Cooperative emission by quantum plasmonic superradiance

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We investigate the correlated spontaneous emission from an assembly of emitters coupled to a plasmonic particle, in a fully quantized model. We prove the existence of cooperative emission, referred to as plasmonic superradiance, for specific emitters positions. Plasmonic superradiance dynamics originates from the strong correlation building up between the dipolar emitters and presents a fast burst of emission. We analyse the impact of the emitter positions and the role of localized surface plasmons on the superradiance. We show that the cooperative behaviour can be inhibited by destructive interferences at small distances. Plasmonic superradiance sources constitutes ultrafast and extremely bright optical nanosources of strong interest for integrated quantum nano-optics platforms.

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Introduction. In a seminal work, Dicke discovered that a large number $N_c$ of atoms radiate collectively when they occupy a subwavelength volume and that their emission is much faster ($\tau_N = \tau_1/N_c$) and stronger ($I_N = N_c^2 I_1$) than for independent atoms. This so-called superradiance originates from spontaneous phase-locking of the atomic dipoles through a same free-space electromagnetic mode and is very similar to the building of cooperative emission in a laser amplifier. Superradiant emission produces original states of light with properties such as superradiant lasers presenting high spectral purity or single photon superradiance for quantum memory. Recently, Putsovits and Shahbazyan identified a new mechanism for cooperative emission by plasmonics Dicke states. They investigated an ensemble of dipoles coupled to a metal nanoparticle particle (MNP), considering a classical approach, which however cannot describe the construction of the cooperative behaviour, a quantum characteristic of superradiance. In this Letter, taking benefit from recent theoretical advances on quantum plasmonics, we derive a quantum approach for plasmonic superradiance and discuss the dynamics of cooperative emission. We highlight the analogy and the differences with superradiance in a cloud of atoms.

Master equation. In the Markov and rotating wave approximations, the dynamics of $N_c$ emitters coupled to a MNP is governed by the effective Hamiltonian

\begin{equation}
\hat{H}_{\text{eff}} = \sum_{n=1}^{N_c} \frac{\hbar n}{2} \left( \delta_n - \frac{\gamma_n}{2} \right) \hat{n}_n + \sum_{n=1}^{N_c} \sum_{i=1}^{N_{\text{ind}}} \left( g_n^{(i)} \hat{a}_+^{(i)} \hat{n}_n^{(i)} - g_n^{(i)} \hat{n}_n^{(i)} \hat{a}_-^{(i)} \right),
\end{equation}

with the detuning $\delta_n = \omega_n - \omega_0$ between the plasmon and emitter angular frequencies $\omega_n$ and $\omega_0$, respectively. In this expression, $\hat{a}_+^{(i)}$ and $\hat{a}_-^{(i)}$ are the transition operators associated to the emitter $i$. Bosonic operators $\hat{n}_n^{(i)}$ and $\hat{a}_+^{(i)}$ describe the creation or annihilation of a localized surface plasmon of order $n$ (LSP$_n$) associated to emitter $i$. $g_n^{(i)}$ refers to the coupling strength between emitter $i$ and mode $\hat{n}_n^{(i)}$.

In the dissipative regime, adiabatic elimination follows $\partial_t \hat{n}_n^{(i)}(t) \approx 0$. It leads to the Lindblad equation

\begin{equation}
\frac{d\hat{\rho}(t)}{dt} = \mathcal{L}[\hat{\rho}(t)] = \sum_{n=1}^{N_c} \sum_{j=1}^{N_{\text{ind}}} \frac{1}{\hbar} \left[ \hat{H}_{jk}, \hat{\rho}(t) \right] + \mathcal{D}_{jk}[\hat{\rho}(t)],
\end{equation}

with

\begin{align}
\hat{H}_{jk} & = -\hbar \Delta_{jk} \hat{a}_+^{(k)} \hat{a}_-^{(j)}, \quad (3a) \\
\mathcal{D}_{jk}[\hat{\rho}(t)] & = \Gamma_{jk} \left( \hat{a}_+^{(k)} \hat{a}_-^{(j)} \hat{\rho}(t) + \hat{\rho}(t) \hat{a}_+^{(k)} \hat{a}_-^{(j)} \right) - \frac{1}{2} \left( \hat{a}_+^{(k)} \hat{a}_-^{(j)} \hat{\rho}(t) + \hat{\rho}(t) \hat{a}_+^{(k)} \hat{a}_-^{(j)} \right).
\end{align}

The parameter $\Gamma_j = \Gamma_{jj}$ ($\Delta_j = \Delta_{jj}$) represents the decay rate (Lamb shift) of the emitter $j$ in presence of the MNP. For $j \neq k$, $\Gamma_{jk}$ and $\Delta_{jk}$ characterize the cooperative decay rate and population transfer

\begin{align}
\Gamma_{jk} & = \sum_{n=1}^{N_c} \frac{\gamma_n}{\delta_n^2 + (\frac{2\hbar}{\gamma_n})^2} g_n^{(j)} g_n^{(k)} \mu_n^{(jk)}, \quad (4a) \\
\Delta_{jk} & = \sum_{n=1}^{N_c} \frac{\gamma_n}{\delta_n^2 + (\frac{2\hbar}{\gamma_n})^2} g_n^{(j)} g_n^{(k)} \mu_n^{(jk)}, \quad (4b)
\end{align}

where we introduced the coupling strength $g_n^{(j)}$ between the emitter $j$ and the mode LSP$_n$. $\mu_n^{(jk)}$ refers to the coupling strength between emitters $i$ and $j$ via LSP$_n$. This plays an important role in the emitters dynamics since depending on its sign it could lead to either enhancement or blockade of the cooperative process as we will discuss later. One can write equivalently $\Gamma_{jk} = 2 \omega_0^2 / (\hbar c 0^2) \mathcal{G}^{(jk)} \cdot \mathbf{m} \left( \mathbf{G} (r_j, r_k, \omega_0) \right)$ where $\mathbf{G}$ is the Green tensor in presence of the MNP (see [15]).
Cooperative emission. Dicke states exist for identical emitters equivalent coupled to an electromagnetic modes so that only a few configurations presenting high symmetry are concerned (see §4 in ref. [1]). Superradiance is degraded if the emitters are located at different points, but still occurs. In the case of plasmonic Dicke states, the coupling to LSPs strongly depends on the emitter orientation and the order of the LSPs modes engaged in the coupling process, which make the understanding of its dynamics more involved. We work in the emitter basis
\[
\{ |ee\ldots ee\rangle, \mathcal{P}_{N_e-1}^N(|\alpha\rangle), \ldots, \mathcal{P}_{N_e}^N(|\alpha\rangle), |gg\ldots gg\rangle \},
\]
where the permutator \( \mathcal{P}_{N_e-1}^N(|\alpha\rangle) \) gives all the states \(|\alpha\rangle\) with \( N_e - l \) excited emitters and \( l \) emitters in their ground state. For instance, \( \mathcal{P}_2^2(|\alpha\rangle) = \{|ee\alpha\rangle, |ege\rangle, |gee\rangle\} \). The radiated emission can be written \( I(t) = \eta \hbar \omega_0 W_{N_e} \) where \( \eta = \gamma_1/\sum \gamma_n \) is the apparent quantum yield and the collective decay rate is defined as
\[
W_{N_e}(t) = \sum_{i=1}^{N_e} \sum_{j=1}^{N_e} \Gamma_{ij} \delta_{ij} \delta_{ij}(t).
\]
This expression generalizes the standard definition to the case of non equal rates \( \Gamma_{ij} \). It can be separated in two contributions: \( W_P(t) \) involving the populations and \( W_C(t) \) involving coherences. For instance, in presence of three emitters, \( W_3(t) = W_P(t) + W_C(t) \) with
\[
W_P(t) = (\Gamma_1 + \Gamma_2 + \Gamma_3) \langle eee|\hat{\rho}(t)|eee\rangle + (\Gamma_1 + \Gamma_2) \langle eeg|\hat{\rho}(t)|eeg\rangle + (\Gamma_1 + \Gamma_3) \langle ege|\hat{\rho}(t)|ege\rangle + (\Gamma_2 + \Gamma_3) \langle geg|\hat{\rho}(t)|geg\rangle + \Gamma_1 \langle eeg|\hat{\rho}(t)|eeg\rangle + \Gamma_2 \langle geg|\hat{\rho}(t)|geg\rangle + \Gamma_3 \langle geg|\hat{\rho}(t)|gge\rangle,
\]
\[
W_C(t) = \Gamma_{12} \langle gee|\hat{\rho}(t)|gee\rangle + \langle gge|\hat{\rho}(t)|gge\rangle + \Gamma_{13} \langle gee|\hat{\rho}(t)|eeg\rangle + \langle gge|\hat{\rho}(t)|eeg\rangle + \Gamma_{23} \langle gee|\hat{\rho}(t)|eeg\rangle + \langle gge|\hat{\rho}(t)|eeg\rangle + \langle gee|\hat{\rho}(t)|gge\rangle + \langle gge|\hat{\rho}(t)|gge\rangle.
\]
Similar expressions can be derived for an arbitrary number of emitters. For independent emitters \( \mu_{i\neq j} = 0 \), the decay rates \( \Gamma_{ij} \) cancel for \( i \neq j \). The emission rate reduces to incoherent emission of the independent emitters \( W_P \). On the contrary, the second term \( W_C \) describes the collective behaviour of the ensemble of emitters when \( \mu_{ij} \neq 0 \). Therefore, the cooperative behaviour originates from the correlation between the states of same excitation \( \mathcal{P}_{N_e-1}^N(|\alpha\rangle) \) in full analogy with free-space superradiance.

The Lindblad equation [18] is numerically solved following ref. [17, 18]. Figure 1(a) represents the dynamics of correlated emission for 6 radial emitters coupled to a silver nanoparticle and its comparison with two limits cases: ideal superradiance and incoherent emission. Ideal superradiance is obtained when all the emitters are located at the same position. We observe a short burst of emission, characteristics of superradiance collective emission. On the opposite, independent emitters emission assumes \( \Gamma_{ij} = 0 \) \( (\forall i \neq j) \) and gives the exponential decay of spontaneous emission with a decay rate given by the Fermi golden rule in presence of the MNP. In case of homogeneous reparation of the emitters around the MNP, we still observe the burst of emission but less pronounced. Close to ideal superradiance behaviour is recovered for emitters located at the two poles of the MNP since the
LSP symmetry leads to identical coupling of the emitters, for this specific configuration. At short distances ($h = 2$ nm, Fig. 4), the cooperative process is not anymore observed and incoherent emission occurs. In case of azimuthal orientation and for $h = 20$ nm, all the emitters equally couple to LSP$_1$ so that close to ideal superradiance is observed (Fig. 3). We analyze in details these behaviours in the following.

**Plasmonic Dicke states.** When all the emitters are located at the same position, $\Gamma_{ij} = \Gamma_{ii} = \Gamma_1$, $\Delta_{ij} = \Delta_{ii} = \Delta_1$ so that we can work with the Dicke ladder ($J = N_e/2$, $M = -J, \ldots, +J$)

$$
|J, M\rangle = \sqrt{\frac{(J + M)!}{N_e!(J - M)!}} |ee \ldots e\rangle
$$

where we have introduced the collective spin operator $J_k = \sum_{i=1}^{N_e} \sigma_i^{(k)}$. $|J, J\rangle$ is the state with all the emitters in their excited state and the symmetrized Dicke state $|J, M\rangle$ is a superposition of the states with $J + M$ excited emitters. The master equation (2) simplifies to

$$
\frac{d\rho_M(t)}{dt} = \Gamma_1 \left[ J (J + 1) - M (M + 1) \right] \rho_{M+1}(t) - \Gamma_1 \left[ J (J + 1) - M (M - 1) \right] \rho_M(t), \tag{9}
$$

where $\rho_M(t) = \langle J, M|\rho(t)|J, M\rangle$ is the population of the state $|J, M\rangle$. This expression is the exact analogue of free-space superradiance except that the decay rate is replaced by its value in presence of the MNP. Finally, the plasmonic superradiance originates from the cascade along the Dicke states ladder. Beginning with the initial condition $|\psi(t = 0)\rangle = |ee \ldots e\rangle = |J, J\rangle$, the system successively goes through the Dicke states $|J, M\rangle$ with $M = J - 1, J - 2, \ldots$, down to the final ground state $|J, -J\rangle = |gg \ldots g\rangle$ for $t \to \infty$. The collective decay rate of state $|J, M\rangle$ is proportional to the number of emitters in their ground state $(J - M + 1)$ and to the number of emitters in their excited ground state $(J + M + 1)$

$$
\Gamma_M = (J + M)(J - M + 1)\Gamma_1. \tag{10}
$$

Similarly to free-space superradiance, the decay rate of the Dicke states starts from $\Gamma_M = N_e\Gamma_1$ for $M = J$, strongly increases up to $\Gamma_M \approx N_e^2\Gamma_1$ for $M = 0$ or $M = \pm 1/2$ (depending on the parity of $J$) and then decreases down to $\Gamma_M = 0$ for the final ground state $(M = -J)$. The build-up of this cooperative behaviour is shown in Fig. 4 (blue curves). It is in close analogy with free-space superradiance. It is worth noticing that since the direct dipole-dipole coupling is negligible compared to LSP mediated dipole-dipole coupling, we avoid the van der Waals dephasing of the symmetric superradiant states observed for free space configurations.

**Superradiant state** Even when the emitters differently couple to LSPs modes, it is possible to define a bright state $|B\rangle$ with one excitation

$$
|B\rangle = \sqrt{\frac{1}{N_e}} \left[ \sqrt{\Gamma_1} |ge \ldots gg\rangle + s_2 \sqrt{\Gamma_2} |ge \ldots gg\rangle + \ldots + s_N \sqrt{\Gamma_N} |gg \ldots ge\rangle \right], \tag{11}
$$

with $\Gamma_T = \sum_{i=1}^{N_e} \Gamma_i$ and the sign $s_j = \Gamma_{ij}/|\Gamma_{ij}|$. The decay rate of this state is

$$
\Gamma_{|B\rangle} = \frac{1}{\Gamma_T} \sum_{i=1}^{N_e} \sum_{j=1}^{N_e} \Gamma_{ij} \sqrt{\Gamma_i \Gamma_j}. \tag{12}
$$

Its maximal value occurs for highly correlated emitters, corresponding to $|\Gamma_{ij}| = \sqrt{\Gamma_i \Gamma_j}$. Then $\Gamma_{|B\rangle} = \Gamma_T$. Moreover, if all the emitters are identically coupled to the MNP, $|B\rangle$ corresponds to the Dicke state $|J, -J+1\rangle$ and we recover $\Gamma_{|B\rangle} = \Gamma_T = N_e \Gamma_1$. For the ring azimuthal distribution discussed in Fig 4, we obtain $\Gamma_{|B\rangle} = 5.9\Gamma_1$ for $h = 20$ nm, close to the ideal situation but $\Gamma_{|B\rangle} = 4.2\Gamma_1$ for $h = 2$ nm. At short distances, the high order LSPs jeopardize the cooperative behaviour as discussed in the following.

**Role of LSPs.** The role of LSPs on the superradiance emission and collective dynamics is first discussed considering two emitters to highlight the analogy and differences with free-space superradiance. Let us consider identical emitters located at the same distance to the MNP (but not necessarily at the same location). We work in the Dicke basis $|ee\rangle, |S\rangle, |A\rangle, |gg\rangle$ with the symmetric and antisymmetric states

$$
|S\rangle = \frac{1}{\sqrt{2}} (|ge\rangle + |eg\rangle), \quad |A\rangle = \frac{1}{\sqrt{2}} (|ge\rangle - |eg\rangle). \tag{13}
$$

Their populations dynamics follows

$$
\begin{align}
\partial_t \rho_{ee}(t) &= -2\Gamma_1 \rho_{ee}(t), \\
\partial_t \rho_{SA}(t) &= \Gamma_{S,AP} \rho_{ee}(t) - \Gamma_{S,AP} \rho_{SA}(t), \\
\partial_t \rho_{gg}(t) &= \Gamma_{S,AP} \rho_{SS}(t) + \Gamma_{A,PS}(t),
\end{align} \tag{14a-c}
$$

as schemed on Fig. 5. The population of the excited state $|ee\rangle$ decays exponentially with the rate $\Gamma_{ee} = 2\Gamma_1$ that does not depend on the relative position (angle) between the two emitters. The symmetric and antisymmetric states are populated from $|ee\rangle$ with the rates...
\[ \Gamma_{S,A} = (\Gamma_1 \pm \Gamma_{12}) \] and relax towards the ground state with the same rates. Finally, superradiant emission follows \[ I(t) = \eta W_2(t) \] with
\[ W_2(t) = 2\Gamma_1\rho_{ee}(t) + \Gamma_S\rho_S(t) + \Gamma_A\rho_A(t). \] (15)

The cooperative relaxation strongly depends on the collective rate \( \Gamma_{12} \), so on the emitters positions. The figure 3a) shows the collective rate \( \Gamma_{12} \) as a function of the angular separation \( \theta \) for two separation distances \( h = 2 \text{ nm} \) or \( h = 20 \text{ nm} \). b, c) Normalized intensity \( I(t)/\eta /\omega_0 \). (b) \( h = 20 \text{ nm} \) (\( \omega_0 = 2.789 \text{ eV} \)) and (c) \( h = 2 \text{ nm} \) (\( \omega_0 = 2.936 \text{ eV} \)). In (b,c), blue curves corresponds to ideal superradiance with the emitters located at the same position, red curves present the uncorrelated emission, black curves refer to the emitter located at the pole \( \theta = \pi \).

\[ |A\rangle = 1/\sqrt{2} (|ge\rangle - |eg\rangle) \] that is not populated and presents a null decay rate \( \Gamma_A = 0 \). For emitter located at the poles of the MNP \( \theta = \pi \) the collective behaviour depends on the distance to the particle. For large separation distances, only the dipolar LSP\(_1\) modes contributes to the emitters-MNP coupling and \( \Gamma_{12}(\pi) \approx -\Gamma_1 \). Superradiant and subradiant states are exchanged (\( |A\rangle \) and \( |S\rangle \), respectively), but the collective dynamics (black curve, Fig. 3b) is close to the ideal case. At smaller distances, high order LSPs modes are also involved in the coupling process and \( \Gamma_{12} \approx 0 \) so that the dynamics of the system closely follows a fully incoherent process (Fig. 3b).

\[
\begin{pmatrix}
-\frac{1}{2}+\Delta
\end{pmatrix} + \sum_{j=1}^{N_c} G(r_{ij}, r_{ij}, \omega) \cdot \vec{p}_j = 0. \tag{16}
\]

The eigenvectors are represented on table I for two emitters. We observe an excellent agreement considering classical and quantum approaches for the collective decay rates of all the modes. The decay rate of the bright state is slightly below the ideal situation \( (\Gamma_2 = 1.7\Gamma_1, \text{instead of} \)

\begin{table}[h]
\centering
\begin{tabular}{|c|c|c|c|c|}
\hline
\text{LSP}_{\alpha} & \text{n=1} & \text{n=2} & \text{n=3} \\
\hline
1/e & 1.71 & 1.61 & 0.39 & 0.29 \\
1/2/e & 1.71 & 1.60 & 0.40 & 0.29 \\
\hline
\end{tabular}
\caption{Plasmonic super/sub-radiant states with 2 emitters \( (h = 2 \text{ nm, } \omega_0 = 2.789 \text{ eV}) \). 25 modes ensure the convergency. The decay rate is normalized with respect to a single emitter decay rate from the same configuration.}
\end{table}
TABLE III: Plasmonic bright states with 6 azimuthal emitters considering single mode MNP response (LSP<sub>n</sub>). The mode field lines are indicated for LSP<sub>1</sub>.

\[ \Gamma_2 = 2\Gamma_1 \]. We check that \( \Gamma_S + \Gamma_A = 2\Gamma_1 \) for all symmetric/antisymmetric pairs in agreement with the Fig. \[. \] In order to deeper understand this behaviour, we consider each mode separately on table \[. \] The alternative charge distribution at the position of the dipole clearly leads to cancel the cooperative behaviour by destructive interferences. Similar behaviour explains the inhibition for six radial emitters (not shown) or for six azimutal emitters (see table \[. \]) The ring arrangement presents an ideal superradiant behaviour when LSP<sub>1</sub> is the only involved mode \( \Gamma_0 = 6\Gamma_1 \) but the presence of high order modes degrades the collective behaviour so that \( \Gamma_0 = 4.2\Gamma_1 \) considering all the modes. This is in agreement with the burst timescale observed in Fig. \[.

Conclusion and outlook. In summary, we derive a quantum approach for plasmonic superradiance and discuss the dynamics of cooperative emission. In full analogy with free-space superradiance, plasmonics superradiance is directly related to the indiscernability of the emitters with respect to LSP excitation. Therefore, the system evolves in a state invariant by emitter permutation (so-called Dicke states) and strong correlations build up between the emitters. However, destructive interference between LSPs could strongly degrade the collective behaviour so that accurate design of the system is necessary to achieve optimized plasmonic superradiant nanosources. Moreover, this work brings deeper understanding of light-matter at the nanoscale and could be helpful in designing surface plasmon laser (SPASER) since it relies on very similar mechanism. Finally, we expect that plasmonic superradiance could be experimentally studied on metallo-dielectric nanohybrids.

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