Multi-emitter stimulated Raman adiabatic passage mediated by plasmons

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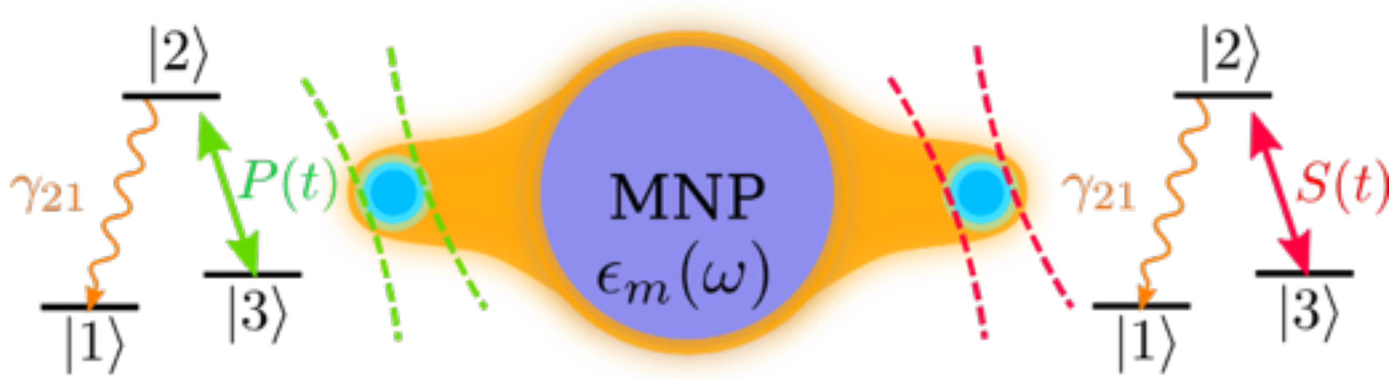


Collaboration: Plasmonics ( ) & Quantum Control ( )

Outline

A. Emitter coupled to a nanosphere

B. Effective model

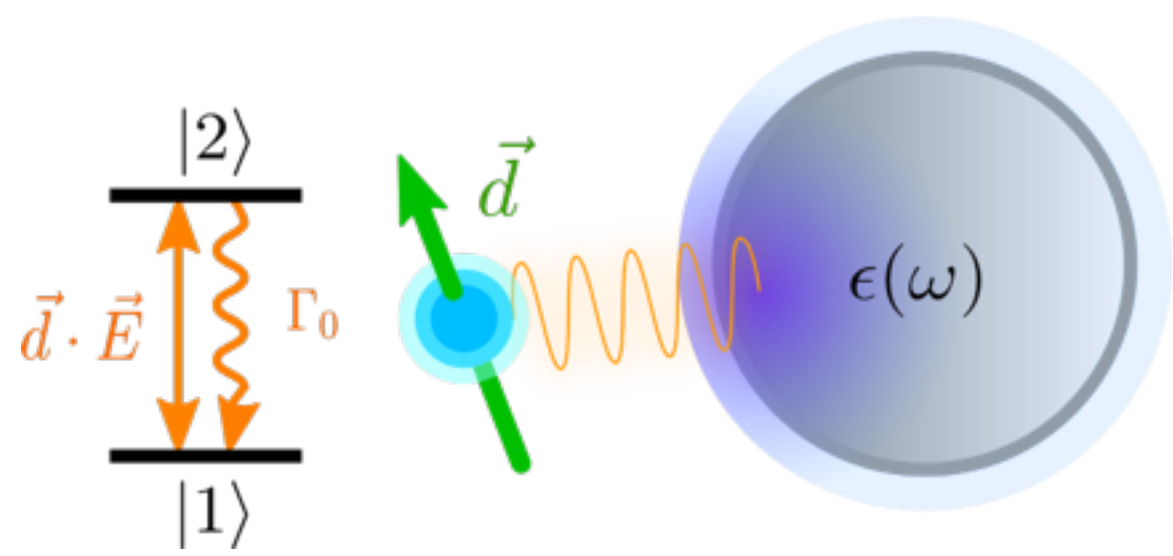
C. Two emitters coupled to a nanosphere

D. Plasmon-mediated STIRAP

Summary

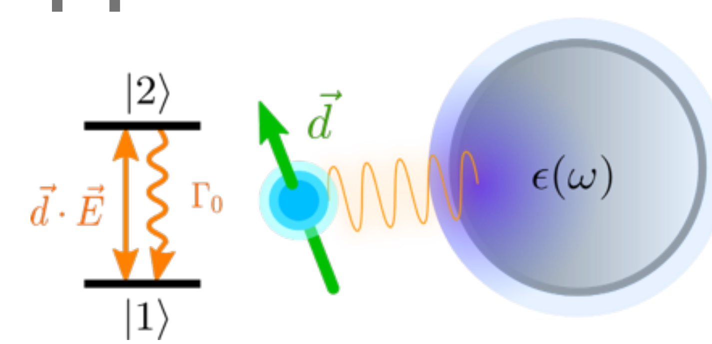
A. Emitter coupled to a nanosphere

- cQED-like models directly with a single mode
- Full quantization & Green’s function approach



A. Emitter coupled to a nanosphere

- Full quantization & Green's function approach



- Electric field operator:

$$\vec{\hat{E}}(\vec{r},\omega) = i\sqrt{\frac{\hbar}{\pi\epsilon_0}}\int \mathrm{d}\vec{r}\,'\frac{\omega^2}{c^2}\sqrt{\epsilon_I(\vec{r}\,',\omega)}\bar{\bar{G}}(\vec{r},\vec{r}\,',\omega)\vec{\hat{f}}_\omega(\vec{r}\,')$$

- Field operators & Green's function:

$$\left[\hat{f}_{\omega,i}(\vec{r}),\hat{f}_{\omega',j}(\vec{r}\,')\right] = \delta_{ij}\delta(\omega-\omega')\delta(\vec{r}-\vec{r}\,')$$

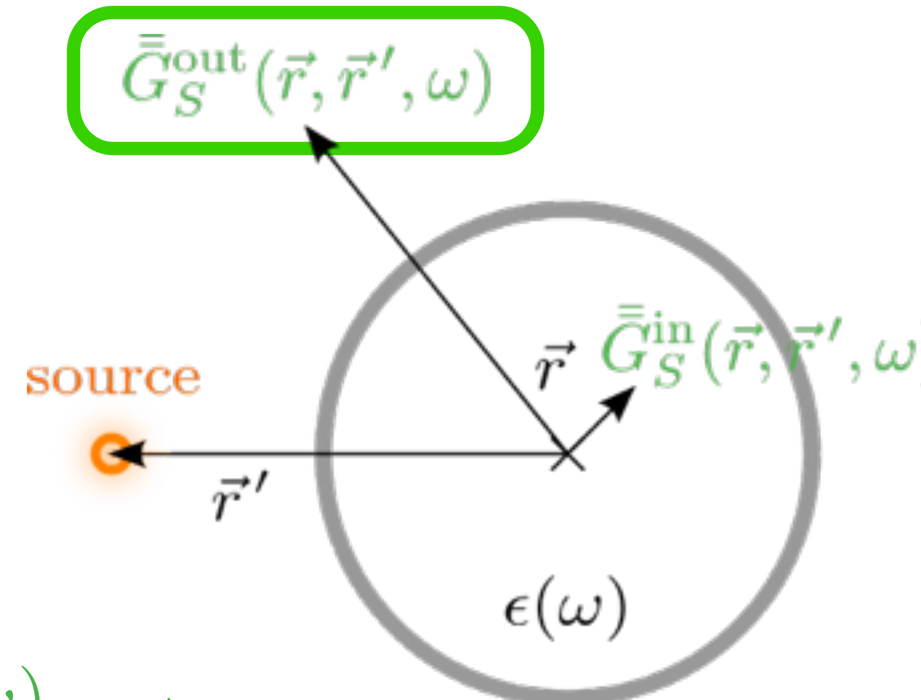
$$\vec{\nabla}\times\vec{\nabla}\times\bar{\bar{G}}(\vec{r},\vec{r}\,',\omega) - \frac{\omega^2}{c^2}\epsilon(\vec{r},\omega)\bar{\bar{G}}(\vec{r},\vec{r}\,',\omega) = \bar{\bar{1}}\delta(\vec{r}-\vec{r}\,')$$

T. Gruner and D.-G. Welsch, Phys. Rev. A **53**, 3 (1996)

A. Emitter coupled to a nanosphere

- The plasmonic modes:

$$\bar{\bar{G}}^{\mathrm{out}}(\vec{r},\vec{r}\,',\omega) = \bar{\bar{G}}_0(\vec{r},\vec{r}\,',\omega) + \bar{\bar{G}}_S^{\mathrm{out}}(\vec{r},\vec{r}\,',\omega)$$



Boundary conditions:

$$\vec{n}_r \times \bar{\bar{G}}_S^{\mathrm{in}}(\vec{r},\vec{r}\,',\omega)_{\vec{r}=\vec{R}^-} = \vec{n}_r \times \bar{\bar{G}}^{\mathrm{out}}(\vec{r},\vec{r}\,',\omega)_{\vec{r}=\vec{R}^+}$$

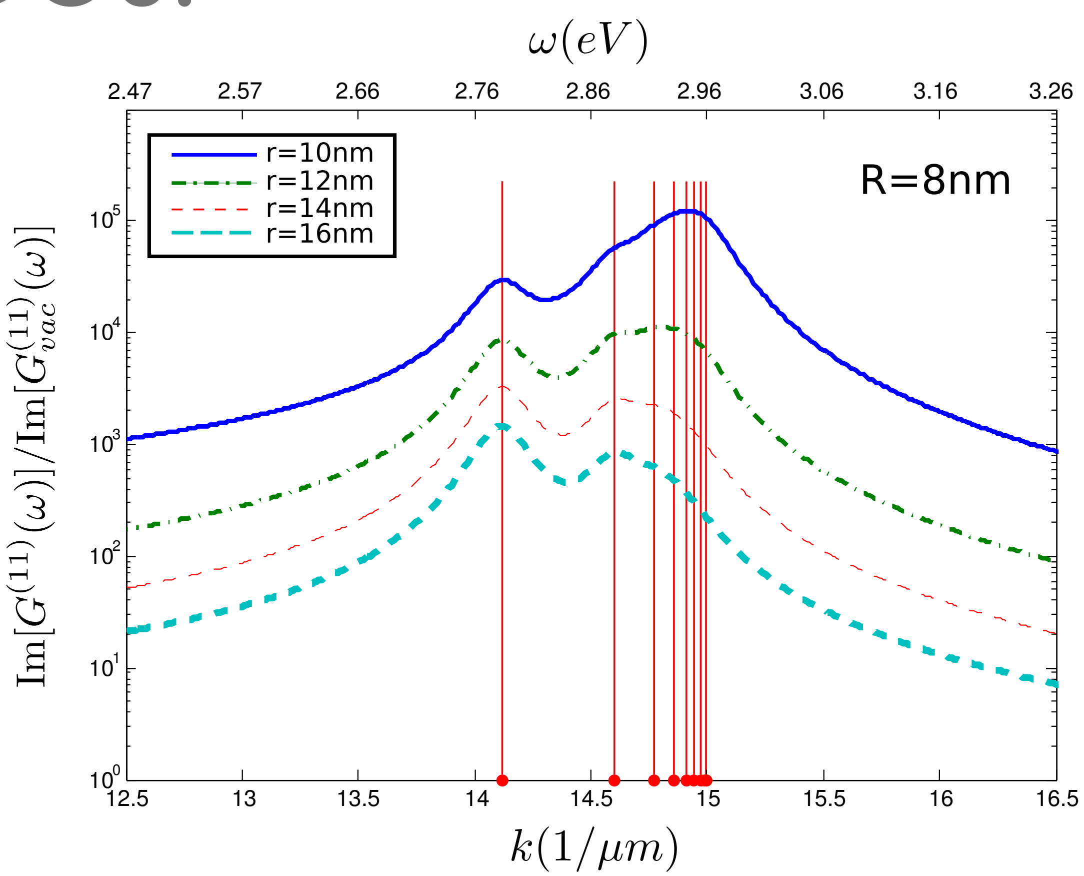
$$\vec{n}_r \times \vec{\nabla} \times \bar{\bar{G}}_S^{\mathrm{in}}(\vec{r},\vec{r}\,',\omega)_{\vec{r}=\vec{R}^-} = \vec{n}_r \times \vec{\nabla} \times \bar{\bar{G}}^{\mathrm{out}}(\vec{r},\vec{r}\,',\omega)_{\vec{r}=\vec{R}^+}$$

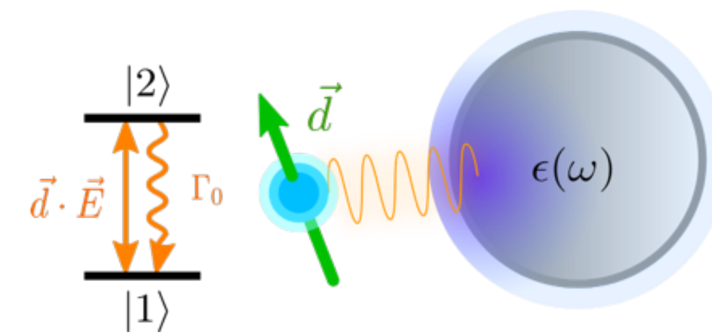
Permittivity model: e.g. Drude model

$$\epsilon(\omega) = \epsilon_\infty - \frac{\omega_p^2}{\omega^2 + i\gamma_e\omega}$$

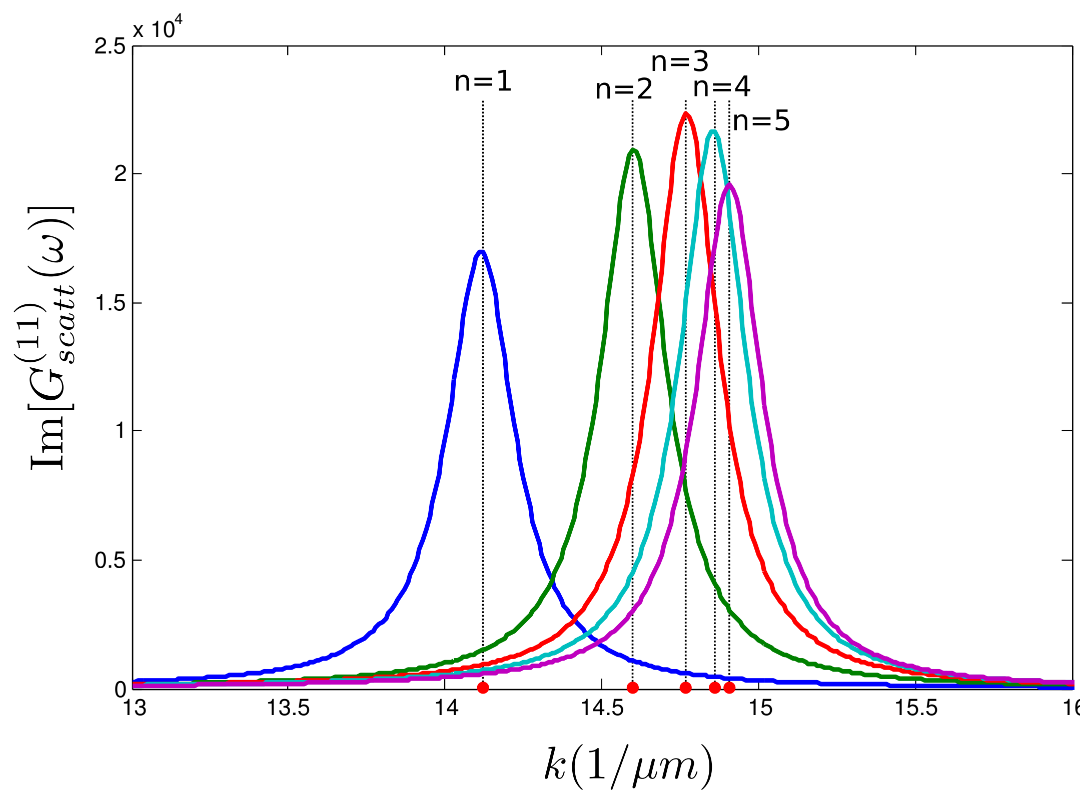
A. Emitter coupled to a nanosphere

- LDOS:





Lorentzian lineshapes

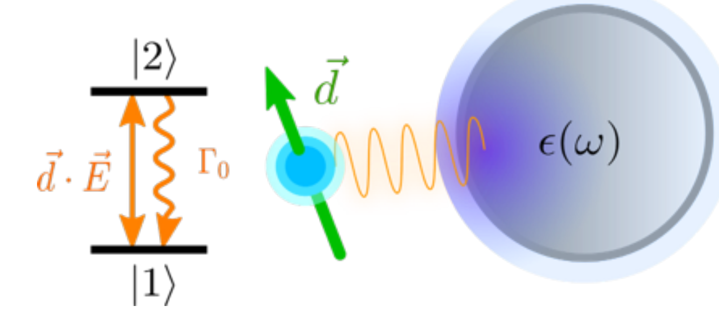


$$\bar{\bar{G}}(\vec{r},\vec{r}\,',\omega) = \sum_n \bar{\bar{G}}^{(n)}(\vec{r},\vec{r}\,',\omega)$$

$$\mathrm{LDOS}(\omega) = \sum_n \frac{(\gamma_n/2)^2\mathrm{LDOS}(\omega_n)}{(\omega-\omega_n)^2 + (\gamma_n/2)^2}$$

B. Effective model

- Full system RWA Hamiltonian:



$$\hat{H} = \hbar\omega_{21}\hat{\sigma}_{22} + \int \mathrm{d}\vec{r} \int_0^{+\infty} \mathrm{d}\omega \hbar\omega \hat{\vec{f}}_\omega^\dagger(\vec{r})\hat{\vec{f}}_\omega(\vec{r}) - \left(\hat{\sigma}_{21}\int_0^{+\infty} \mathrm{d}\omega \vec{d}\cdot\hat{\vec{E}}(\vec{r}_{\mathrm{em}},\omega) + \mathrm{h.c.}\right)$$

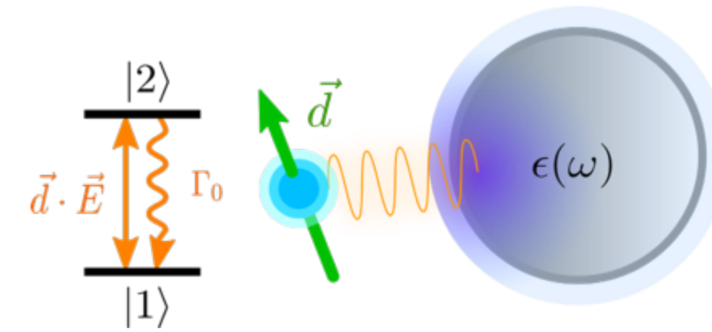
$$\hat{H} = \hbar\omega_{21}\hat{\sigma}_{22} + \int_0^{+\infty} \mathrm{d}\omega \hbar\omega \hat{a}_\omega^\dagger \hat{a}_\omega + i\hbar \int_0^{+\infty} \mathrm{d}\omega \left(\kappa^*(\omega)\hat{a}_\omega^\dagger\hat{\sigma}_{12} - \mathrm{h.c.}\right)$$

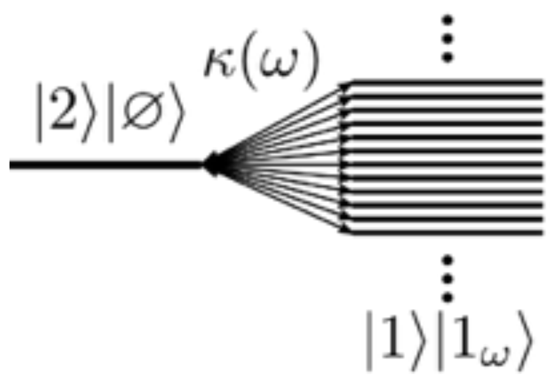
with

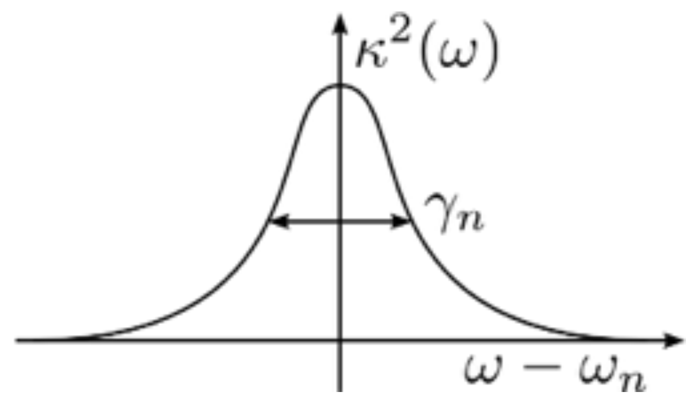
$$|\kappa(\omega)|^2 = \frac{1}{\hbar\pi\epsilon_0}\frac{\omega^2}{c^2}\vec{d}\cdot\mathfrak{Im}\left\{\bar{\bar{G}}(\vec{r}_{\mathrm{em}},\vec{r}_{\mathrm{em}},\omega)\right\}\vec{d}^* = \sum_{n=1}^{N}\frac{\gamma_n}{2\pi}\frac{|\bar{\Omega}_n|^2}{(\omega-\omega_n)^2+\left(\frac{\gamma_n}{2}\right)^2}$$

B. Effective model

- Atom-continuum single Lorentzian coupling:

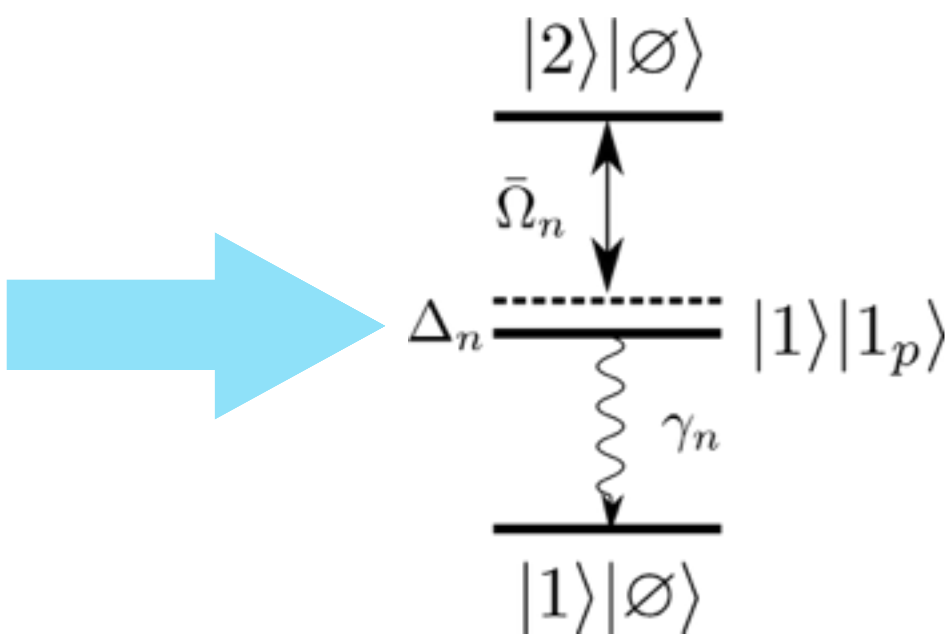






$$\kappa(\omega)=\sqrt{\frac{\gamma_n}{2\pi}}\frac{\bar{\Omega}_n}{\omega-\omega_n+i\frac{\gamma_n}{2}}$$

Plasmon dressed state basis: $|\mathrm{emitter}\rangle\otimes|\mathrm{plasmons}\rangle$

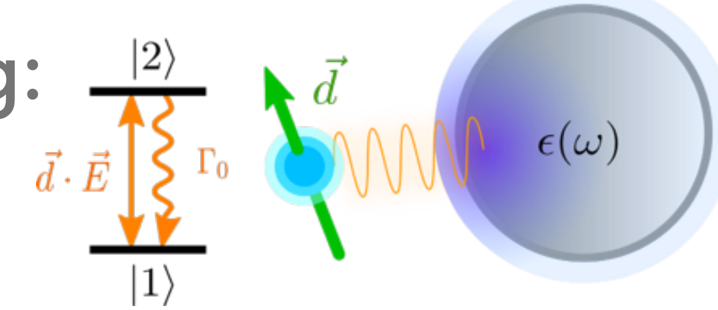


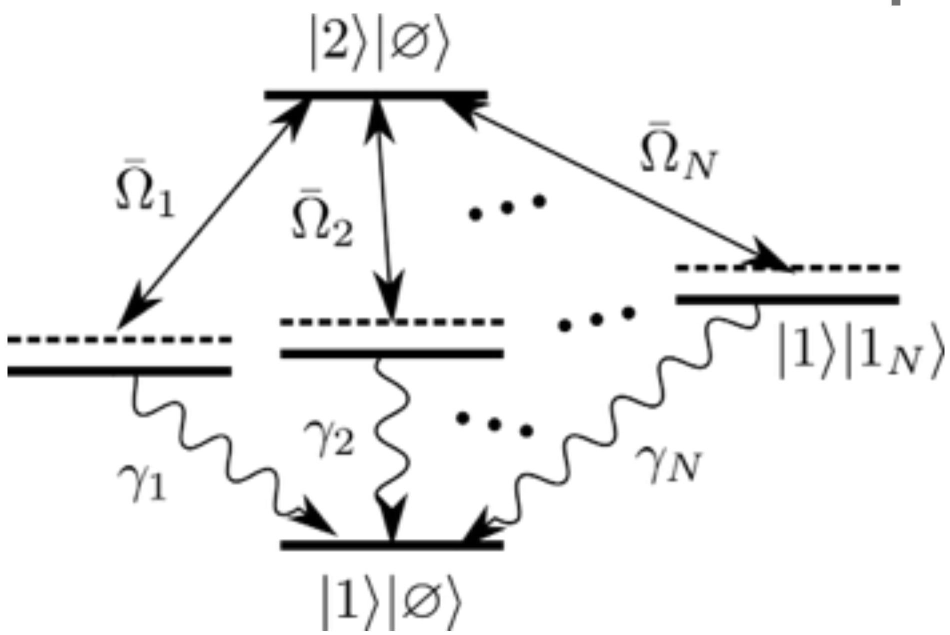
$$|1_p\rangle=\frac{1}{\bar{\Omega}_n^*}\int_0^{+\infty}\mathrm{d}\omega\,\kappa^*(\omega)\hat{a}_\omega^\dagger|\varnothing\rangle$$

$$\begin{array}{ccc} & |2\rangle|\varnothing\rangle & |1\rangle|1_p\rangle \\ \hat{H}_{\mathrm{eff}}= & \multicolumn{2}{c}{\begin{pmatrix}\omega_{21} & \bar{\Omega}_n \\ \bar{\Omega}_n^* & \omega_n-i\frac{\gamma_n}{2}\end{pmatrix}}\end{array}$$

B. Effective multi-mode model

- Atom-continuum multiple Lorentzian coupling:





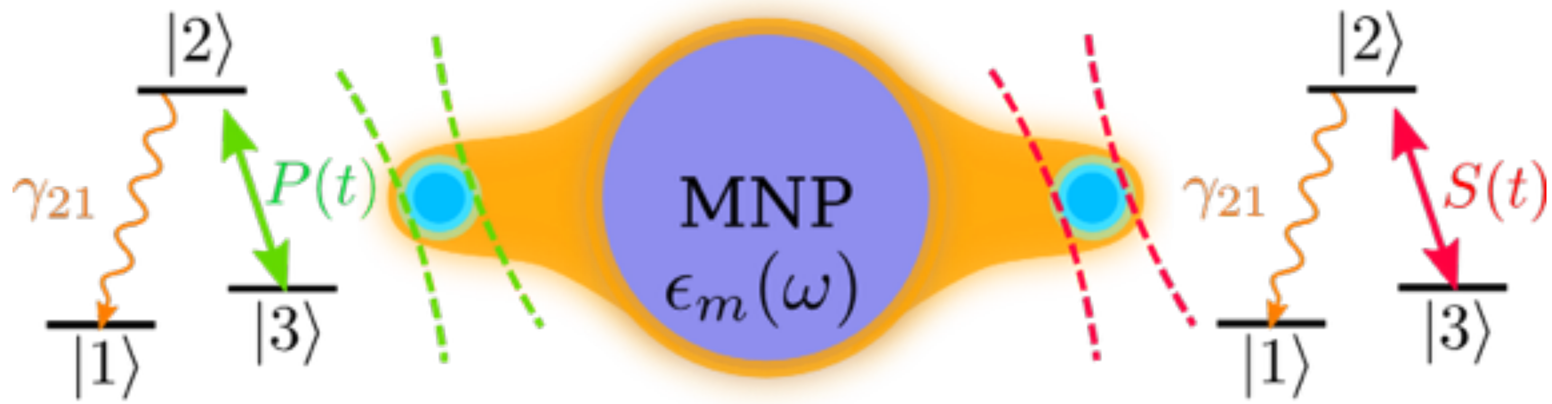
$$|\kappa(\omega)|^2 = \sum_{n=1}^{N} \frac{\gamma_n}{2\pi} \frac{|\bar{\Omega}_n|^2}{(\omega-\omega_n)^2 + \left(\frac{\gamma_n}{2}\right)^2}$$

$$\hat{H}_{\text{eff}} = \begin{array}{c} \begin{array}{ccccc} \color{red}{|2\rangle|\varnothing\rangle} & \color{red}{|1\rangle|1_1\rangle} & \color{red}{|1\rangle|1_2\rangle} & & \color{red}{|1\rangle|1_N\rangle} \end{array} \\ \begin{pmatrix} \omega_{21} & \bar{\Omega}_1 & \bar{\Omega}_2 & \dots & \bar{\Omega}_N \\ \bar{\Omega}_1^* & \omega_1 - i\frac{\gamma_1}{2} & 0 & \dots & 0 \\ \bar{\Omega}_2^* & 0 & \omega_2 - i\frac{\gamma_2}{2} & \ddots & \vdots \\ \vdots & \vdots & \ddots & \ddots & 0 \\ \bar{\Omega}_N^* & 0 & \dots & 0 & \omega_N - i\frac{\gamma_N}{2} \end{pmatrix} \end{array}$$

For more details see the poster of David Dzsotjan

C. Two emitters coupled to a nanosphere

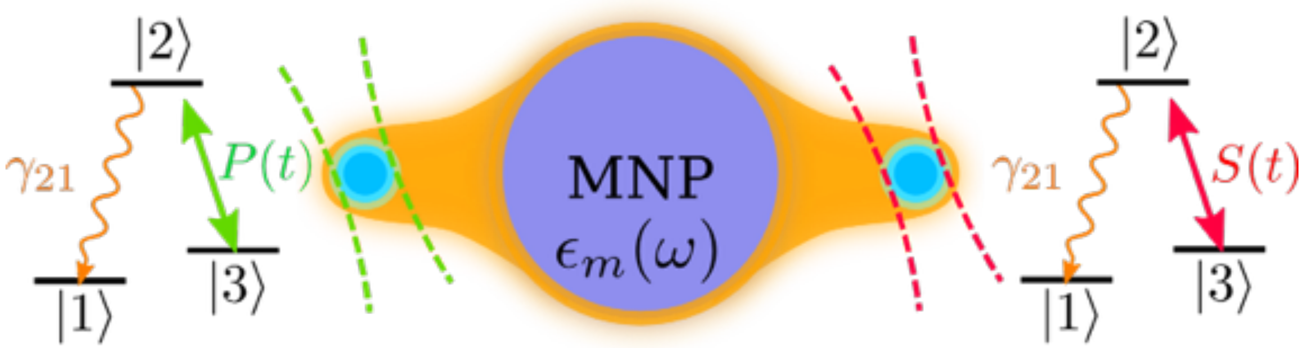
- two 3-level emitters coupled to a sphere:



- both atoms couple equally to the MNP
- $|2\rangle \leftrightarrow |3\rangle$ off-resonant with the plasmon modes and coherently driven by laser pulses, each addressing a single emitter

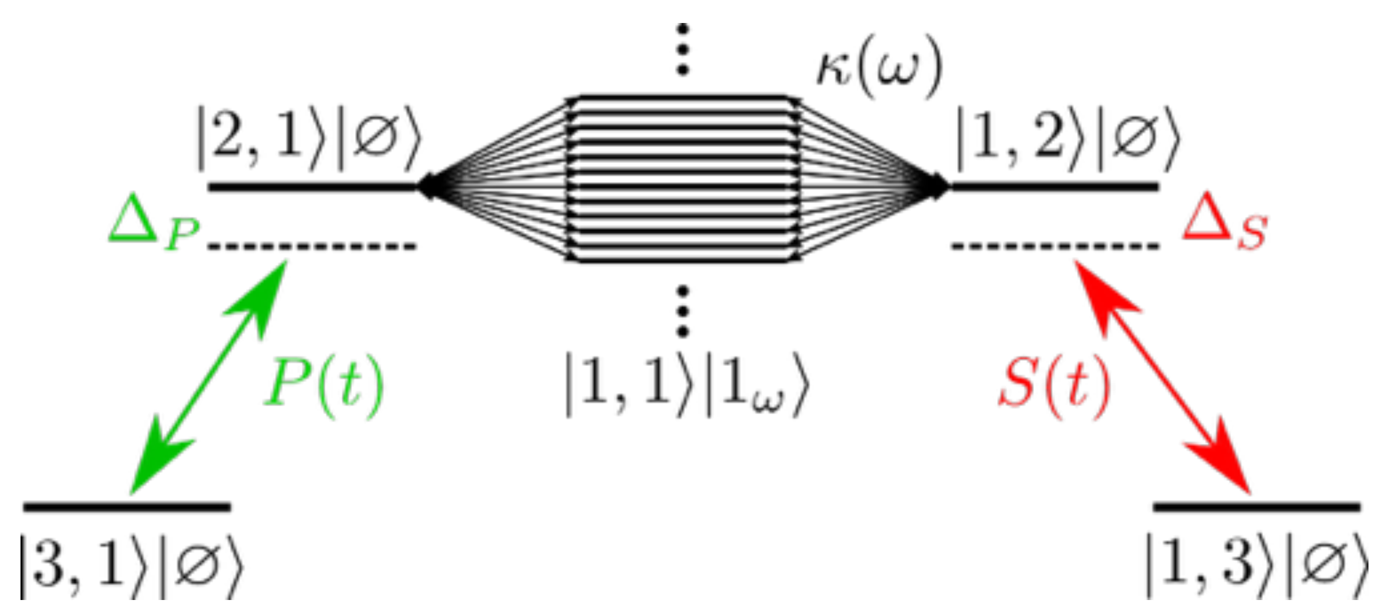
C. Two emitters coupled to a nanosphere

$$\hat{H} = \sum_{\alpha=1}^{2}\left(\hbar\omega_{21}\hat{\sigma}_{22}^{(\alpha)} + \hbar\omega_{23}\hat{\sigma}_{33}^{(\alpha)}\right) + \int \mathrm{d}\vec{r}\int_{0}^{+\infty}\mathrm{d}\omega\,\hbar\omega\,\hat{\vec{f}}^{\dagger}_{\omega}(\vec{r})\,\hat{\vec{f}}_{\omega}(\vec{r})$$
$$-\sum_{\alpha=1}^{2}\hat{\sigma}_{21}^{(\alpha)}\left(\int_{0}^{+\infty}\mathrm{d}\omega\,\vec{d}_{21}^{(\alpha)}\cdot\hat{\vec{E}}(\vec{r}_{\alpha},\omega) + \mathrm{h.c.}\right)$$
$$+\left(\hbar P(t)\mathrm{e}^{-i\omega_L t}\hat{\sigma}_{23}^{(1)} + \mathrm{h.c.}\right)$$
$$+\left(\hbar S(t)\mathrm{e}^{-i\omega_L t}\hat{\sigma}_{23}^{(2)} + \mathrm{h.c.}\right)$$



product basis

$|\mathrm{emitter}\rangle_1 \otimes |\mathrm{emitter}\rangle_2 \otimes |\mathrm{plasmons}\rangle$



C. Two emitters coupled to a nanosphere

- Effective multi-mode discrete Hamiltonian:

$$\hat{H}_{\text{eff}} = \begin{pmatrix} 0 & P & \cdots & & & \cdots & 0 \\ P & \Delta_P & \cdots & \bar{\Omega}_n & \cdots & & \vdots \\ \vdots & \vdots & \ddots & & \mathbf{0} & \vdots & \\ & \bar{\Omega}_n^* & & \Delta_n - i\frac{\gamma_n}{2} & & \bar{\Omega}_n & \\ & \vdots & \mathbf{0} & & \ddots & \vdots & \vdots \\ \vdots & & \cdots & \bar{\Omega}_n^* & \cdots & \Delta_S & S \\ 0 & \cdots & & & \cdots & S & 0 \end{pmatrix}$$

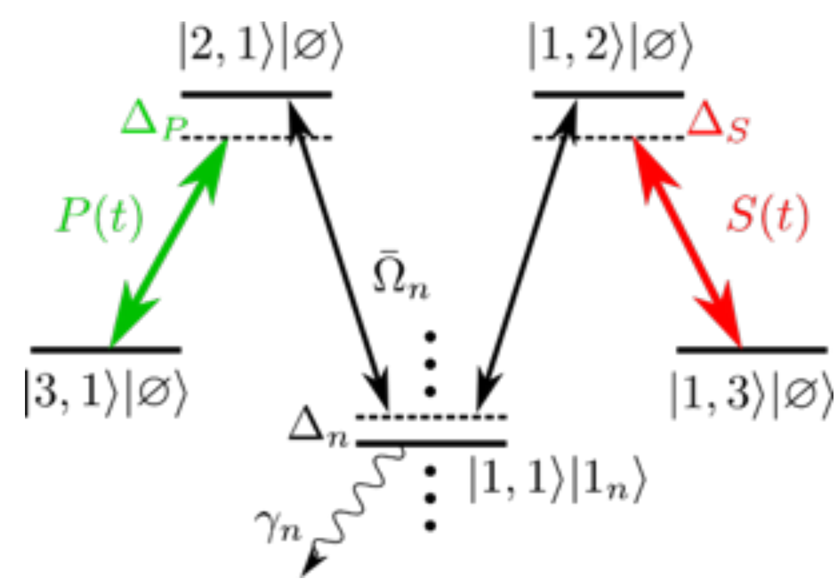
- discrete basis
- modal expansion

<u>Computation in two parts:</u>

Mode structure computation

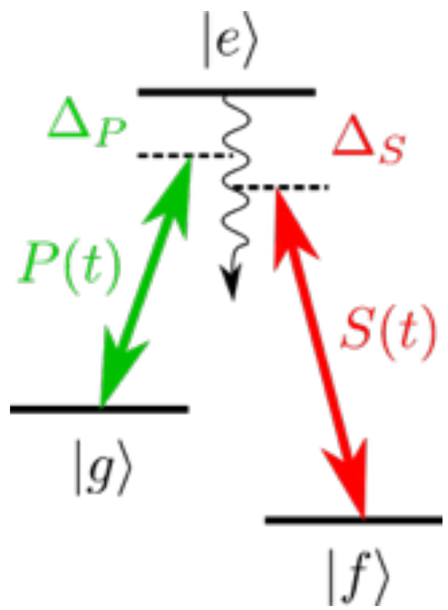
input

Quantum dynamics



D. Plasmon-mediated STIRAP

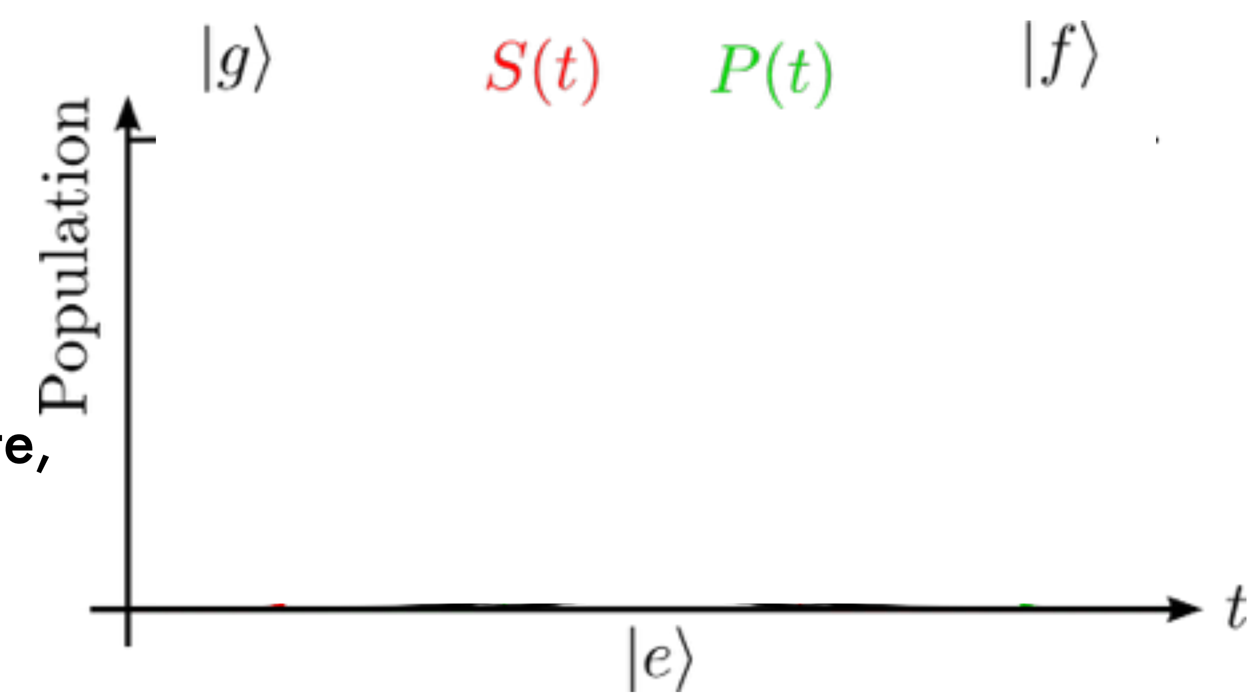
- STImulated Raman Adiabatic Passage



$$\hat{H} = \begin{pmatrix} 0 & P & 0 \\ P & \Delta_P - i\frac{\gamma}{2} & S \\ 0 & S & \Delta_P - \Delta_S \end{pmatrix}$$

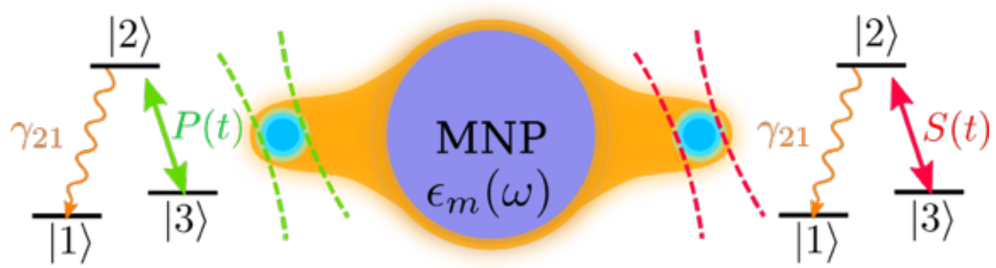
- Population transfer process
- Well-known with 3-level sys.

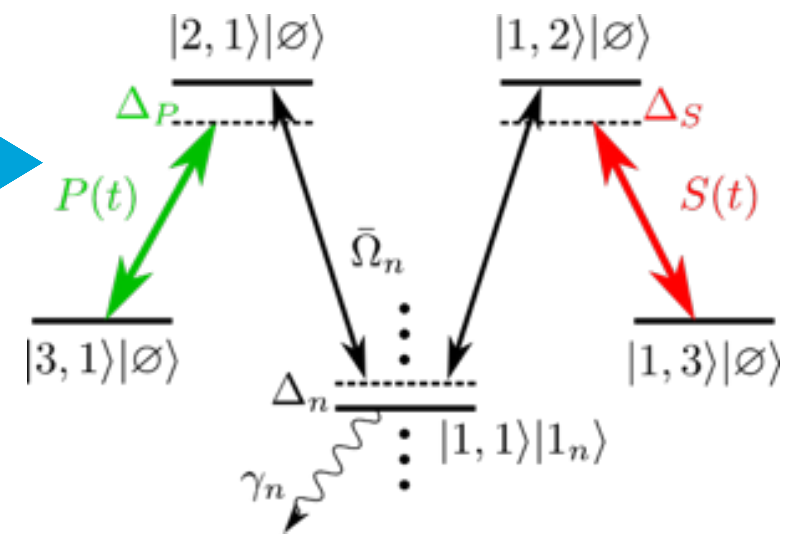
- Counter-intuitive sequence
- Adiabatic pulses
- $\Delta_P = \Delta_S$
- Passage via a non-lossy dark eigenstate, adiabatic limit $P_{\max}T \sim S_{\max}T \gg 1$



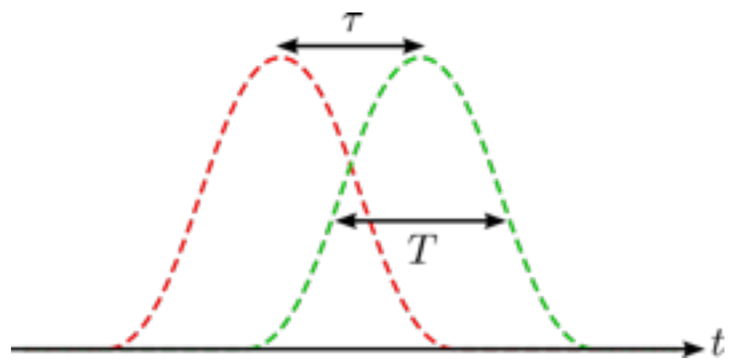
U. Gaubatz, P. Rudecki, S. Schiemann and K. Bergmann, J. Chem. Phys. **92**, 5363 (1990)

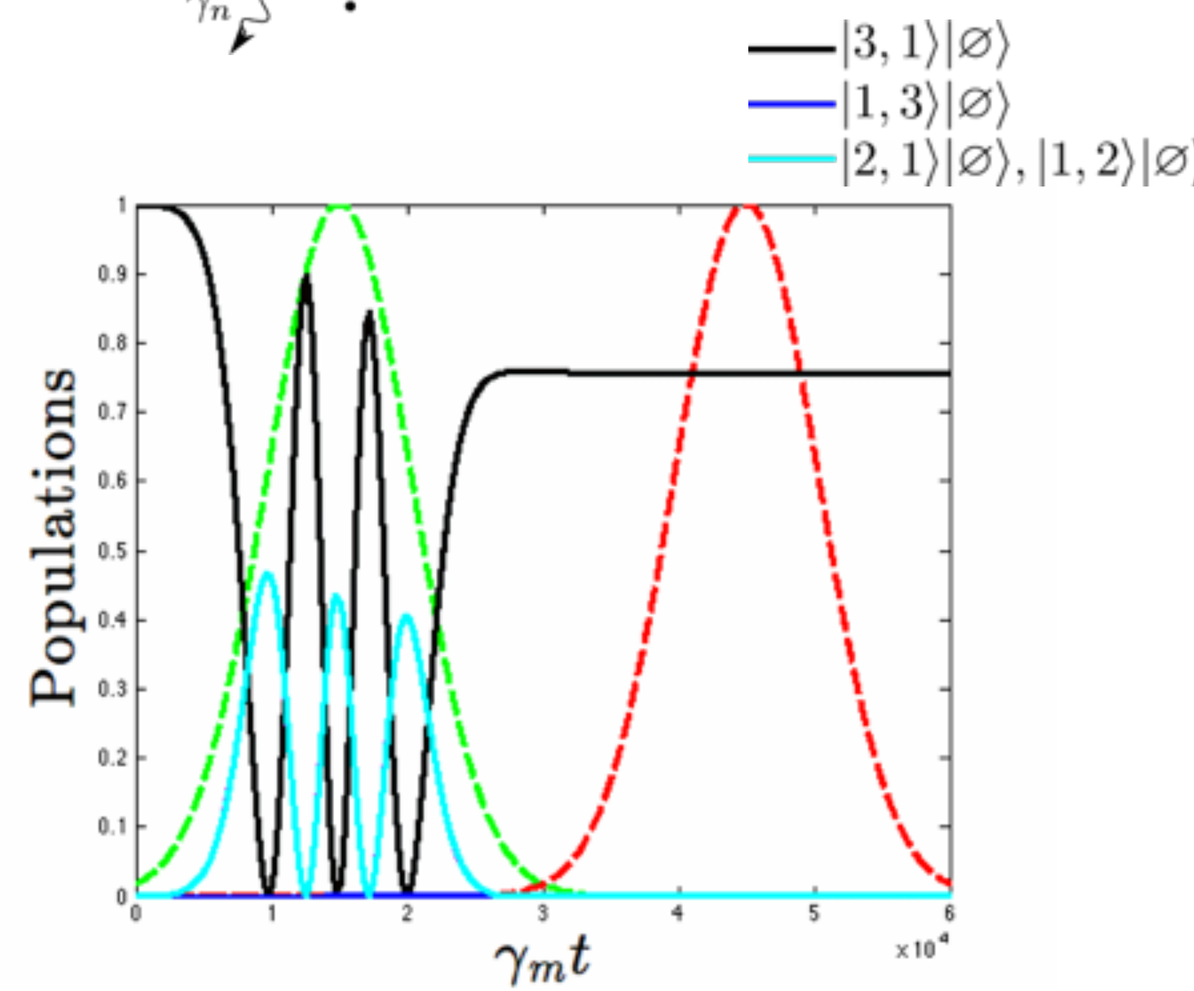
D. Plasmon-mediated STIRAP





$$|\psi_i\rangle = |3,1\rangle|\varnothing\rangle \rightarrow |\psi_f\rangle = |1,3\rangle|\varnothing\rangle$$



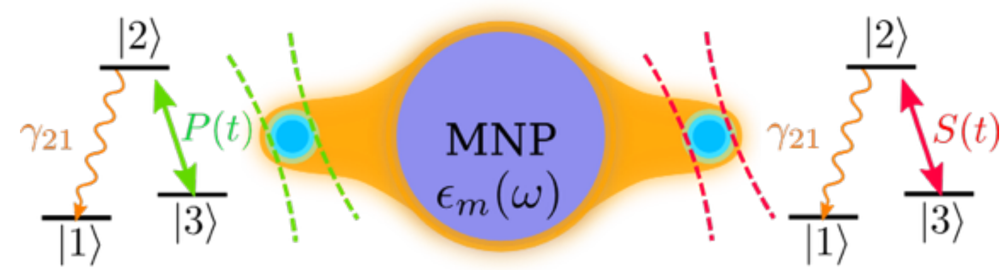


Parameters:

- 10 modes
- silver MNP R = 8nm
- $\omega_{21} \sim \omega_n$
- $d_{21}^{(\alpha)} = 0.5\text{nm} \times e$
- pulse length in the ps-ns regime
- emitters close 2 nm from MNP

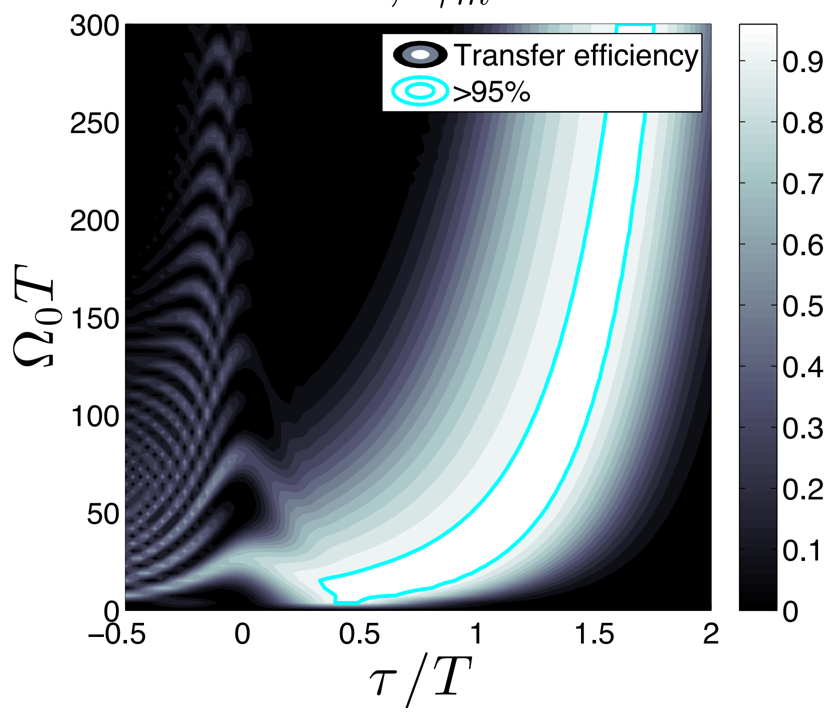
D. Plasmon-mediated STIRAP

Parameters:
- 10 modes (R = 5nm)
- 25 modes (R = 8 or 20 nm)
- $\omega_{21} \sim \omega_n$
- $d_{21}^{(\alpha)} = 0.5\text{nm} \times e$
- pulse length in the ps-ns regime
- emitters close 2 nm from MNP

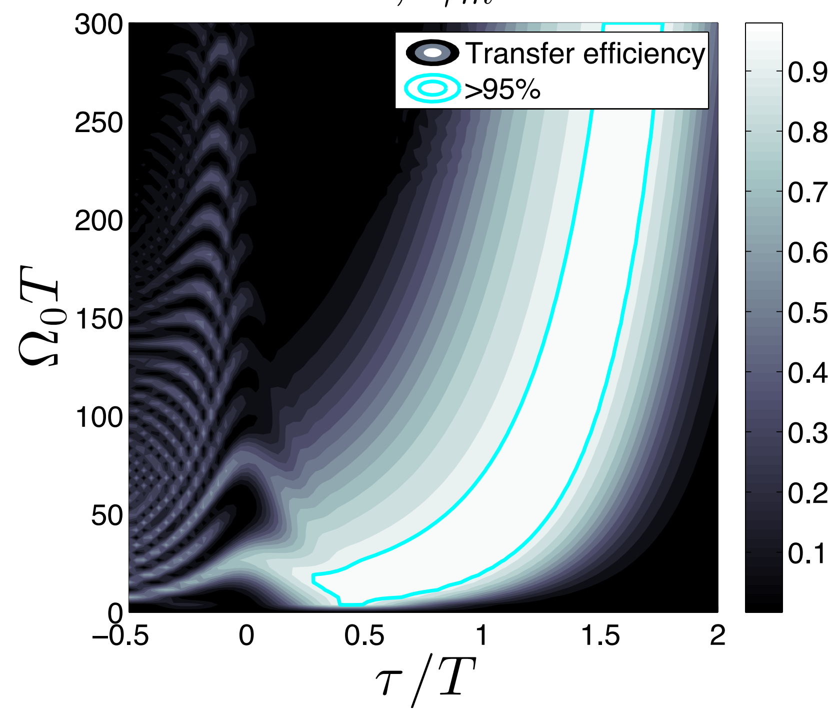


$$|\psi_i\rangle = |3,1\rangle|\varnothing\rangle \rightarrow |\psi_f\rangle = |1,3\rangle|\varnothing\rangle$$

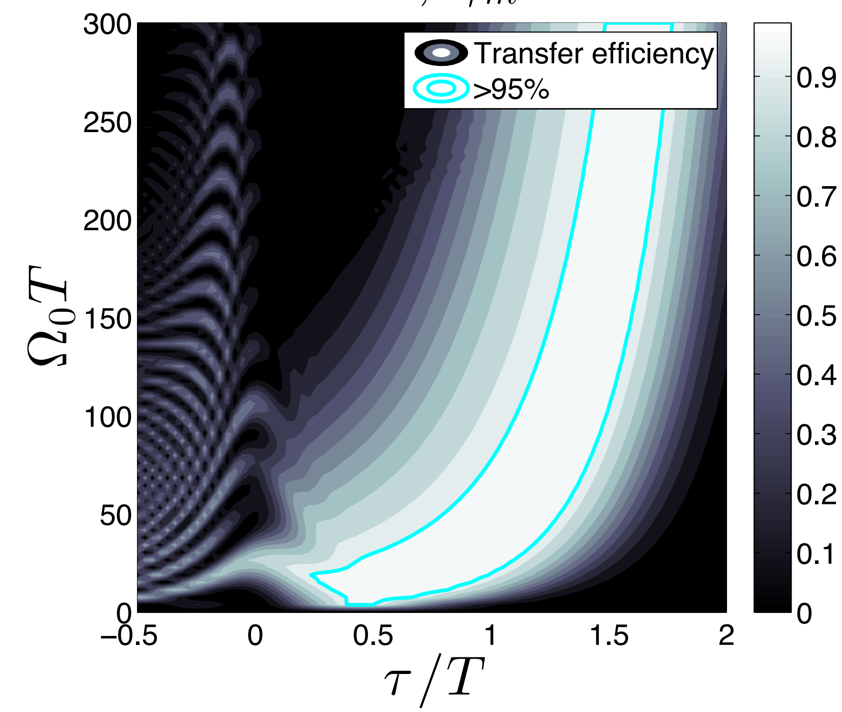
$R = 5$ nm, $\gamma_m T = 7500$



$R = 8$ nm, $\gamma_m T = 7500$

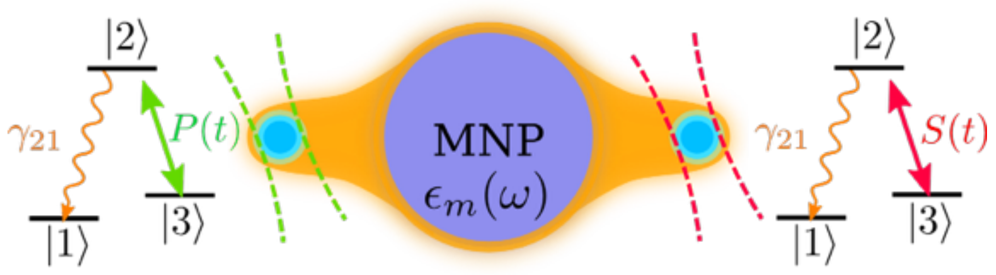


$R = 20$ nm, $\gamma_m T = 7500$



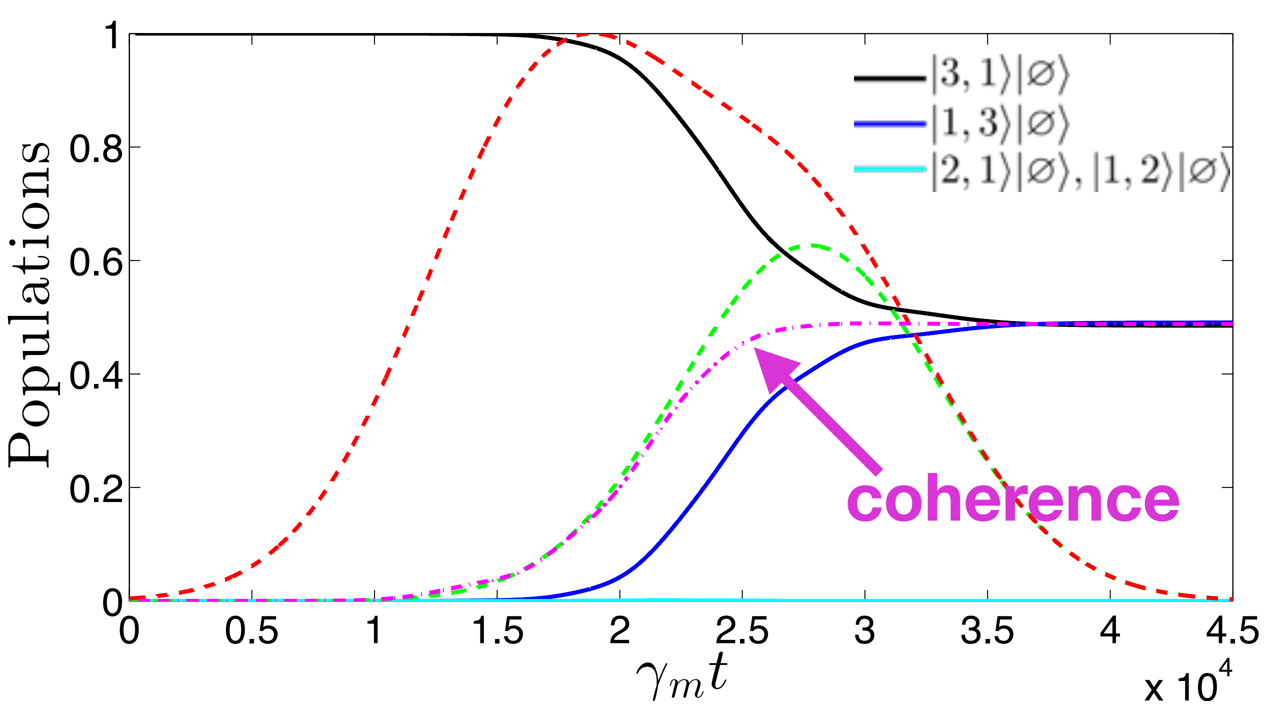
D. Plasmon-mediated STIRAP

- Entanglement using fractional STIRAP:



$$|\psi_i\rangle = |3,1\rangle|\varnothing\rangle \rightarrow |\psi_f\rangle = \frac{1}{\sqrt{2}}\left(|3,1\rangle + \mathrm{e}^{i\phi}|1,3\rangle\right)|\varnothing\rangle$$

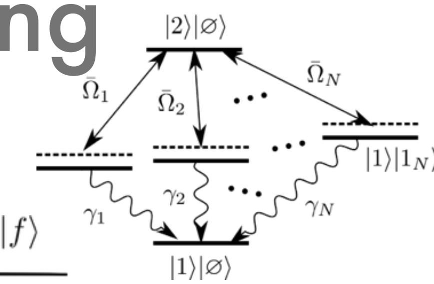
Parameters:
- 10 modes
- silver MNP R = 8nm
- $\omega_{21} \sim \omega_n$
- $d_{21}^{(\alpha)} = 0.5\text{nm} \times e$
- pulse length in the ps-ns regime
- emitters close 2 nm from MNP



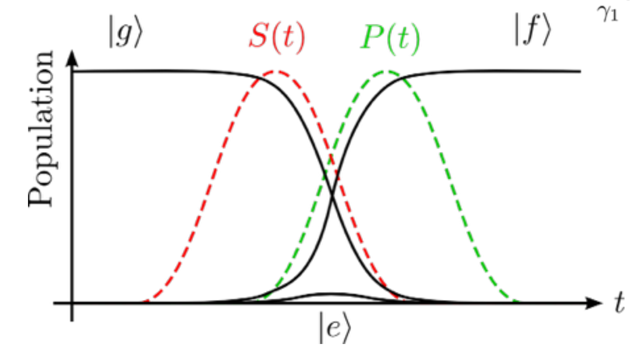
N. V. Vitanov, K.-A. Suominen and B. W. Shore, J. Phys. B **32**, 4535 (1999)
L. P. Yatsenko, S. Guérin and H.-R. Jauslin, Phys. Rev. A **70**, 043402 (2004)

Summary

- Simple effective model for Lorentzian coupling



- Robustness of the STIRAP



- Multi-emitter population transfer
- Multi-emitter entanglement $\frac{1}{\sqrt{2}}\left(|3,1\rangle + \mathrm{e}^{i\phi}|1,3\rangle\right)|\varnothing\rangle$
- Applicable for other geometries: localized (nanoprism, ellipsoid…) & delocalized (nanowire…)

25th birthday of STIRAP !
STIRAP symposium on sept. 22–25, Kaiserslautern, Germany