Dynamical quantum theory of heat transfer between plasmonic nanosystems

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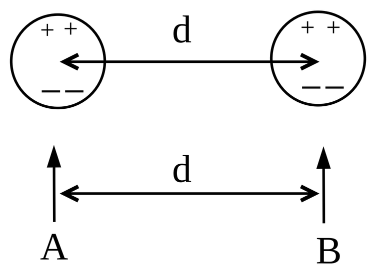
Abstract

We develop a dynamical theory of heat transfer between two nanosystems. In particular, we consider the resonant heat transfer between two nanoparticles due to the coupling of localized surface modes having a finite spectral width. We model the coupled nanosystem by two coupled quantum mechanical oscillators, each interacting with its own heat bath, and obtain a master equation for the dynamics of heat transfer. The damping rates in the master equation are related to the lifetimes of localized plasmons in the nanoparticles. We study the dynamics towards the steady state and establish connection with the standard theory of heat transfer in steady state. For strongly coupled nanoparticles we predict Rabi oscillations in the mean occupation number of surface plasmons in each nanoparticle.

System

- Consider two metallic nanoparticles



- Polarizability and permittivity of the nanoparticles

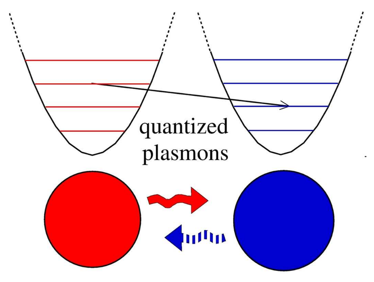
$$\alpha(\omega) = 4\pi r_0^3 \frac{\epsilon(\omega) - 1}{\epsilon(\omega) + 2} \quad \text{and} \quad \epsilon(\omega) = \epsilon_\infty - \frac{\omega_p^2}{\omega(\omega + i\gamma)}.$$

- Localized surface plasmons for ($\omega_p \gg \gamma$):

$$\omega_{sp} = \frac{\omega_p}{\sqrt{\epsilon_\infty + 2}} \quad \text{and} \quad \Gamma = \frac{\gamma}{2}.$$

Modelling the plasmonic nanosystem

- Describing localized surface plasmons as harmonic oscillators



- Hamiltonian of the system

$$H = H_0 + H_I + H_{B1} + H_{B2} + H_{A-B1} + H_{B-B2}.$$

- Hamiltonian for both plasmonic particles a and b

$$H_0 = \hbar\omega_{sp} a^\dagger a + \hbar\omega_{sp} b^\dagger b.$$

- Hamiltonian for the interaction between the particles

$$H_I = \hbar g (b^\dagger a + a^\dagger b).$$

- Hamiltonian of the heat bath oscillators

$$H_{B1/B2} = \sum_j \hbar\omega_{1/2j} a^\dagger_{1/2j} a_{1/2j}.$$

- Hamiltonian for the coupling to the heat baths

$$H_{A-B1} = \hbar i \sum_j g_{1j}(a + a^\dagger)(a_{1j} - a^\dagger_{1j}),$$
$$H_{B-B2} = \hbar i \sum_j g_{2j}(b + b^\dagger)(a_{2j} - a^\dagger_{2j}).$$

- Heat baths are in thermal equilibrium at T_1 and T_2, i.e.

$$\rho_{B1/B2} = \frac{e^{-\beta_{1/2} H_{B1/B2}}}{\mathrm{Tr}\big(e^{-\beta_{1/2} H_{B1/B2}}\big)}.$$

Master Equation

- Born-Markov + rotating wave approximation and tracing out the bath variables using the density operators ρ_{B1} and ρ_{B2}:

$$\frac{\partial \rho_S}{\partial t} = -i\omega_a[a^\dagger a, \rho_S] - i\omega_b[b^\dagger b, \rho_S] - ig[a^\dagger b + b^\dagger a, \rho_S]$$
$$- \kappa_1(\overline{n}_1 + 1)\big(a^\dagger a \rho_S - 2a\rho_S a^\dagger + \rho_S a^\dagger a\big) - \kappa_1 \overline{n}_1 \big(a a^\dagger \rho_S - 2a^\dagger \rho_S a + \rho_S a a^\dagger\big)$$
$$+ (1 \to 2, a \to b).$$

- Introducing the mean occupation numbers

$$\overline{n}_{1/2} = \frac{1}{e^{\hbar\beta_{1/2}\omega_{a/b}} - 1}.$$

- The coupling constants $\kappa_{1/2}$ can be identified as the linewidths Γ of the localized surface modes.

Dynamical Equations

- The dynamical equations follow from the master equation:

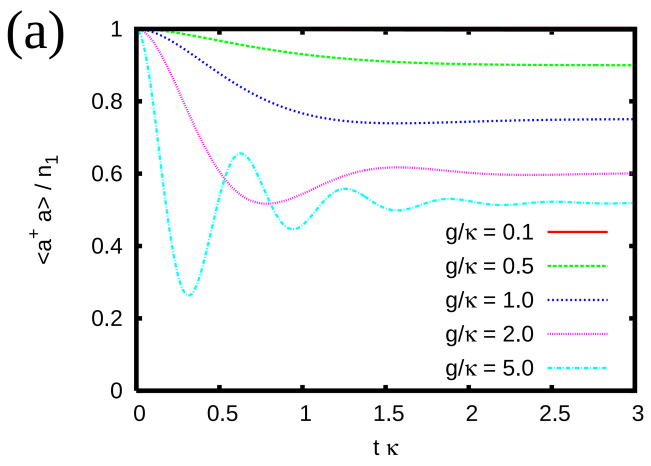
$$\frac{d}{dt}\langle a^\dagger a\rangle = -ig\big(\langle a^\dagger b\rangle - \langle b^\dagger a\rangle\big) - 2\kappa_1\langle a^\dagger a\rangle + 2\kappa_1\overline{n}_1,$$
$$\frac{d}{dt}\langle b^\dagger b\rangle = -ig\big(\langle b^\dagger a\rangle - \langle a^\dagger b\rangle\big) - 2\kappa_2\langle b^\dagger b\rangle + 2\kappa_2\overline{n}_2,$$
$$\frac{d}{dt}\langle b^\dagger a\rangle = \Omega_{ab}\langle b^\dagger a\rangle - ig\big(\langle b^\dagger b\rangle - \langle a^\dagger a\rangle\big),$$
$$\frac{d}{dt}\langle a^\dagger b\rangle = \Omega_{ba}\langle a^\dagger b\rangle - ig\big(\langle a^\dagger a\rangle - \langle b^\dagger b\rangle\big).$$

- Introducing the new quantities:

$$\Omega_{ab} = -i(\omega_a - \omega_b) - \kappa_1 - \kappa_2 \quad \text{and} \quad \Omega_{ba} = +i(\omega_a - \omega_b) - \kappa_1 - \kappa_2.$$

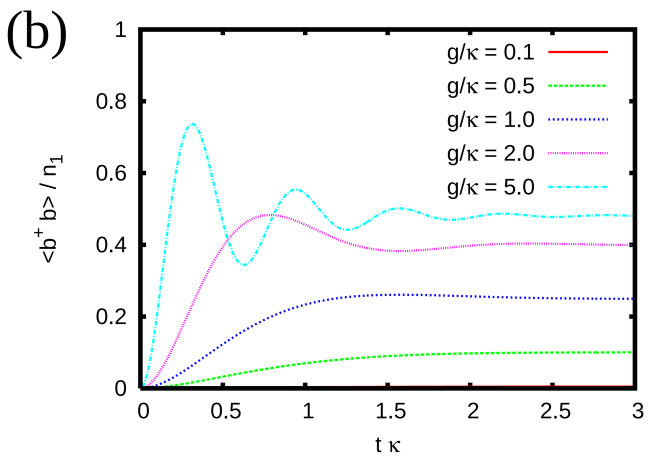
- Numerical results ($\kappa_1 = \kappa_2 \equiv \kappa$, $\omega_a = \omega_b$, and $\overline{n}_2 = 0$):

(a)



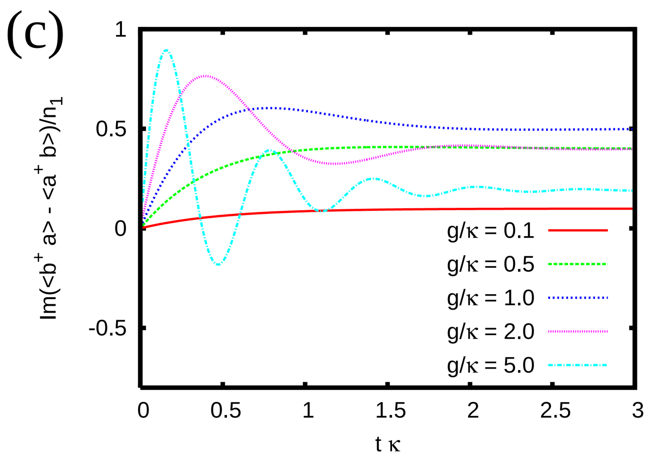
$\langle a^\dagger a\rangle/\overline{n}_1$

(b)

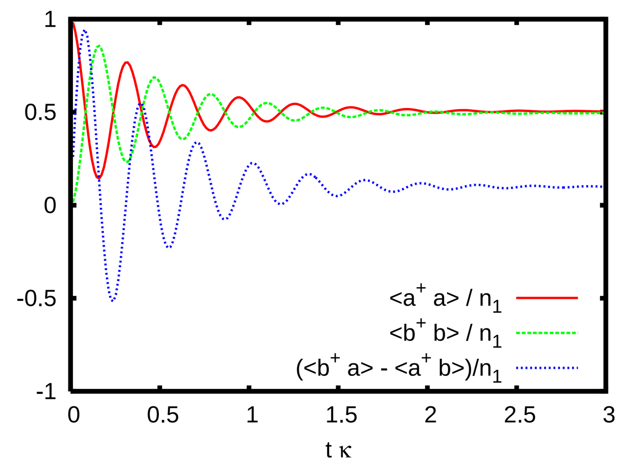


$\langle b^\dagger b\rangle/\overline{n}_1$

(c)



$\mathrm{Im}(\langle b^\dagger a\rangle - \langle a^\dagger b\rangle)/\overline{n}_1$



$\mathrm{Im}(\langle b^\dagger a\rangle - \langle a^\dagger b\rangle)/\overline{n}_1$, $\langle a^\dagger a\rangle/\overline{n}_1$, and $\langle b^\dagger b\rangle/\overline{n}_1$ for $g/\kappa = 10$

Heat transfer rate

- The mean transfered power from oscillator a to oscillator b is

$$\langle P\rangle = \hbar\omega_{sp}(-ig)\big[\langle ab^\dagger\rangle - \langle a^\dagger b\rangle\big] \equiv \hbar\omega_{sp} R.$$

- From the steady-state result of fluctuational electrodynamics [1]

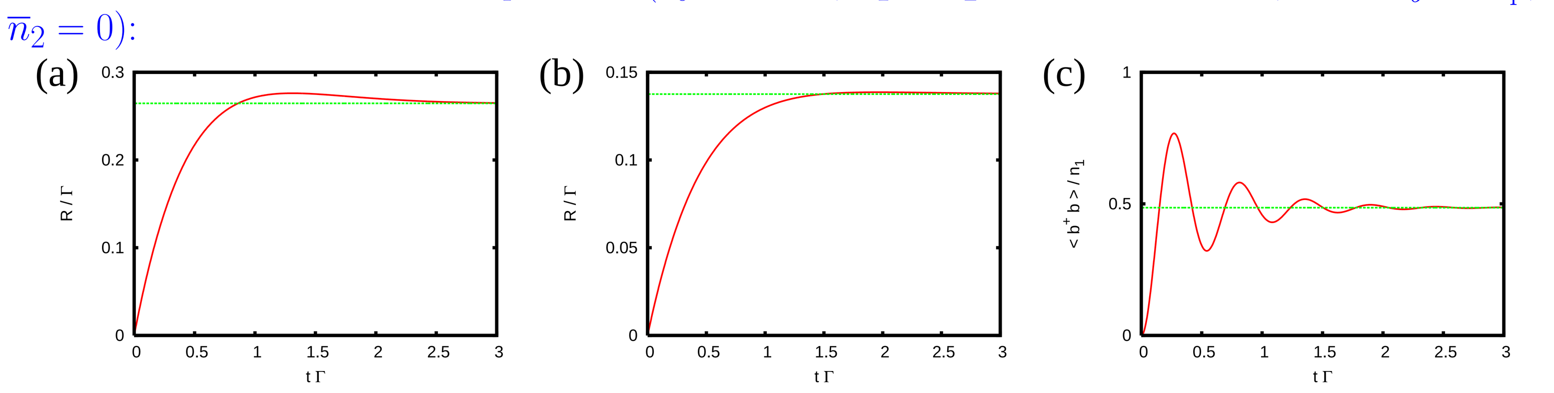
$$P = \int_0^\infty \frac{d\omega}{4\pi^3}\hbar\omega\overline{n}_1 \mathrm{Im}(\alpha)^2 \frac{\omega^6}{c^6}\left[\frac{3}{(\frac{\omega}{c}d)^6} + \frac{1}{(\frac{\omega}{c}d)^4} + \frac{1}{(\frac{\omega}{c}d)^2}\right]$$

$$\Rightarrow R_{st-st} = \overline{n}_1 \frac{\omega_{sp}^2 r_0^6}{\Gamma \; 2} F(\omega_{sp}) \left(\frac{3}{\epsilon_\infty + 2}\right)^2 \quad \text{with} \quad F(\omega) = \frac{\omega^6}{c^6}\left[\frac{3}{(\frac{\omega}{c}d)^6} + \frac{1}{(\frac{\omega}{c}d)^4} + \frac{1}{(\frac{\omega}{c}d)^2}\right].$$

- Comparison with quantum mechanical steady-state expression [2] gives ($g^2 \ll \kappa_1\kappa_2$ and $\kappa_1 = \kappa_2 = \Gamma$)

$$R = \frac{g^2}{\Gamma}\overline{n}_1 \Rightarrow g = \omega_{sp}\frac{r_0^3}{\sqrt{2}}\sqrt{F(\omega_{sp})}\frac{3}{\epsilon_\infty + 2}.$$

- Numerical results for two silver particles ($r_0 = 20\,\mathrm{nm}$, $\kappa_1 = \kappa_2 \equiv \Gamma = 1.4 \cdot 10^{13} s^1$, $\omega_a = \omega_b = \omega_{sp}$, and $\overline{n}_2 = 0$):



(a) $d = 800\,\mathrm{nm}$, i.e. $g/\Gamma = 0.6$ (b) $d = 1200\,\mathrm{nm}$, i.e. $g/\Gamma = 0.4$ (c) $d = 100\,\mathrm{nm}$, i.e. $g/\Gamma = 5.8$

1. [1] P.-O. Chapuis, M. Laroche, S. Volz, and J.-J. Greffet, Appl. Phys. Lett. **92**, 201906 (2008).
2. [2] S.-A. Biehs and G. S. Agarwal, J. Opt. Soc. Am. B **30**, 700 (2013).