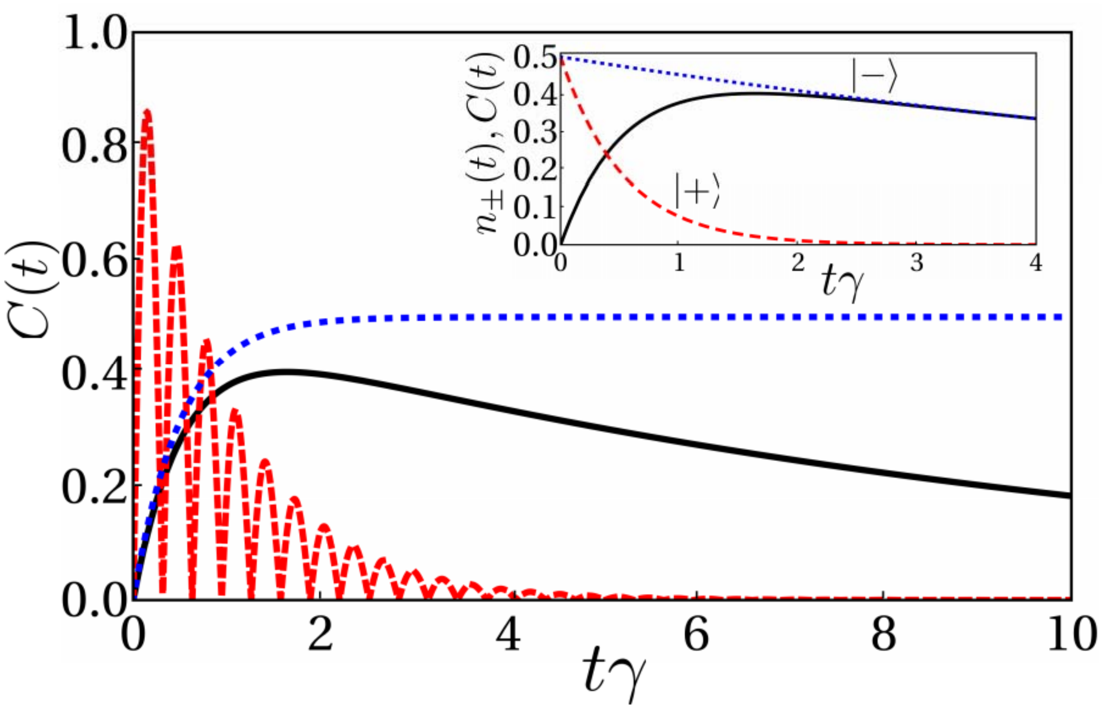


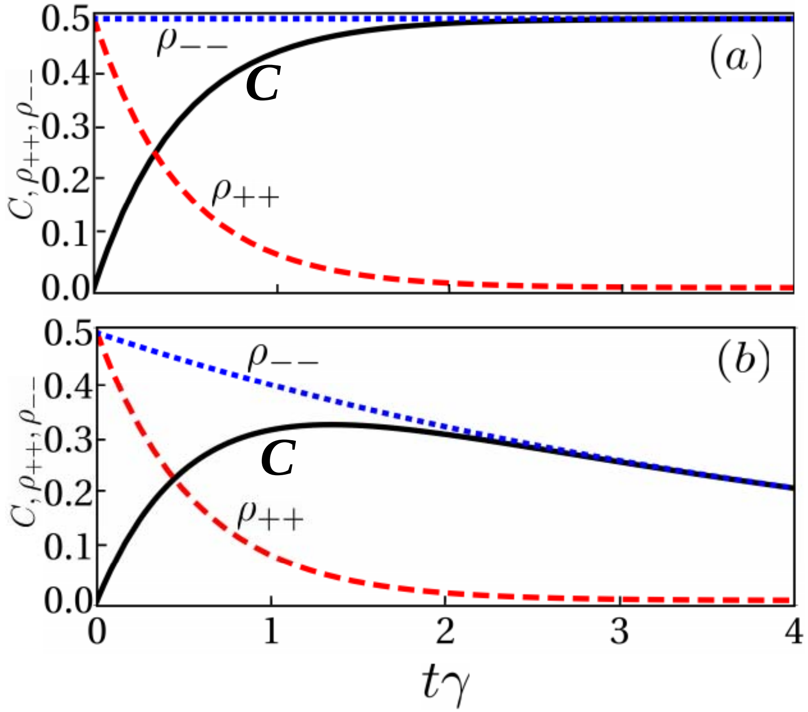
⇔ maximally entangled mixed states

Spontaneous decay of a single excitation

$$|\psi(t=0)\rangle = |1\rangle = |e_1 g_2\rangle = \frac{1}{\sqrt{2}}(|+\rangle + |-\rangle) \quad \Rightarrow$$ **Concurrence becomes:**

$$C(t) = \sqrt{[\rho_{++}(t) - \rho_{--}(t)]^2 + 4Im[\rho_{+-}(t)]^2} = e^{-\gamma t}\sinh[\gamma\beta e^{-\lambda_{\mathrm{pl}}/(2\ell)} t]$$



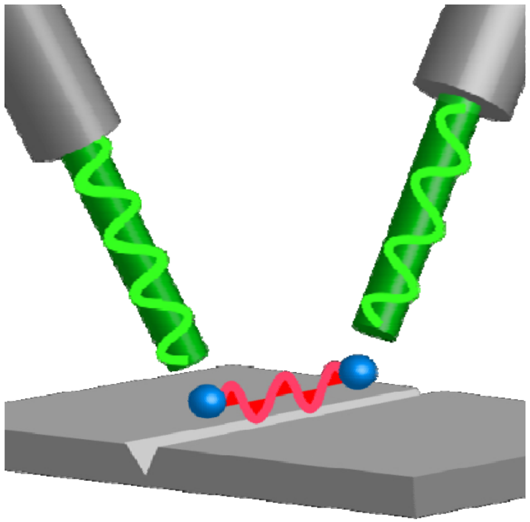


Stationary entanglement

(in the previous viewgraph) Spontaneous decay mediated by plasmons produces finite-time entanglement starting from an unentangled state

$$|\psi(t=0)\rangle = |1\rangle = |e_1 g_2\rangle = \frac{1}{\sqrt{2}}(|+\rangle + |-\rangle)$$

But one wants both to ***obtain*** and ***manipulate*** **stationary entanglement.**
This can be done by means of lasers**:**

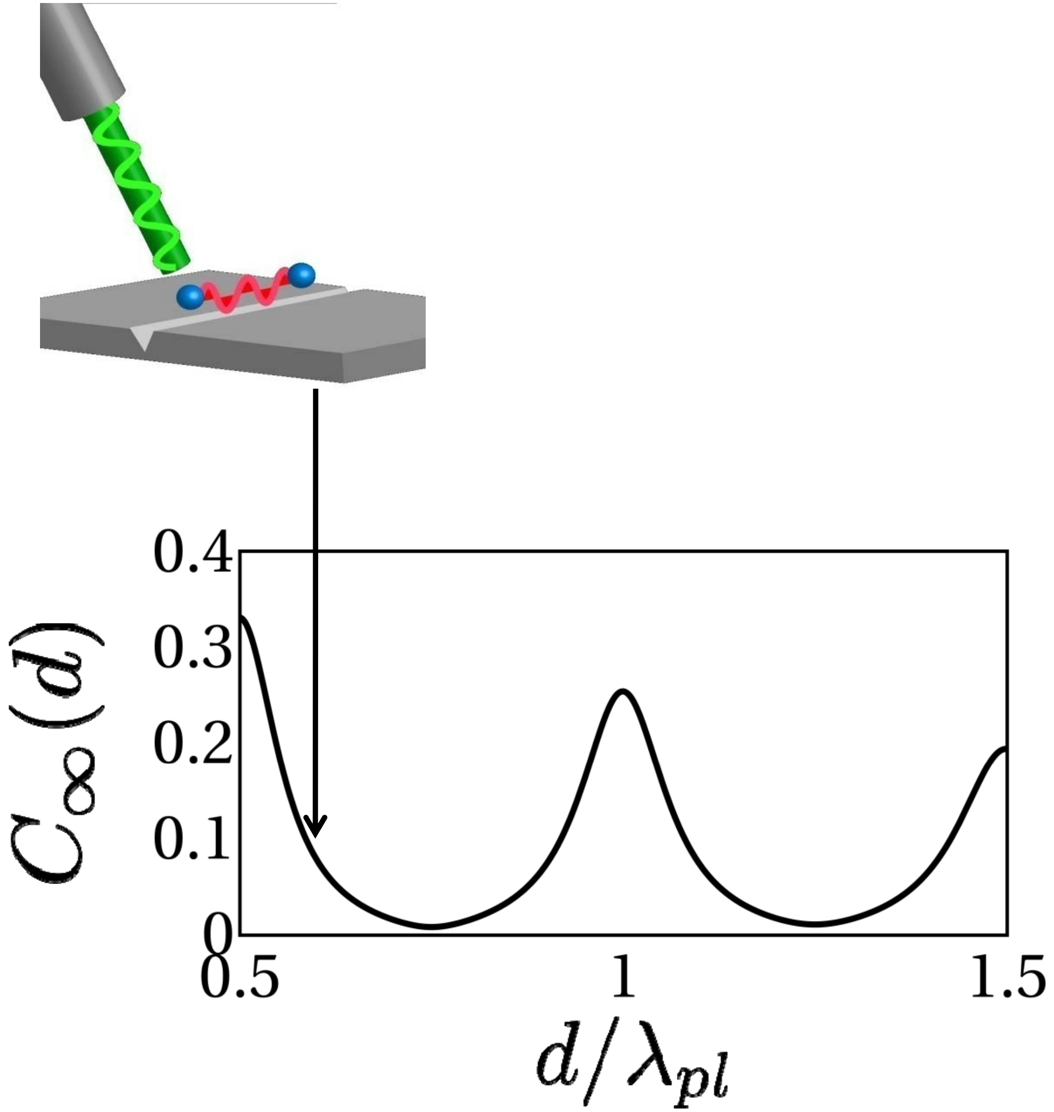


$$H_{las} = \sum_{i=1}^{2} \Omega_i(\sigma_i + \sigma_i^\dagger)$$

In the coherent part of the master equation

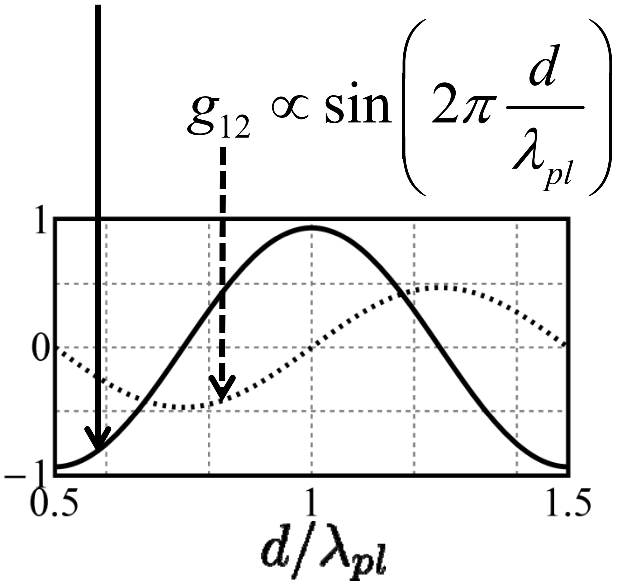
Stationary entanglement

$\Omega_1 \neq 0, \Omega_2 = 0$

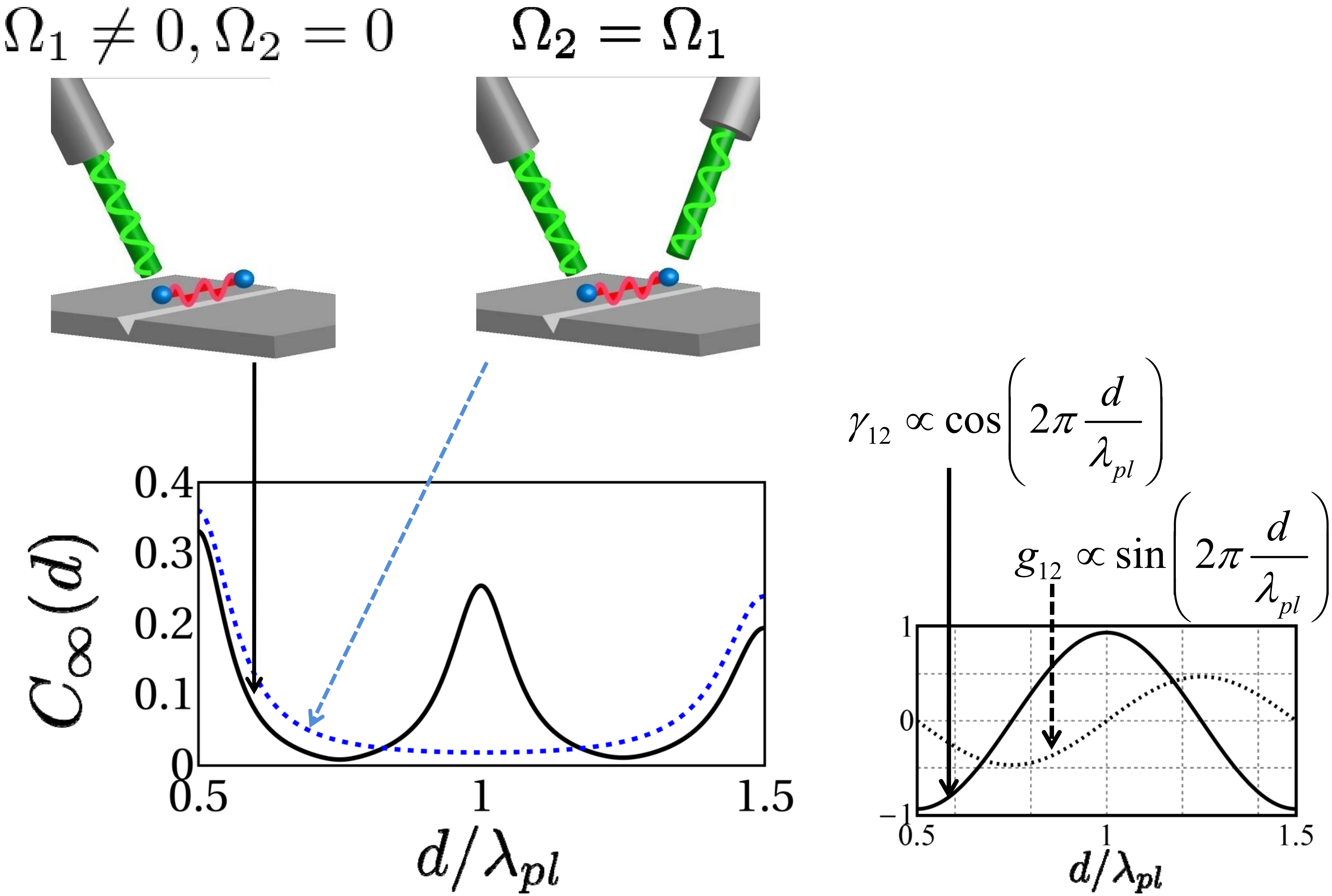


$\gamma_{12} \propto \cos\left(2\pi \frac{d}{\lambda_{pl}}\right)$

$g_{12} \propto \sin\left(2\pi \frac{d}{\lambda_{pl}}\right)$



Stationary entanglement



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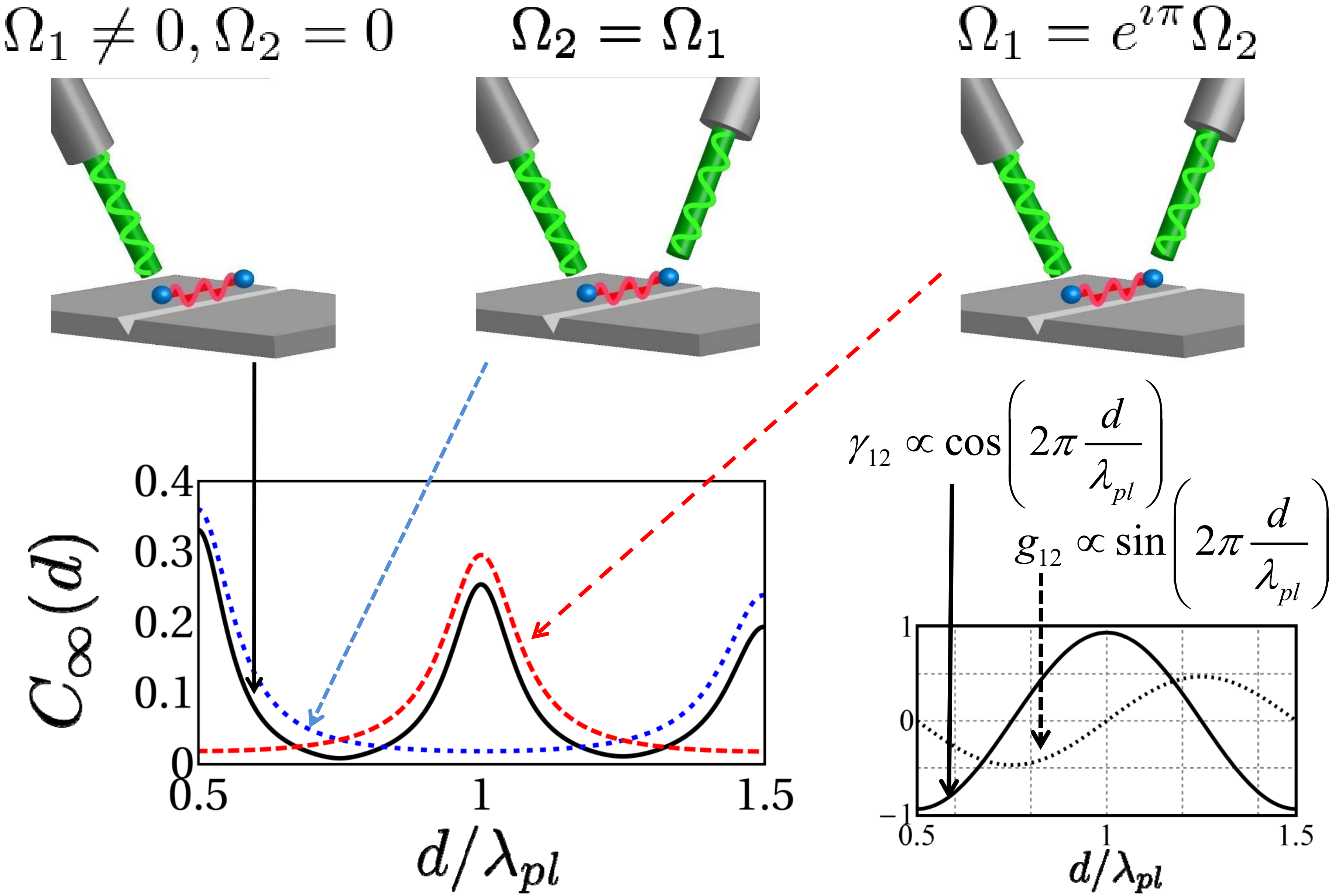


How is stationary entanglement generated?

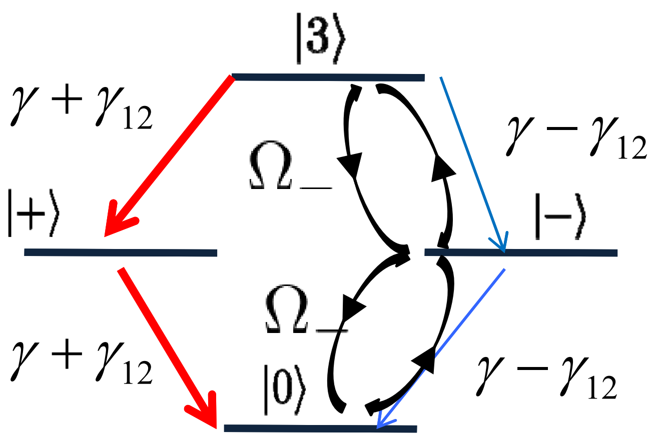
$$\gamma_{12} \propto \cos\left(2\pi \frac{d}{\lambda_{pl}}\right)$$

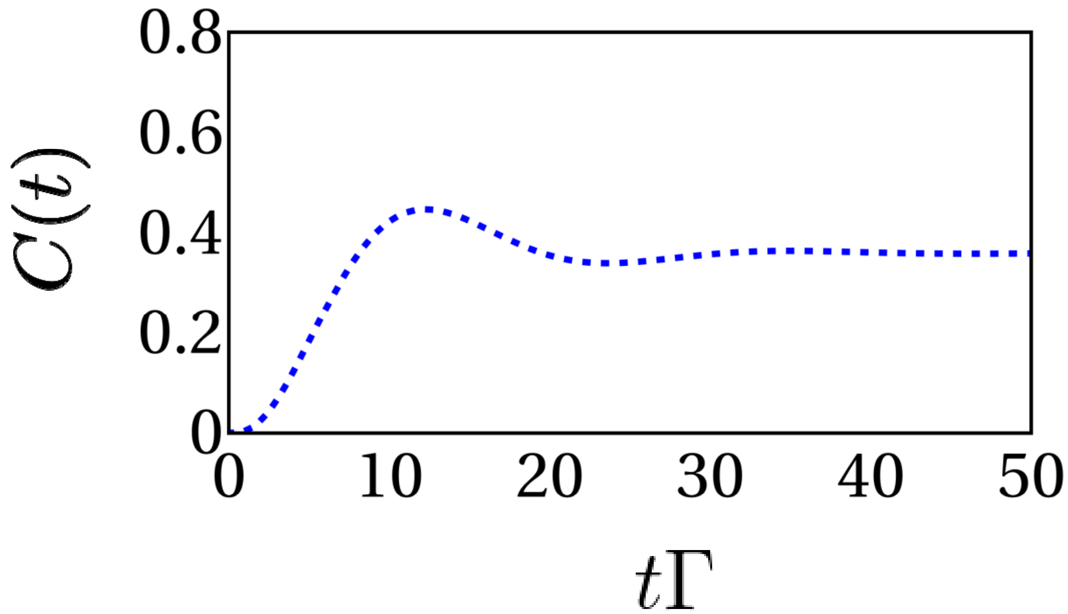
$$d \approx \lambda_{pl}$$

$$\Omega_1 = e^{i\pi}\Omega_2$$

$$\Omega_- = \frac{(\Omega_1 - \Omega_2)}{\sqrt{2}}$$

$$\begin{cases} H_{las}|0\rangle = \Omega_-|-\rangle \\ H_{las}|-\rangle = \Omega_-(|0\rangle + |3\rangle) \\ H_{las}|+\rangle = 0 \\ H_{las}|3\rangle = \Omega_-|-\rangle \end{cases}$$





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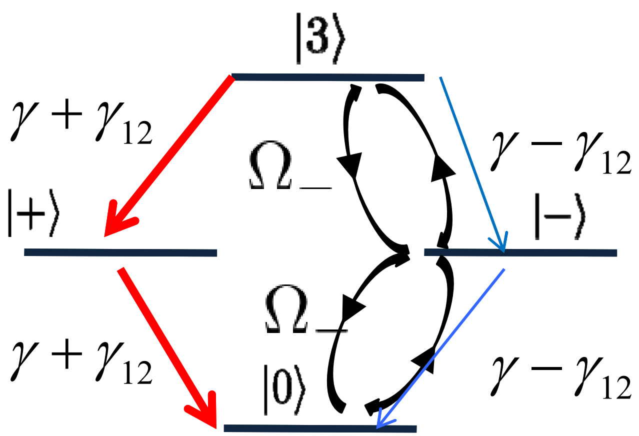

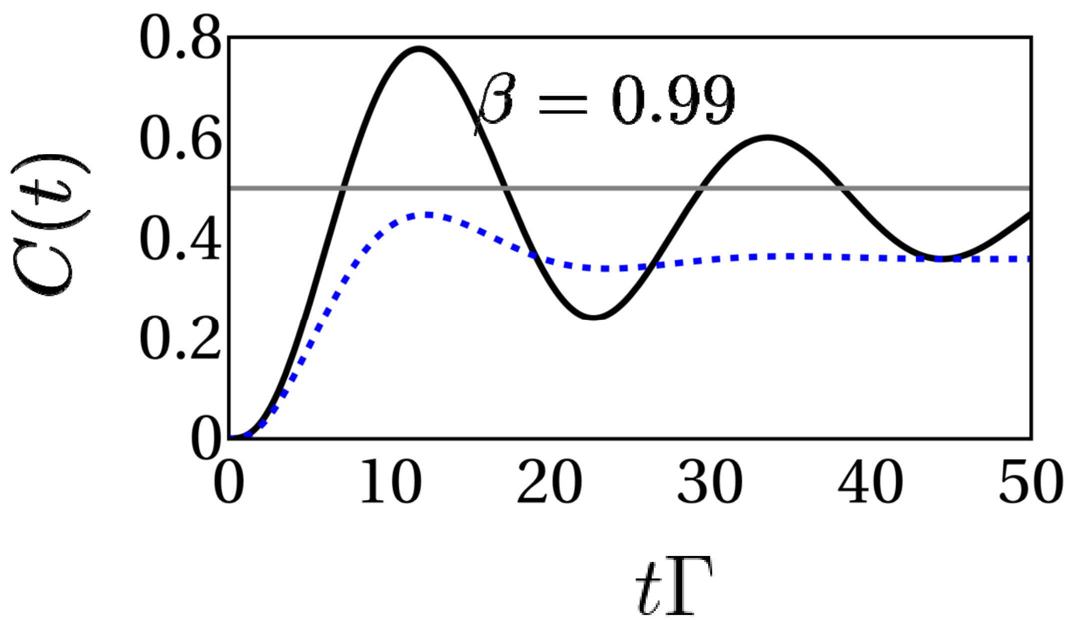

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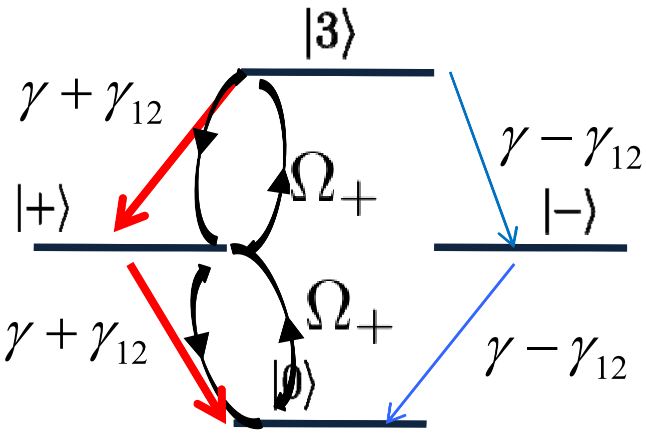
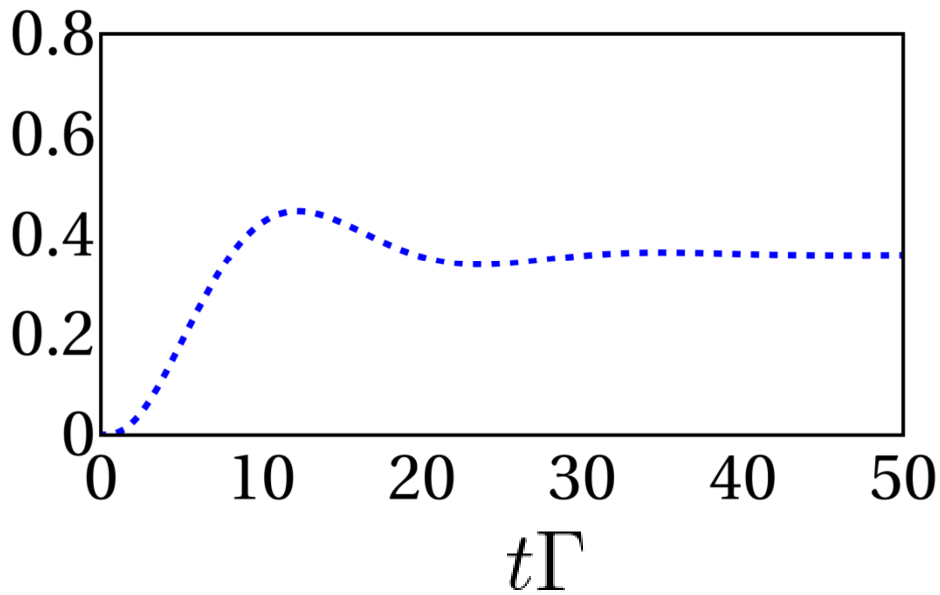
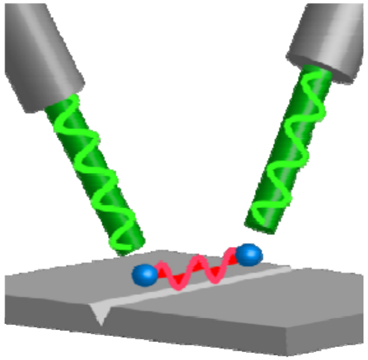
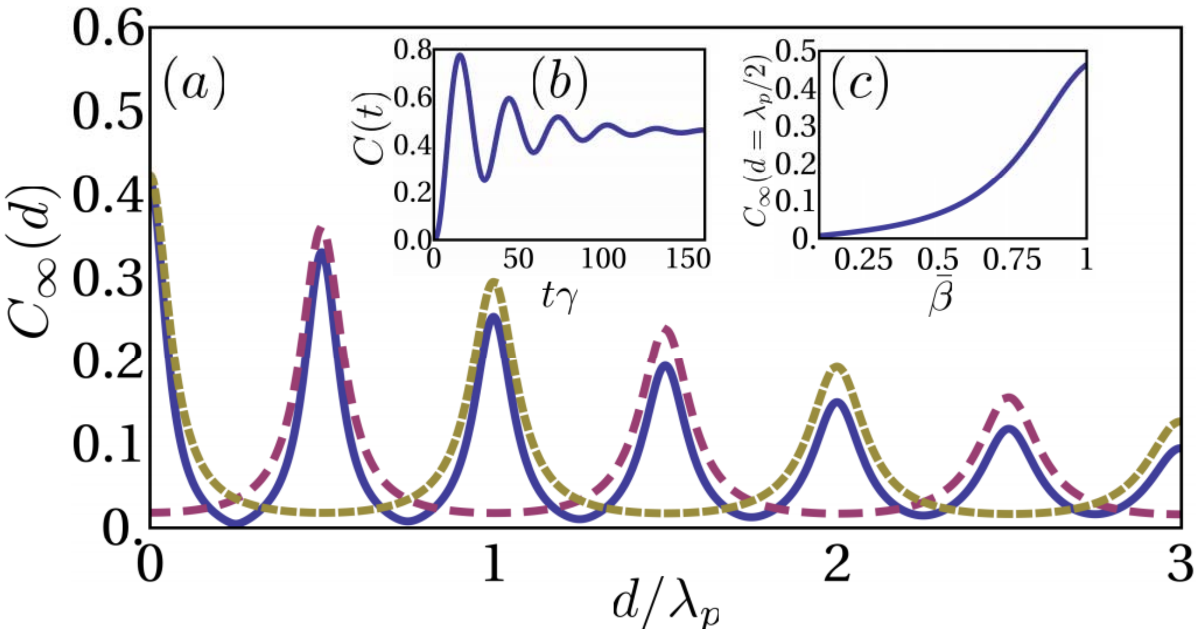
$$\begin{cases} H_{las}|0\rangle = \Omega_+|+\rangle \\ H_{las}|-\rangle = 0 \\ H_{las}|+\rangle = \Omega_+(|0\rangle + |3\rangle) \\ H_{las}|3\rangle = \Omega_+|+\rangle \end{cases}$$





Stationary state concurrence

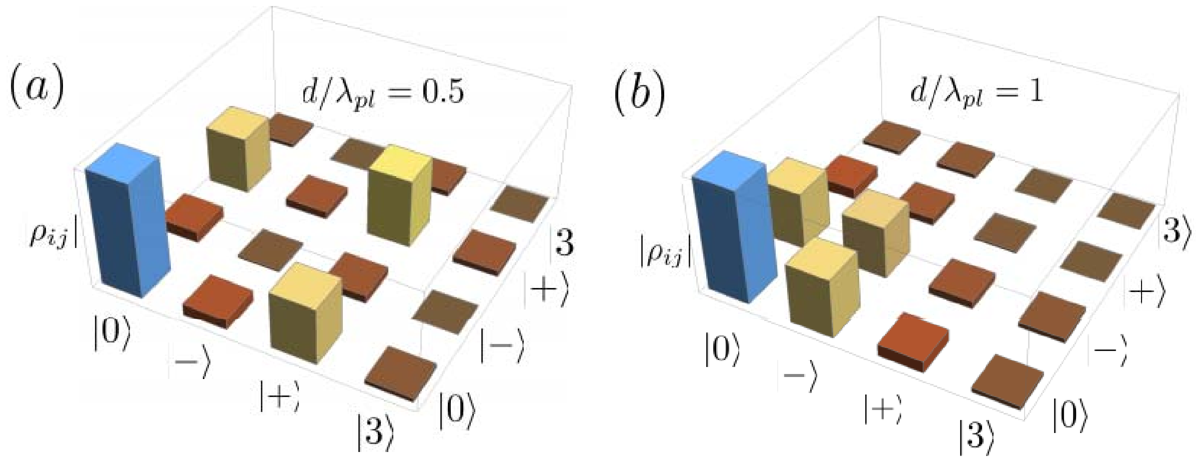




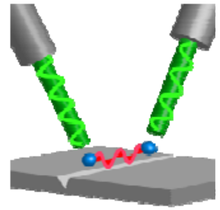
Stationary state tomography

Stationary density matrix

$\Omega_1 = 0.15\gamma,\ \Omega_2 = 0$



How to measure stationary concurrence: QE-QE correlation



Second order cross-coherence

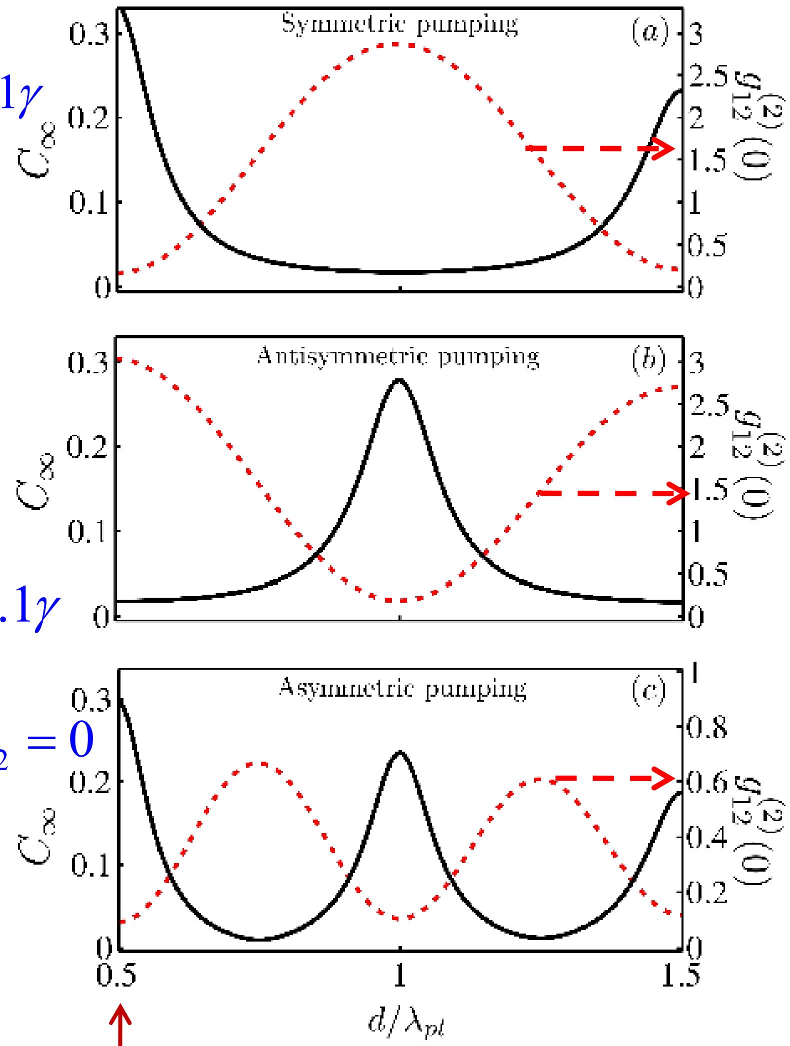
between the two QE's

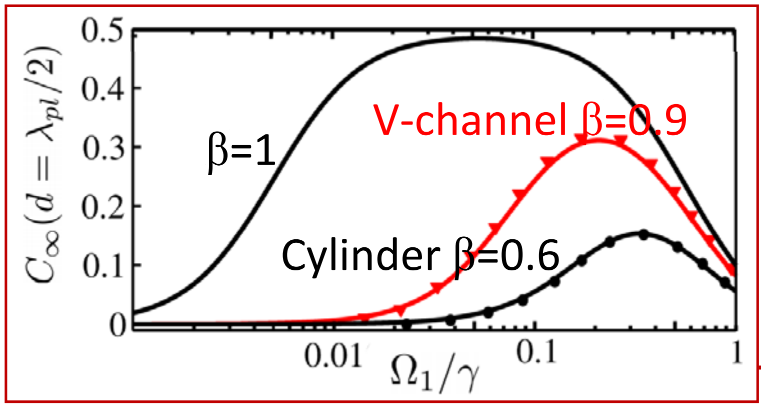
$$g_{12}^{(2)} = \frac{<\sigma_1^\dagger \sigma_2^\dagger \sigma_2 \sigma_1>}{<\sigma_1^\dagger \sigma_1><\sigma_2^\dagger \sigma_2>}$$

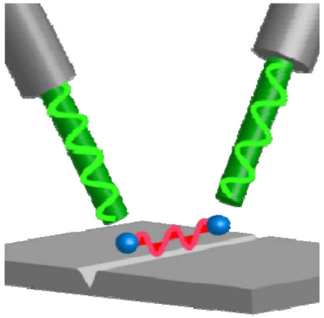
$\Omega_1 = \Omega_2 = 0.1\gamma$

$\Omega_1 = -\Omega_2 = 0.1\gamma$

$\Omega_1 = 0.15\gamma, \Omega_2 = 0$



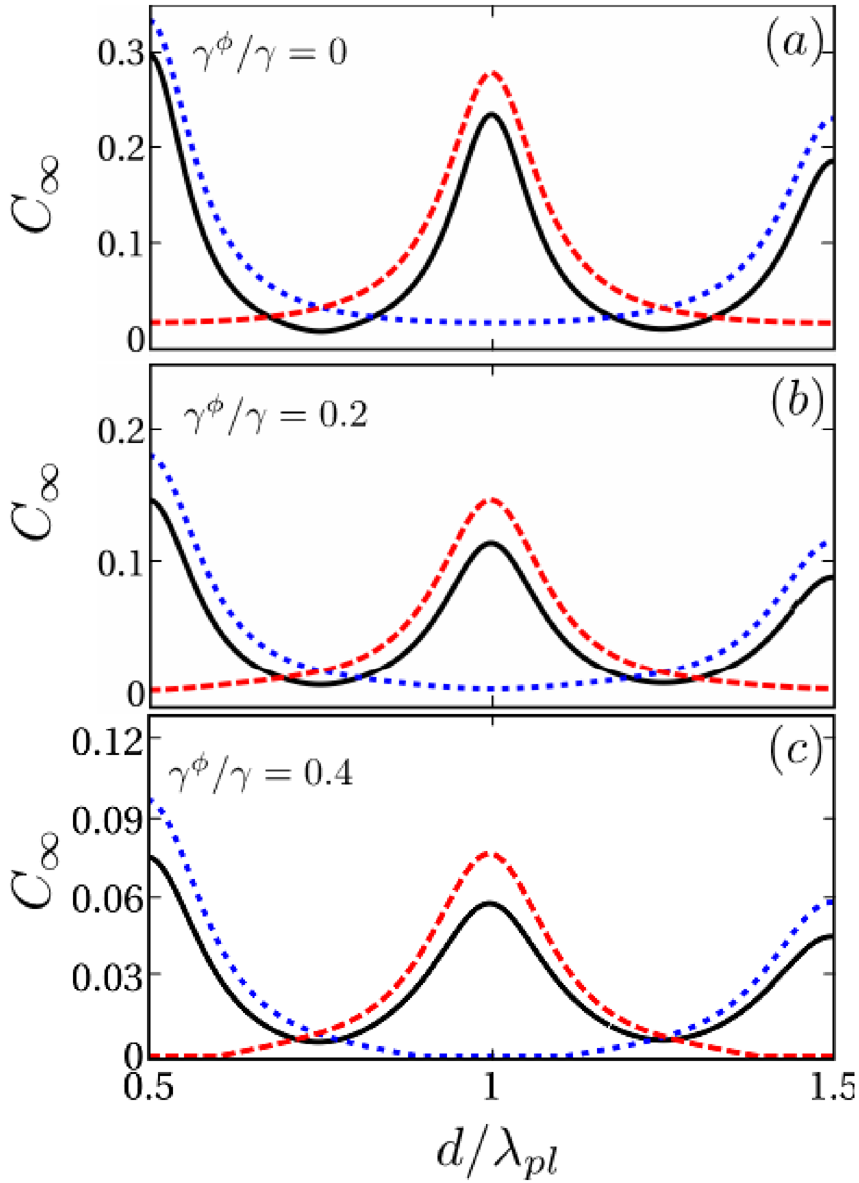




Effect of pure dephasing

$$\mathcal{L}_{\text{deph}}[\hat{\rho}] = \frac{\gamma^{\phi}}{2} \sum_i \left[[\hat{\sigma}_i^{\dagger}\hat{\sigma}_i, \hat{\rho}], \hat{\sigma}_i^{\dagger}\hat{\sigma}_i \right]$$

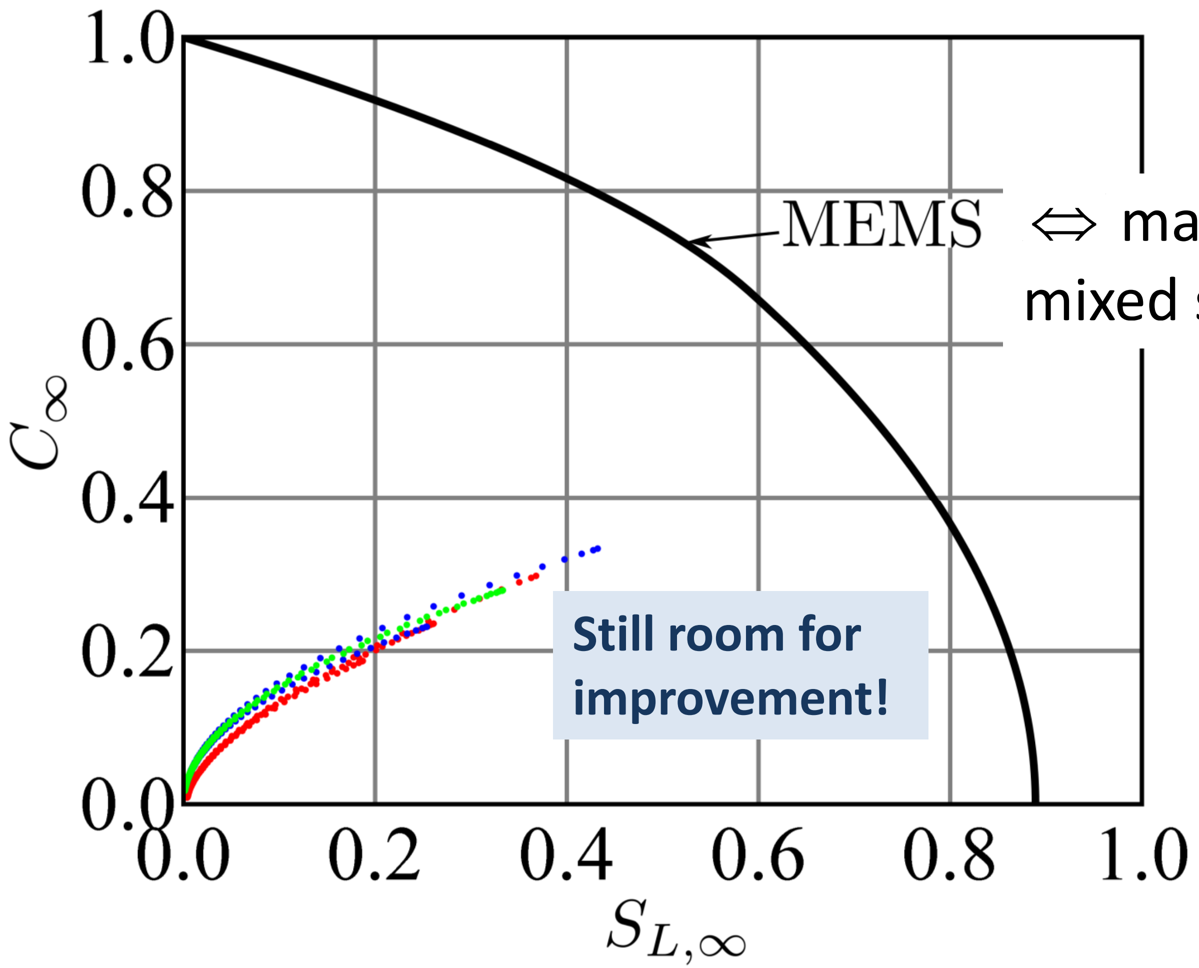
Pure dephasing reduces, but not critically, both correlations & concurrence



Purity

Concurrence - Linear entropy diagram

$$S_L = \frac{4}{3}\left(1 - Tr\,\rho^2\right)$$

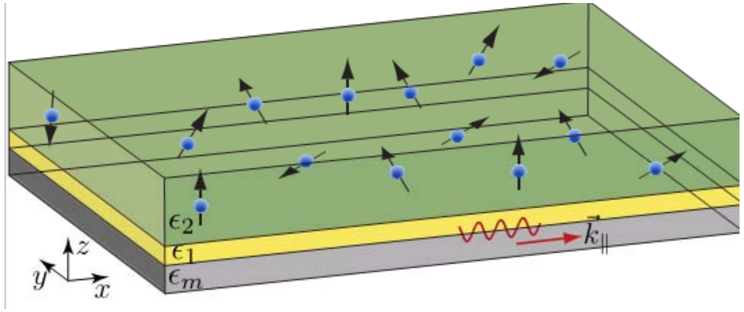


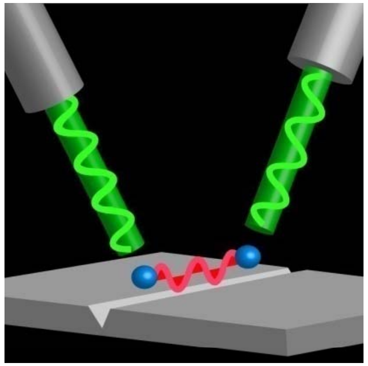
⇔ maximally entangled mixed states

β=0.94
L=2μm
d/λ_{SPP}=0.5-1.5
$\Omega_1=0.15\gamma$; $\Omega_2=0$
$\Omega_1=\Omega_2=0.1\gamma$
$\Omega_1=-\Omega_2=0.1\gamma$

SPP

Strong coupling to excitons & Intermediary for quantum entanglement





Outline

- Introduction to surface plasmon polaritons (SPP)
- Coupling of 1 quantum emitter (QE) to SPP
- Collective mode (excitons) strongly coupled to SPP
- Coupling & entanglement of 2 QE mediated by SPP
- **Conclusion**

Conclusion: Plasmon-polaritons share many properties & capabilities with cavity exciton-polaritons (Cav-QED).

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SPP

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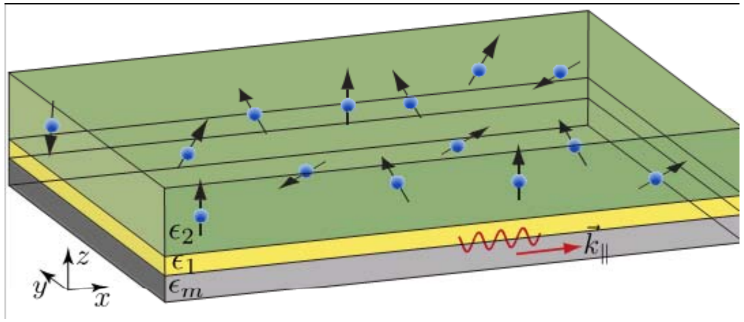

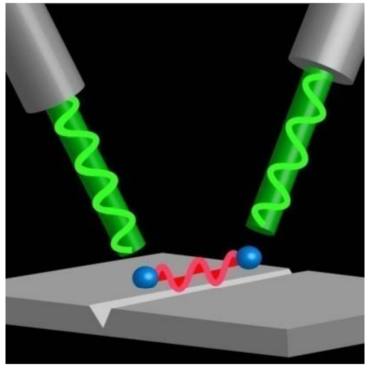

- A. Gonzalez-Tudela *et al*, Phys. Rev. Lett. **110** , 126891 (2013)

- A. Gonzalez-Tudela *et al*, Phys. Rev. Lett. **106** , 020501(2011)
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SPP

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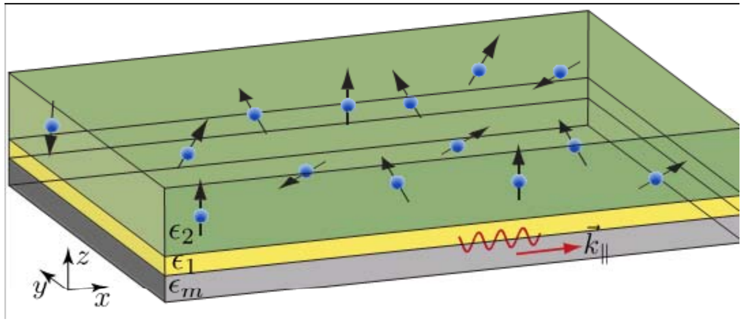

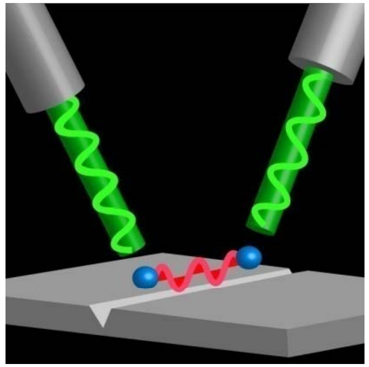

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Thanks for your attention