Role of nonlocality and Landau damping in the dynamics of a Quantum dot coupled to surface plasmons.

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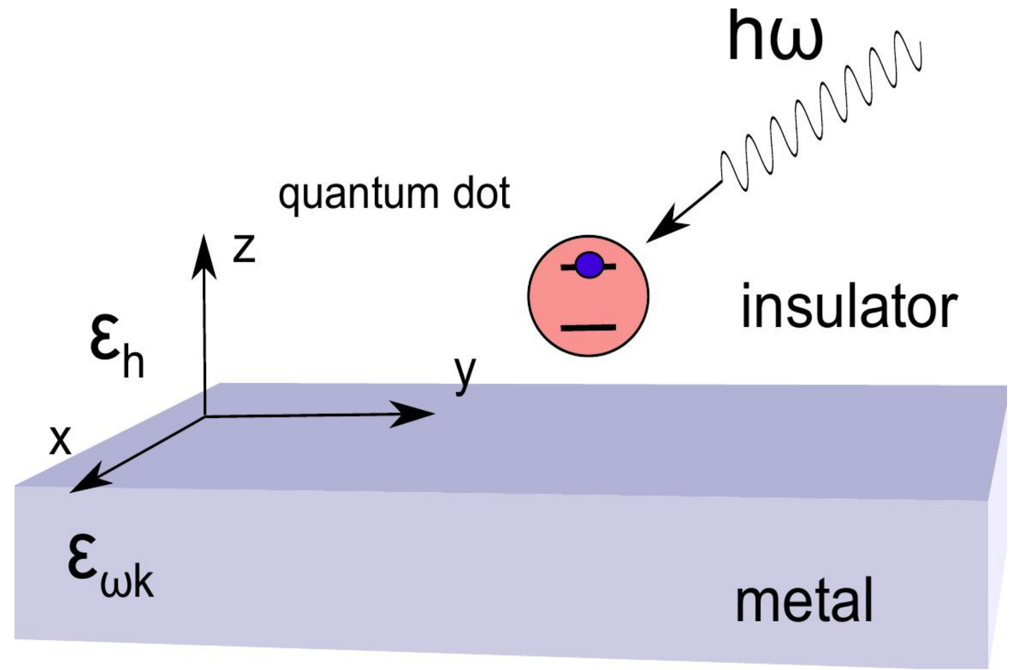


Metal slab (thick), and **quantum dot** (quantum scatterer with well separate levels). Exciton in the dot is created by external laser pulse.

Non-local electromagnetic response

Delocalized carriers in the metal → spatially non-local response

Bulk dielectric susceptibility →

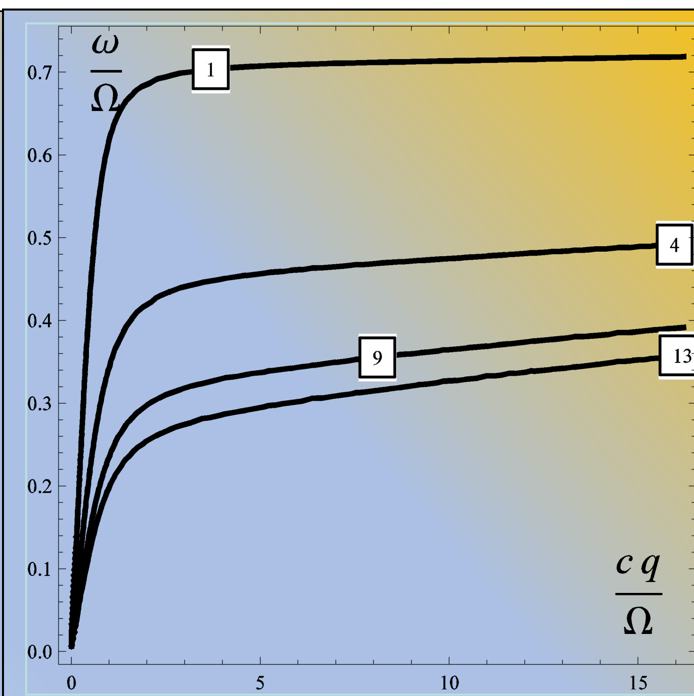
Lindhard function

$$\varepsilon(\omega,k)=1+\frac{3\Omega_p^2}{(kv_F)^2}[1-\frac{\omega}{2kv_F}\ln\frac{\omega+kv_F}{\omega-kv_F}]$$

$$\varepsilon(\omega,k)=1-\frac{\Omega_p^2}{\omega^2}\,,\qquad \mathrm{Im}\,\varepsilon=\frac{3\pi\Omega_p^2\omega}{2(kv_F)^3}\sim\frac{\Omega_p^2\omega r_D^3}{v_F{}^3}$$

$k\to 0$ $\qquad$ $k v_F>\omega$

N. W. Ashcroft and N. D. Mermin, *Solid State Physics*, (Thomson, Toronto, 1976)



Dispersion of the Surface Plasmon at the boundary of metal with dielectric media. Numbers in the boxes are dielectric constants of the host media.

Plasmon's quantization:

In general, plasmon has arbitry amlitude, for quantization we need to make it such that total energy of SP W=ħω.

Hamiltonian of the system

$$H=\hbar\left[\frac{1}{2}\Omega_{ex}(\sigma_z+1)+\sum_{\vec{k}}\omega_k b_k^+ b_k\right]+\hbar\sum_{\vec{k}} g_k\sigma_- b_k^+ + g_k^*\sigma_+ b_k$$

Ω_{ex} is the frequency of the exciton transition, ω_k is the frequency of the surface plasmon, and g_k are the coupling constants.

Quantum state $|\Psi\rangle = a(t)|\Psi_e\rangle+\sum c_k(t)|\Psi_k\rangle$

Dynamic equations $i\partial_t a(t)=\Omega_{ex}a(t)+\sum g_k c_k(t),$ $\quad i\partial_t c_k(t)=\omega_k c_k(t)+g_k^* a(t)$

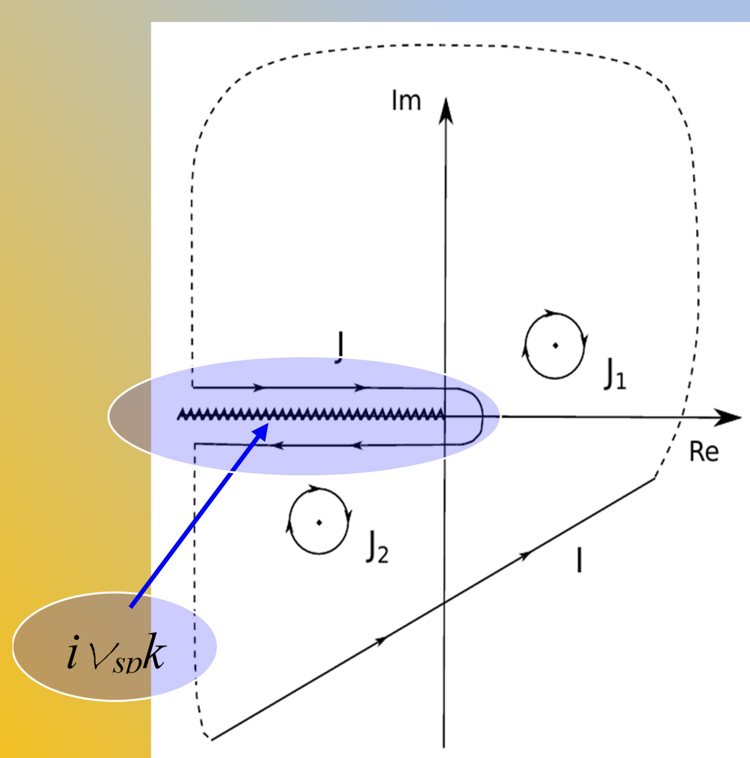
$$a(s)=\frac{1}{s+i\Omega_{ex}+K(s)}$$ Laplas transform solution

$$K(s)=\sum_k\frac{|g_k|^2}{s+i\omega_k}\approx\int_0^\infty\frac{k^2e^{-2kd}dk}{s+i\Omega_{sp}+\Gamma+iv_{sp}k}$$

- Incomplet gamma function

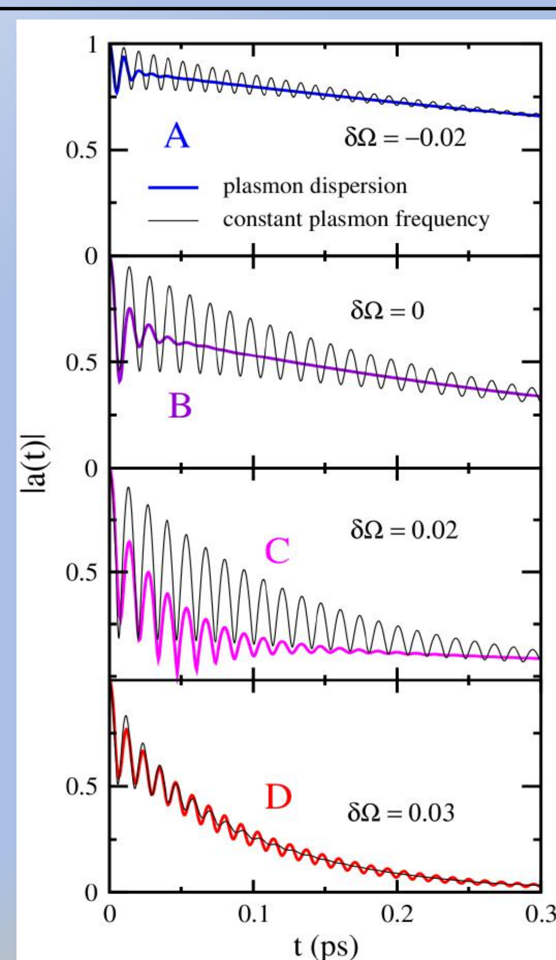
Linear spectrum generates brunch cut -- without linear spectrum only poles exist, no branch-cut

Inverse Laplace transform contour:



$$|a(t)|=\left|a_1e^{-i\Omega_R t-\gamma_1 t}+a_2e^{-\gamma_2 t}\right| \text{ or}$$

$$|a(t)|=\left|a_1e^{-i\Omega_R t-\gamma_1 t}+a_2t^{-3}\right|$$



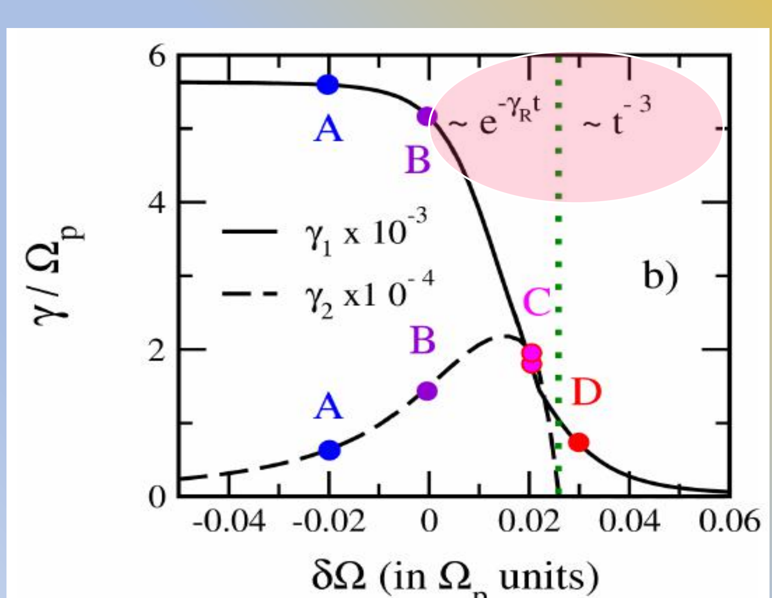
Two type relaxations

1) fast relaxation of the Rabi oscillations ~0.1ps

2) slow monotonous relaxation ~100 ps

Power law relaxation

When the branch-cut contribution is important (single pole), the time asymptote may change from exponential to a power law. Consequence: much longer Rabi oscillations



Summary

Non-locality in the electro-magnetic response:

1. is important in systems of finite size
2. changes the frequency dispersion (spectrum) of surface plasmons
3. for a flat surface leads to the two-velocity frequency dispersion (velocities are very different)
4. modifies the damping rate of surface plasmons (Landau damping)
5. leads to notable changes in the dynamics of a nearby quantum system (two time relaxation pattern, change in the long-time asymptotic, etc.)

References

1. Larkin, I.A., Stockman, M.I., Achermann, M., Klimov, V.I.; Dipolar emitters at nanoscale proximity of metal surfaces: Giant enhancement of relaxation in microscopic theory Phys. Rev. B 69, 121403(R), (2004).
2. A. Vagov, I. A. Larkin, M. D. Croitoru, V. M. Axt “Role of nonlocality and Landau damping in the dynamics of a quantum dot coupled to surface plasmons”; Phys. Rev. B **93**, 195414, (2016)
3. I. A. Larkin, K. Keil, A. Vagov, M. D. Croitoru, and V. M. Axt Superanomalous Skin Effect for Surface Plasmon Polaritons Phys. Rev. Lett. **119**, 176801 (2017)

Outline

1. Theory with the non-local electro-magnetic response
   a) Lindhard response
   b) finite size corrections
2. Surface plasmon modes
   a) electromagnetic problem
   b) frequency dispersion and non-locality
   c) Landau damping
3. Dynamics of a quantum dot induced by surface plasmons
   a) effective quantum Hamiltonian
   b) dynamical patterns & non-locality

Theoretical description for dot-metal coupling

There are three theory components – dot, material carriers, el.-m. fields

1. Dot – quantum system with few (two) levels

2. Charge carriers in materials – material equations, for example Boltzmann kinetic equation

3. Maxwell equations for the fields

2. + 3. Maxwell equations for the field in the matter

surface plasmons – the modes localized near the metal-insulator surface

Non-local electromagnetic response

Delocalized carriers in the metal → spatially non-local response

Bulk dielectric susceptibility → Lindhard function

$$\varepsilon(\omega,k)=1+\frac{3\Omega_p^2}{(kv_F)^2}\left[1-\frac{\omega}{2kv_F}\ln\frac{\omega+kv_F}{\omega-kv_F}\right]$$

$$\varepsilon(\omega,k)=1-\frac{\Omega_p^2}{\omega^2}\,,\quad k\to 0 \qquad\qquad \operatorname{Im}\varepsilon=\frac{3\pi\Omega_p^2\omega}{2(kv_F)^3}\sim\frac{\Omega_p^2\omega r_D^3}{v_F^{\;3}}\,,\quad k\,v_F>\omega$$

$$i\mathbf{k}\cdot\mathbf{P}_{\rm k}=en_k\,;\quad \delta f(\mathbf{k})=\frac{\mathbf{E}(\mathbf{k})\cdot\nabla_{\mathbf{p}}f_0}{\mathbf{k}\cdot\mathbf{v}-\omega+i/\tau}$$

$$4\pi\,\mathbf{P}(\mathbf{k})=(\varepsilon-1)\cdot\mathbf{E}(\mathbf{k})$$

$$(\varepsilon_{k,\omega}-1)=\frac{4\pi e^2}{k^2}\oiiint_{\mathbf{p}}\frac{\mathbf{k}\cdot\nabla_{\mathbf{p}}f_0(\mathbf{p})d^3p}{\mathbf{k}\cdot\mathbf{v}-\omega+i/\tau}$$

Plasmon's quantization: In general, plasmon has arbitrary amplitude, for quantization we need to make it such that total energy of SP $\hbar\omega_k$

$$\hbar\omega_k=\iiint_V w\,dV;\quad w=\frac{1}{16\pi}\left(\frac{\partial}{\partial\omega}\big(\omega\varepsilon(\omega)\big)E^2+H^2\right)$$

Hamiltonian of the system: dot + SP

$$H=\hbar\left[\frac{1}{2}\Omega_{ex}(\sigma_z+1)+\sum_{\vec{k}}\omega_k b_k^+ b_k\right]+\hbar\sum_{\vec{k}}g_k\sigma_- b_k^+ + g_k^*\sigma_+ b_k$$

Where Ω_{ex} is the frequency of the exciton transition, ω_q is the frequency of the surface plasmon with the wave vector q, c^+ and c are the creation and annihilation operators of electron in the quantum dot in conduction band, d^+ and d are creation and annihilation operators of the electron in valence band, and b_q^+ and b_q are creation and annihilation operators of the SP. H_I is the interaction Hamiltonian of the two-level dipole with SP and g_q are the coupling constants. To solve Schrödinger equation, we apply Laplace transform:

Quantum state $\left|\Psi\right\rangle = a(t)\left|\Psi_e\right\rangle + \sum c_k(t)\left|\Psi_k\right\rangle$

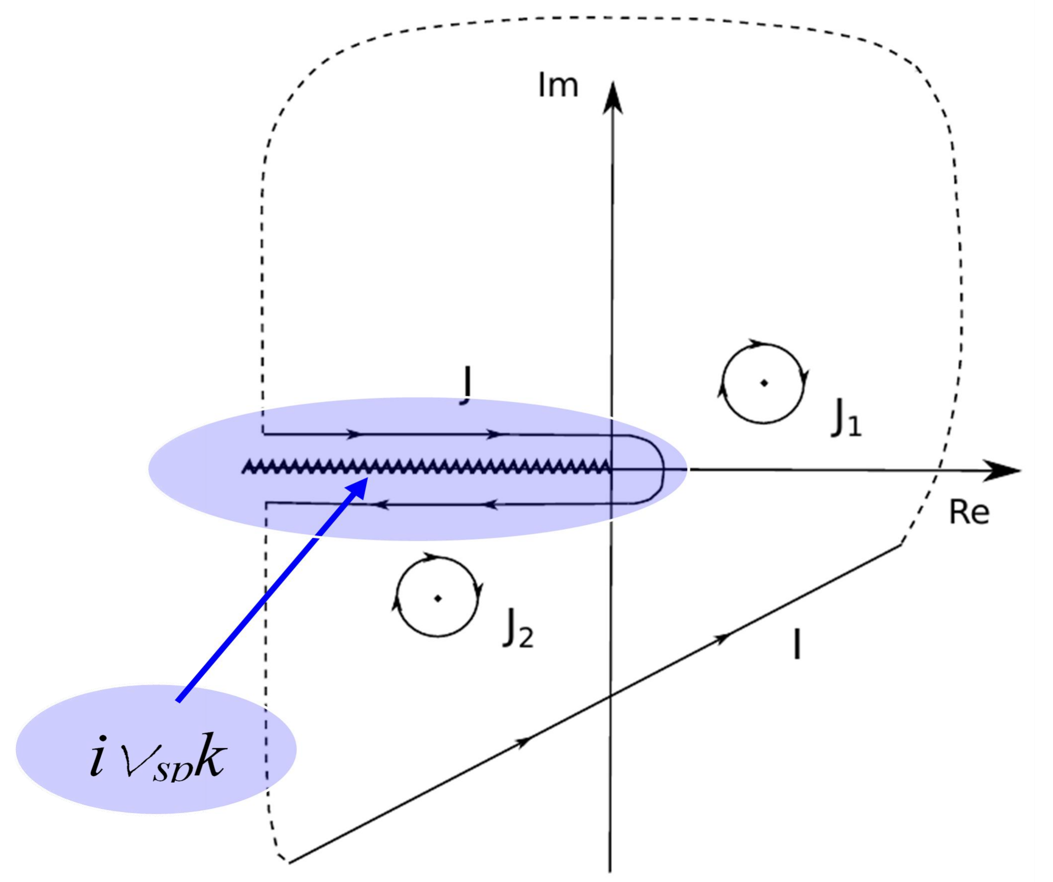
Dynamic equations $i\partial_t a(t) = \Omega_{ex} a(t) + \sum g_k c_k(t),$ $i\partial_t c_k(t) = \omega_k c_k(t) + g_k^* a(t)$

$$a(s) = \frac{1}{s + i\Omega_{ex} + K(s)}$$ Laplace transform solution

$$K(s) = \sum_k \frac{|g_k|^2}{s + i\omega_k} \approx \int_0^\infty \frac{g^2 k^2 e^{-2kd} dk}{s + i\Omega_{sp} + \Gamma + iv_{sp}k}$$

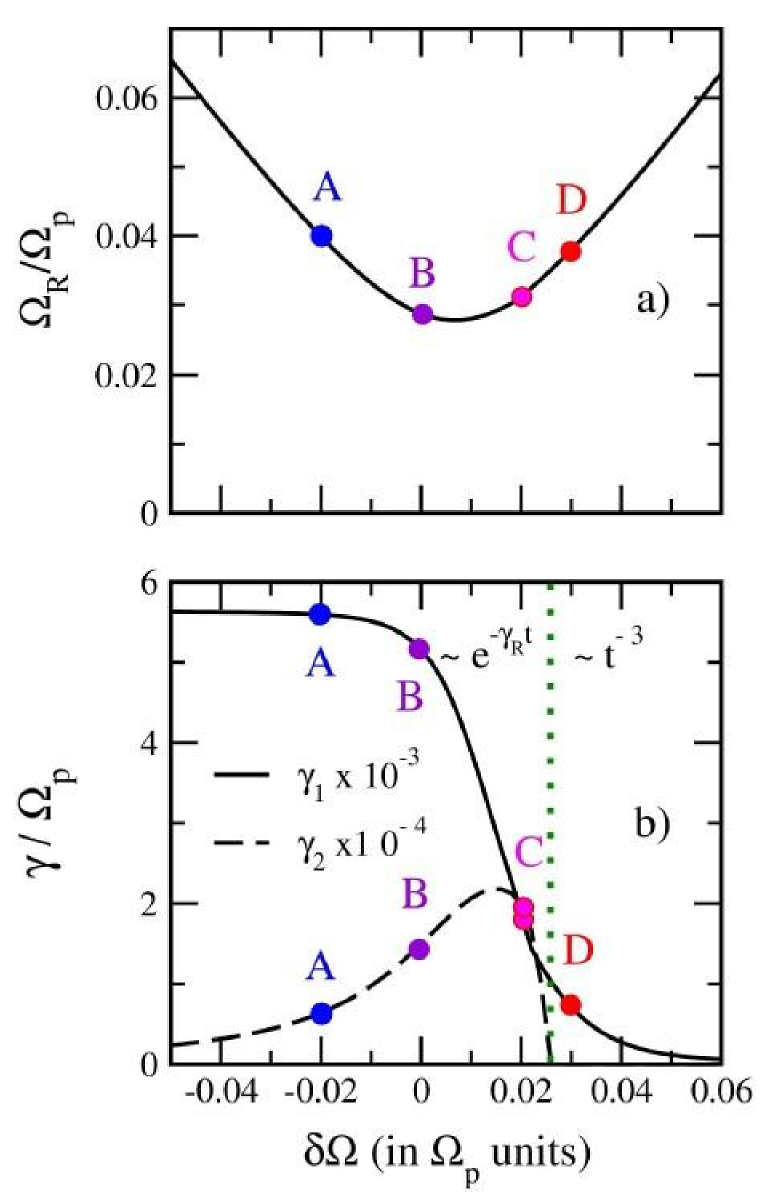
$K(s)$ - Incomplet gamma function

With the cut from – infinity to zero.

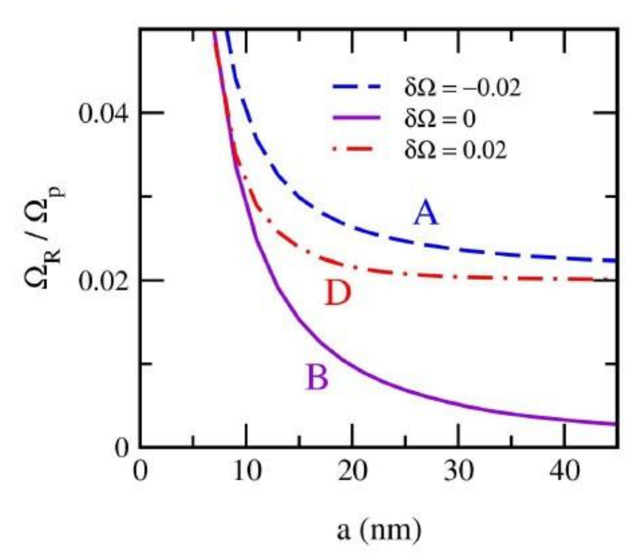


Inverse Laplace transform contour

Linear spectrum generates brunch cut: without linear spectrum only poles exist, no branch-cut. As a result $|a(t)| = \left|a_1 e^{-i\Omega_R t - \gamma_1 t} + a_2 e^{-\gamma_2 t}\right|$ when contribution from two poles dominates, or $|a(t)| = \left|a_1 e^{-i\Omega_R t - \gamma_1 t} + a_2 t^{-3}\right|$. First term is the Rabi oscillations that have fast relaxation ~ 0.1ps, whereas at $\Omega_{ex} > \Omega_{sp} + g^2$ the branch-cut contribution is important (single pole), the time asymptote may change from exponential to a power law. At this regime system has much longer Rabi oscillations.

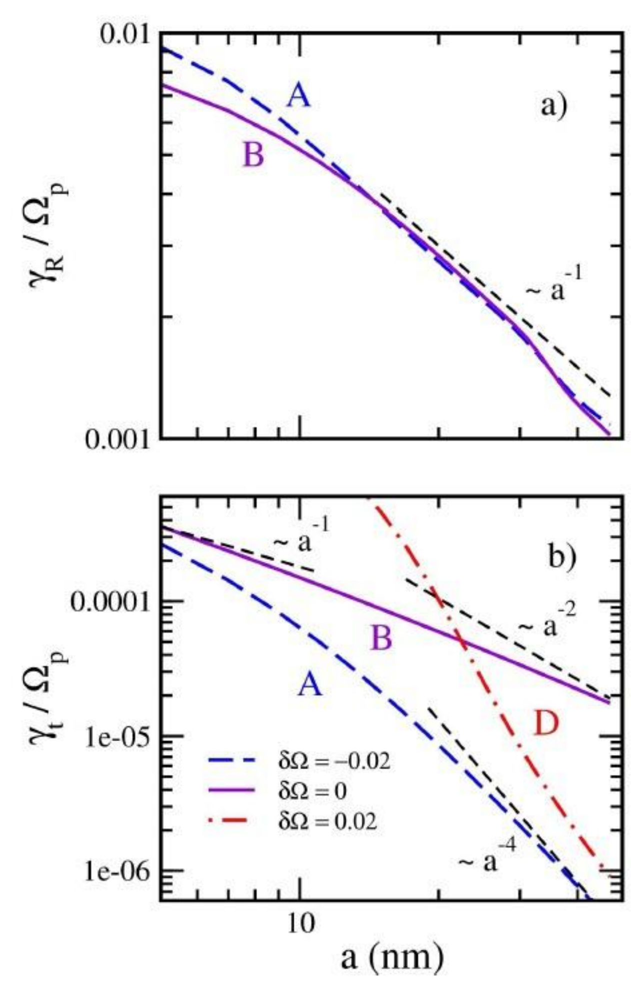


Dependence of the characteristics of the system dynamics on the detuning $\delta\Omega = \Omega_{ex} - \Omega_{sp}$.
Panel a): frequency of the Rabi oscillations Ω_R.
Panel b): two decay rates, $\gamma_{1,2}$.
The QD-surface distance is a = 10nm.



Frequency of the Rabi oscillations Ω_R as a function of the QD to surface distance a = 10nm.

A, B, D correspond to $\delta\Omega = -0.02, 0, 0.02$ (in Ω_p units).



Log-log plots of the decay rates γ_R (panel a) and γ_t (panel b) as functions of the QD-surface distance a calculated at $\delta\Omega = -0.02, 0, 0.02$ (in Ω_p units).
The case $\delta\Omega = 0.02$ (D) is absent for γ_R in panel a) because the corresponding contribution has a power law asymptotic.
Panel a) reveals a more complicated dependence $\gamma_R(a)$. At small a the additional contribution disappears and one obtains $\gamma_R \sim a^{-1}$. However, the numerical curves in panel a) are not close to this asymptotes, showing instead a more complicated pattern.

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