Non-hermitian Hamiltonian description for quantum plasmonics: from dissipative dressed atom picture to Fano states

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Abstract
We derive effective Hamiltonians for a single dipolar emitter coupled to a metal nanoparticle (MNP) with particular attention devoted to the role of losses. For small particles sizes, absorption dominates and a non-hermitian effective Hamiltonian describes the dynamics of the hybrid emitter-MNP nanosource. We discuss the coupled system dynamics in the weak and strong coupling regimes offering a simple understanding of the energy exchange, including radiative and non-radiative processes. We define the plasmon Purcell factors for each mode. For large particle sizes, radiative leakages can significantly perturbate the coupling process. We propose an effective Fano Hamiltonian including plasmon leakages and discuss the link with the quasi-normal mode description. We also propose Lindblad equations for each situation and introduce a collective dissipator for describing the Fano behavior.

Keywords: quantum plasmonics, non-hermitian, Fano states

1. Introduction

Cavity quantum electrodynamics (cQED) benefits from the long duration of the light–matter interaction in optical microcavities. This has opened the door to important applications including a low threshold laser [1], supercontinuum laser [2], or indistinguishable single photon sources [3]. In the strong coupling regime, the Jaynes–Cummings ladder anharmonicity can lead to photon blockade [4] and the coherence of the hybrid polariton states permits the realization of a low power laser [5]. Optical microcavities present extremely high quality factors but at the price of diffraction limited sizes, limiting integration capabilities. It is therefore of strong interest to transpose cQED to nanophotonics and plasmonics [6–13]. Particular attention has been devoted to the strong coupling regime [14–18] since it offers the possibility of controlling the dynamics of the light emission, as e.g. photon blockade [19, 20] or coherent control [21–23]. Moreover, quadrupolar forbidden atomic transitions can occur in plasmonic cavities thanks to the strong plasmon field gradient [24, 25]. In addition, in the weak coupling regime, the acceleration of single photon source cadency by coupling to plasmons opens the door to high operation speed quantum functionalities beyond the limited bandwidth (high Q) of optical microcavities systems [9, 26–28].

Quantum plasmonic systems behave like open quantum systems because of strong losses originating from absorption into the metal or from radiation leakages to the far-field. The dynamics of open quantum systems can be described considering either a master equation or a non-hermitian effective
Hamiltonian. Generally, the master equation is derived from a hermitian Hamiltonian by tracing out the baths into which the energy is lost. A non-hermitian effective Hamiltonian can also be derived describing the full dynamics of the same open system except the ground state of the system. More precisely, the non-hermitian Hamiltonian lacks the feeding term (e.g., laser pump) appearing in the master equation but is fully equivalent when no pumping occurs [29]. Compared to the master equation approach, the use of a non-hermitian Hamiltonian strongly reduces the required numerical resources when the number of atomic and plasmonic states increase. In addition, it is worthwhile to note that it is difficult to separate the plasmon contribution from the absorption and radiation baths so that deriving a master equation can be delicate. However, a master equation can be inferred from the non-hermitian Hamiltonian on the basis of its similarity with other open quantum systems. Therefore, we investigate in detail the properties of the non-hermitian effective Hamiltonian we recently derived for localized surface plasmons (LSPs) [22, 30]. We specifically discuss the role of losses in the effective Hamiltonian construction, leading to non-hermitian behaviors. LSPs constitute a benchmark for investigating non-hermitian behaviors by an analytical description and by experimental direct characterization of their response. For instance, self-hybridization of LSPs of different orders due to non-hermiticity has been recently demonstrated [31]. In the following, we investigate the dynamics of hybrid nanosources on the basis of the eigenmodes of the non-hermitian Hamiltonian and discuss the link with quasi-normal mode (QNM) approaches [32–34].

In section 2, we describe the main steps leading to the definition of an effective Hamiltonian. We identify the coupling strength and illustrate the procedure considering a quantum emitter coupled to a silver nanoparticle. We demonstrate that the hybrid nanosource optical response can be described in full analogy with a QED representation where dissipative localized surface plasmons (LSPs) play a role analogous to leaky cavity modes. The strong and weak coupling regimes are discussed in sections 3 and 4, respectively. Finally, in section 5 we investigate the impact of LSP leakage on the effective Hamiltonian structure, notably by introducing Fano states that originate from coupling the LSP discrete states to the free-space continuum. For this purpose, we start from a bath model inferred from the effective Hamiltonian derived in section 2 and discuss the role of the leakages in this model.

2. LSP field quantization and effective model

We represent in figure 1 the hybrid system that consists of a dipolar emitter close to a metal nanoparticle (MNP). The dielectric constant of the background medium is \( \varepsilon_b \). The dipolar quantum emitter is a two-level system (TLS) with ground and excited states \( |g\rangle \) and \( |e\rangle \) of energy \( \hbar \omega_g \) and \( \hbar \omega_e \), respectively. The dipole moment of the optical transition is denoted by \( \mathbf{d}_{eg} \).

The decay rate of the excited state is denoted by \( \gamma_0 = \gamma_{0\text{rad}} + \gamma_{0\text{NR}} \), including the radiative and intrinsic non-

\[
\begin{align*}
-\mathbf{d}_{eg} \cdot \mathbf{E}(r) \quad &\quad |e\rangle \\
\gamma_d \quad &\quad |g\rangle
\end{align*}
\]

Figure 1. Schematic of the hybrid system embedded in a background material with permittivity \( \varepsilon_p \). A spherical MNP of radius \( R \) and permittivity \( \varepsilon_m(\omega) \) is coupled to a TLS dipolar emitter. \( r_d \) and \( r \) refer to the emitter and detector positions, respectively. The inset describes the TLS system.

radiative contributions. \( \gamma_{0\text{rad}} \) is the radiative contribution in the homogeneous medium of optical index \( n_b = \sqrt{\varepsilon_b} \)

\[
\gamma_{0\text{rad}} = n_b \frac{\omega_0^3}{3\pi\varepsilon_0 c^3}
\]

and we define the intrinsic quantum yield \( \eta = \gamma_{0\text{rad}} / \gamma_0 \). The MNP is characterized by the dielectric constant \( \varepsilon_m(\omega) = \varepsilon_m(\omega) + i\varepsilon_m^{*}(\omega) \) that can be extracted from tabulated data or modeled with a Drude model. Without loss of generality, we assume a Drude-like behavior \( \varepsilon_m(\omega) = \varepsilon_\infty - \omega_p^2/(\omega^2 + i\Gamma_p \omega) \) with \( \varepsilon_\infty \approx 6 \), \( \hbar \omega_p = 7.90 \) eV, and \( \hbar \Gamma_p = 51 \) meV for silver.

The Hamiltonian of the coupled system reads as

\[
\hat{H} = \hbar \omega_0 \hat{\sigma}_e - i \hbar \frac{\gamma_0}{2} \hat{\sigma}_e + \int dr \int_0^{+\infty} d\omega \omega \hat{f}_e^\dagger(r) \cdot \hat{f}_e(r) - \left[ \hat{\sigma}_g \int_0^{+\infty} d\omega \hat{d}_{eg} \cdot \hat{E}_g^\dagger(r) + \text{H.c.} \right].
\]

\( \omega_0 = \omega_e - \omega_p \) is the transition angular frequency and we introduce the excited state population and raising operators of the emitter \( \hat{\sigma}_e = |e\rangle \langle e| \) and \( \hat{\sigma}_g = |e\rangle \langle g| \), respectively. In equation (2), the first term refers to the TLS energy and we have phenomenologically introduced the decay rate \( \gamma_0 \) of the excited state. The third term describes the total energy of the electromagnetic field where \( \hat{f}_e^\dagger(r) \cdot \hat{f}_e(r) \) is the polaritonic vector field operator at the position \( r \) associated with the creation (annihilation) of a quantum in the presence of the MNP. The last term describes the emitter-field interaction under the rotating-wave approximation.

The electromagnetic field must be quantized by taking into account the dispersing and absorbing nature of the metal [35–37]. Within the Langlevin type model of [35], the electric field operator can be written as

\[
\hat{E}_g(r) = i \sqrt{\frac{\hbar}{\pi \varepsilon_0}} k_0 \int dr' \sqrt{\frac{\omega_0}{\hbar}} G_e(r, r') \hat{f}_e^\dagger(r'),
\]

where \( k_0 = \omega/c \) is the wavenumber and \( G_e(r, r') \) is the Green tensor associated to the electric field response at position \( r \) from an excitation localized at \( r' \) in the medium. This expressions fails in describing the electric field operator in free-space (for which \( \varepsilon'' = 0 \)). Recent works discuss a general definition of the electric field operator including the free-space contribution [38].
In the following, we investigate the optical response of the hybrid system. The wavefunction of the hybrid system can be written at time \( t \) as

\[
|\psi(t)\rangle = C_e(t) e^{-i\omega t} |\varepsilon, \varnothing\rangle + \int \text{d} \mathbf{r} \ e^{-i\mathbf{p}\cdot\mathbf{r}} \ C_\omega(\mathbf{r}, t) \cdot |g, I_\omega(\mathbf{r})\rangle,
\]

(4)

where \( |\varepsilon, \varnothing\rangle \) refers to the emitter in its excited state and no LSP mode excited whereas \( |g, I_\omega(\mathbf{r})\rangle \) refers to the emitter in its ground state and a single excited polariton of energy \( h\omega \). The elementary excitation at position \( \mathbf{r} \) is defined through the action of the bosonic vector field operator on the vacuum state \( |\mathbf{r}_0(\mathbf{r})\rangle \otimes |\varnothing\rangle = |I_\omega(\mathbf{r})\rangle \).

Until now, we consider a continuous description for polaritons. However, the dynamics of the coupled system deserve attention regarding the excitation of LSP modes of the MNP. The Green tensor \( G \) governs this dynamics and contains all the modal information of the MNP in terms of the Mie expansion

\[
\hat{E}_\omega(\mathbf{r}) = \sum_{n=1}^{\infty} \hat{E}_{\omega,n}(\mathbf{r}),
\]

\[
\hat{E}_{\omega,n}(\mathbf{r}) = \int \frac{\hbar^2}{\sqrt{\varepsilon_0 k_0}} \int \text{d}r' \text{e}^{-i\mathbf{p}\cdot\mathbf{r}'} G_{\omega,n}(\mathbf{r}, \mathbf{r}') \hat{F}_{\omega,n}(\mathbf{r}')
\]

\[
G_{\omega,n}(\mathbf{r}, \mathbf{r}') = \sum_{n=1}^{\infty} G_{\omega,n}(\mathbf{r}, \mathbf{r}'),
\]

(5)

where \( G_n \) refers to the contribution of the \( n \)th plasmon \( \text{LSP}_n \) (dipolar plasmon for \( n = 1 \), quadrupolar plasmon for \( n = 2 \), etc.) so that \( \hat{\mathcal{E}}_{\omega,n} \) is the electric field operator associated to the \( \text{LSP}_n \) mode. This leads us to define the bosonic creation operator for a given position of the atomic system, satisfying the commutation relation \([\hat{a}_{\omega,n}(\mathbf{r}_d), \hat{a}_{\omega,n}(\mathbf{r}_d)'\rangle] = \delta(\omega - \omega')\delta_{n,n'}[22, 30]

\[
\hat{a}_{\omega,n}(\mathbf{r}_d) = \frac{1}{i\hbar \omega_{\omega,n}} \text{d}_{\omega,n} \cdot \hat{E}_{\omega,n}(\mathbf{r}_d),
\]

(6)

\[
|\omega_{\omega,n}(\mathbf{r}_d)|^2 = \frac{k_0^2}{2\pi \varepsilon_0} \text{im}[\text{d}_{\omega,n} \cdot G_{\omega,n}(\mathbf{r}_d', \mathbf{r}_d) \cdot \text{d}_{\omega,n}^*].
\]

(7)

The excitation at the frequency \( \omega \) of a single plasmon of order \( n \) is \( \omega_{\omega,n}(\mathbf{r}_d) \). Moreover \( \omega_{\omega,n} \) is the emitter-LSP \( n \) coupling which is the key parameter to build the effective model. Truncating the modal decomposition to the number \( N \) of modes involved in the coupling process, the full Hamiltonian (equation (2)) becomes (see [30, 39] for details)

\[
\hat{H} = \hbar \omega_0 \hat{\sigma}_e - i \hbar \frac{\gamma_0}{2} \hat{\sigma}_e + \sum_{n=1}^{N} \hbar \omega_{\omega,n} \hat{a}_{\omega,n}^\dagger(\mathbf{r}_d) \hat{a}_{\omega,n}(\mathbf{r}_d)
\]

\[
+i \hbar \sum_{n=1}^{N} \int_0^{+\infty} \text{d} \omega \omega_{\omega,n}(\mathbf{r}_d) \hat{a}_{\omega,n}(\mathbf{r}_d) \hat{\sigma}_e - \text{H.c.}
\]

(8)

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure2.png}
\caption{Coupling constant spectra for the six first \text{LSP}_n modes calculated using equation (7). The emitter is located 2 nm from the silver MNP. The Lorentzian fits follow equation (9).}
\end{figure}

Figure 2. Coupling constant spectra for the six first \text{LSP}_n modes calculated using equation (7). The emitter is located 2 nm from the silver MNP. The Lorentzian fits follow equation (9).

To finalize the effective model, we take benefit from the Lorentzian profile for each resonance. In figure 2, we plot the coupling constant \( |\omega_{\omega,n}|^2 \) for several modes. We observe an excellent agreement with a Lorentzian profile so that \( \omega_{\omega,n} \) can be written as

\[
\omega_{\omega,n} = \frac{\Gamma_n}{2\pi} \omega - \omega_n + \frac{i\Gamma_n}{2}.
\]

(9)

\( \omega_n \) and \( \Gamma_n \) are the mode resonance frequency and width, respectively. These parameters are deduced from a Lorentzian fit, but analytical expressions are also available in the near-field regime, revealing the radiative and non-radiative contributions to the mode’s rate of losses \( \Gamma_n \).

Finally, the effective Hamiltonian is obtained by integrating over the angular frequency \( \omega_0 \) in order to establish a set of \( N \) discrete modes. For this purpose, we define the \( n \)th plasmonic operator

\[
\hat{a}_{\omega,n}(\mathbf{r}_d) = \frac{1}{i\hbar \omega_{\omega,n}} \int_0^{+\infty} \text{d} \omega \omega_{\omega,n}(\mathbf{r}_d) \hat{a}_{\omega,n}(\mathbf{r}_d)
\]

(10)

and the wavefunction of the hybrid system takes the form

\[
|\psi_{\text{eff}}(t)\rangle = C_e(t) |\varepsilon, \varnothing\rangle + \sum_{n=1}^{N} C_n(t) |g, I_\omega(\mathbf{r}_d)\rangle,
\]

(11)

such that \( |I_\omega(\mathbf{r}_d)\rangle = \hat{a}_{\omega,n}(\mathbf{r}_d)|\varnothing\rangle \) defines the excitation of a single plasmon \( \text{LSP}_n \). This effective wavefunction lets the contribution of the TLS excited state and all plasmon modes explicitly appear. We explicitly indicate the dependence on the emitter position \( \mathbf{r}_d \) to indicate that the parameters of the effective model are defined for a given position of the atomic system. Since we are considering a single emitter coupled to the MNP, we can safely omit the explicit dependence on \( \mathbf{r}_d \) in the following. Finally, we define the effective Hamiltonian \( \hat{H}_{\text{eff}} \) so that \( \hat{i} \hbar \partial_t |\psi_{\text{eff}}(t)\rangle = \hat{H}_{\text{eff}} |\psi_{\text{eff}}(t)\rangle \). Identifying the dynamics of the wavefunctions in the discrete and continuum descriptions, we obtain the matrix representation of the effective Hamiltonian in...
the basis \(|e, \emptyset, |g, 1\rangle, \ldots, |g, N\rangle\) \([30, 39]\)

\[
H_{\text{eff}} = \hbar \begin{pmatrix}
-i\frac{\Delta_0}{2} & g_1 & g_2 & \cdots & g_N \\
g_1 & \Delta_1 - i\frac{\Gamma_1}{2} & 0 & \cdots & 0 \\
g_2 & 0 & \Delta_2 - i\frac{\Gamma_2}{2} & \cdots & \vdots \\
\vdots & \vdots & \ddots & \ddots & \vdots \\
g_N & 0 & \cdots & 0 & \Delta_N - i\frac{\Gamma_N}{2}
\end{pmatrix}
\]

(12)

where \(\Delta_n = \omega_n - \omega_0\) is the detuning of the LSP\(_n\) resonance from the TLS emission. The effective Hamiltonian describes the evolution of a sub-system (atomic states + plasmons) of a larger configuration (atom + phonon bath + radiation bath) so that it is non-hermitian and presents losses on its diagonal. This effective Hamiltonian provides a very practical representation of the hybrid configuration, presenting a direct analogy with a cQED system. \(g_n\) defines the coupling strength of the emitter to the MNP \(n\)th mode. \(\omega_n, \Gamma_n\) are the LSP\(_n\) frequency and rate of losses, respectively. \(\omega_0, \Gamma_0\) and \(g_0\) depend on the MNP material and its size but the coupling strength \(g_n\) also depends on the distance to the MNP. These parameters are deduced from a Lorentzian fit of the coupling constant \(\kappa_{\omega,\text{LM}}\) as presented in figure 2. We plot in figure 3 the coupling strength to the LSP\(_n\) \((n = 1-4)\) modes as a function of the distance. We superimpose the mode losses rate \(\Gamma_n\). We observe that mode losses are governed by Joule losses in the metal \((\hbar \Gamma_n \approx \hbar \Gamma_p \approx 51 \text{ meV})\).

For larger particles, the dipolar mode losses are larger due to higher radiative losses. This will lead us to propose a modified Fano effective Hamiltonian in section 5.

Finally, the coupling strength \((\hbar g_n)\) can overcome the plasmon losses \((\hbar \Gamma_n \approx 51 \text{ meV})\) and TLS losses \((\hbar \gamma_0 = 15 \text{ meV})\) at very short distances so that a strong coupling regime occurs \([40]\). Note that higher coupling strength can be expected for smaller separation distances. However, we have to keep in mind that for distances below 1 nm, non-local effects can occur and the dielectric function presents a \(k\) dependence \(\epsilon_{\omega,k}(\omega, k)\), not taken into account here, that can screen the coupling strength \([41-43]\).
It is worth noting that this effective Hamiltonian presents a one to one mapping with a non-hermitian Hamiltonian of cQED. By analogy to the cQED treatment, we can describe LSPs’ dissipation by the coupling to a continuum bath (see figure 4). We separate the system $S$ composed of emitter and LSPs from the environment $E$ associated with the bath. Hence, we introduce a new Hamiltonian leading to the same dynamics as the original one

\[ \hat{H}_{SE} = H_S + H_E + H_I, \]
\[ H_S = \sum_{n=1}^{N} \hbar \Delta_n \hat{a}_n^\dagger \hat{a}_n + \hbar \sum_{n=1}^{N} (g_n \hat{\sigma}_+ \hat{a}_n + g_n^* \hat{a}_n^\dagger \hat{\sigma}_-), \]
\[ H_E = \int d\omega \, \hbar \omega \sum_{n=1}^{N} \hat{b}_{\omega,n}^\dagger \hat{b}_{\omega,n}, \]
\[ H_I = i\hbar \int d\omega \sum_{n=1}^{N} \beta_n(\omega) (\hat{b}_{\omega,n}^\dagger \hat{a}_n - \hat{a}_n^\dagger \hat{b}_{\omega,n}). \]

The system Hamiltonian $H_S$ describes the interaction between the emitters and the LSPs and is hermitian. The environment Hamiltonian $H_E$ involves all the bath oscillators of energies $\hbar \omega$. For each cavity pseudo-mode of order $n$ we define a reservoir. The interaction between the system and the environment is described by the Hamiltonian $H_I$ where $\beta_n(\omega)$ characterizes the coupling between the pseudo-modes and their associated reservoirs. For flat coupling $\beta_n(\omega) \approx \beta_0(\omega) = \sqrt{1/2\pi}$, this new Hamiltonian leads to the following Lindblad equation [44–47]

\[
\frac{d\hat{\rho}(t)}{dt} = \frac{1}{i\hbar} [\hat{H}_S, \hat{\rho}(t)] + \mathcal{D}_0[\hat{\rho}(t)] + \mathcal{D}_{LSP}[\hat{\rho}(t)],
\]

where $\hat{\rho}$ is the density matrix of the emitter-LSPs system. The LSP dissipator naturally appears and is of the form

\[
\mathcal{D}_{LSP}[\hat{\rho}(t)] = -\sum_{n=1}^{N} \frac{\Gamma_n}{2} \left[ \hat{a}_n^\dagger \hat{a}_n \hat{\rho}(t) + \hat{\rho}(t) \hat{a}_n^\dagger \hat{a}_n - 2 \hat{a}_n \hat{\rho}(t) \hat{a}_n^\dagger \right],
\]

but we phenomenologically introduce the emitter dissipator

\[
\mathcal{D}_0[\hat{\rho}(t)] = -\frac{\gamma_0}{2} \left[ \hat{\sigma}_+ \hat{\sigma}_- \hat{\rho}(t) + \hat{\rho}(t) \hat{\sigma}_+ \hat{\sigma}_- - 2 \hat{\sigma}_- \hat{\rho}(t) \hat{\sigma}_+ \right].
\]

The direct derivation of $\mathcal{D}_0$ will be discussed in section 5.

The master equation (14) is a model for the same dynamics as the non-hermitian effective Hamiltonian (12) but also includes the dynamics of the fundamental state $|g, \varnothing\rangle$. However, it is worth noting that for the $N$ LSPs + 1 emitter states, the Lindblad master equation operates in a space of dimension $(N + 1)^2$ whereas the effective Hamiltonian work in a space of dimension $(N + 1)$, so that the effective Hamiltonian should be privileged when possible.

3. Dissipative dressed atom picture

Considering all the $N$ LSP modes plus the TLS excited state in the effective Hamiltonian (equation (12)), we define $N + 1$ hybrid modes that are the eigenvectors of $H_{eff}$. We denote their complex angular frequency by

\[
\lambda_m = \omega_m - i\frac{\gamma_m}{2}, \quad (m = 1, \ldots, 26),
\]

where $\lambda_m$ is the eigenvalue of the effective Hamiltonian.

For such dissipative systems, we have to define the right and left eigenvectors $|\Pi_m^R\rangle$ and $|\Pi_m^L\rangle$, respectively, satisfying $H_{eff}|\Pi_m^R\rangle = \lambda_m |\Pi_m^R\rangle$ and $H_{eff}^\dagger |\Pi_m^L\rangle = \lambda_m^* |\Pi_m^L\rangle$, $|\Pi_m^R\rangle, |\Pi_m^L\rangle = \delta_{mn}$. For a Hamiltonian of the form (12), one can simply connect them as follows (see appendix D) [48]:

\[
|\Pi_m^R\rangle = m_0 |e, \varnothing\rangle + \sum_{n=1}^{N} m_n |g, l_n\rangle,
\]
\[
|\Pi_m^L\rangle = -m_0^* |e, \varnothing\rangle + \sum_{n=1}^{N} m_n^* |g, l_n\rangle,
\]

where $m_0$ and $m_n$ give the weight of each mode $|e, \varnothing\rangle$ or $|g, l_n\rangle$. 

Figure 5. (a) Energy diagram of the dressed states. A thicker line indicates a stronger weight of the atom $|e, \varnothing\rangle$, left part of the diagram). (b) Polarization spectrum. Black lines indicate the dressed states frequencies. In (b) the green line corresponds to the emission frequency of the emitter ($\hbar \omega_0 = 2.94$ eV) leading to the strong coupling. The blue (magenta) line refers to the frequency $\Omega_2 (\Omega_3)$ of the dressed state $\Pi_2 (\Pi_3)$. The cyan line near $\hbar \omega \approx 2.9$ eV indicates the frequency of the $\Pi_3$ state. Reprinted with permission from [40]. Optical Society of America.
Finally, the wavefunction (expression 11) can be represented at time \( t \) by:

\[
|\psi_{\text{eff}}(t)\rangle = C_e(t)|e, \varnothing\rangle + \sum_{n=1}^{N} C_n(t) \cdot |g, L_n\rangle
\]

\[
= \sum_{m=1}^{N+1} \eta_m^\text{eff}\Pi_{m}^\text{eff} e^{-i\omega_{m}t},
\]

(20)

with \( \eta_m = \langle \Pi_m | \psi(0) \rangle = -m_0 \) if we assume an emitter initially in its excited state and no LSP mode populated. Therefore the hybrid system wavefunction is expanded on the non-hermitian Hamiltonian eigenmodes defining the atomic states dressed by LSP modes, as depicted in figure 5(a) [40]. It defines a Jaynes–Cummings ladder, that has a same form as a cQED model, and that can be probed considering the near-field emission spectrum. Indeed, for an emitter initially in its excited state, the polarization spectrum takes the form

\[
P(\omega) = \left[ \int_{0}^{\infty} dt e^{i\omega t} |C_e(t)|^2 \right]^2
\]

\[
= \sum_{m=1}^{N+1} \frac{m_0^2}{\omega - (\omega_0 + \omega_m) + i\gamma_m/2}^2.
\]

(21)

If the dressed states are well separated in energy, the near-field spectrum is approximated by a sum of Lorentzians peaked at their resonance energy and with a full-width at half maximum (FWHM) \( \gamma_m \). In the present case, some of the dressed states are not sufficiently separated so that the exact expression (equation (21)) has to be used but still the effective model gives a clear understanding of the Rabi splitting (see figure 5(b)).

However, the polarization spectrum lacks information on radiative and non-radiative emission processes. Moreover, experimental characterization generally relies on far-field emission spectrum. Qualitative understanding of the far-field behavior can be achieved considering the dipolar LSP_1 mode population \( |G_1(\omega)|^2 \). Indeed, the far-field radiated signal (in the whole space) can be written as

\[
P_{\text{rad}} = \frac{1}{2\pi}\gamma_{\text{rad}} P(\omega) \sim |G_1(\omega)|^2,
\]

(22)

where \( \gamma_{\text{rad}} \) is the radiative decay rate (at the angular frequency \( \omega \)) in presence of the MNP (see appendix C for the details). We compare in figure 6 the far-field emission at the detector position and the bright dipolar mode population spectrum. We obtain very good agreement, justifying that far-field emission is governed by the dipolar LSP_1 mode scattering. We observe again a Rabi splitting of 144 meV that is a reminiscence of the strong coupling regime observed in the polarization spectrum. However, the main contribution comes from the bright LSP_1 scattering near \( \omega = 2.79 \) eV.

### 4. Weak coupling regime: Purcell factor and Fermi’s golden rule

We are particularly interested in the role of losses on the hybrid system dynamics. In the previous section, we discussed the strong coupling regime and the effect of both absorption and radiative losses on the near-field and far-field spectra. Before investigating in detail the effective Hamiltonian modification in the presence of important radiative losses, it is worthwhile to discuss the dynamics of the coupled system in the weak coupling regime. This permits to introduce notably the Purcell factor before discussing how it is modified and why Fano states are introduced in the presence of radiative leakages.

From the effective Hamiltonian (equation (12)), the emitter and LSP population dynamics obeys

\[
\dot{C}_e(t) = -\frac{\gamma_0}{2}C_e(t) - i \sum_{n=1}^{N} \gamma_n C_n(t),
\]

\[
\dot{C}_n(t) = -i\gamma_n C_e(t) - \left( i\Delta_n + \frac{\Gamma_n}{2} \right) C_n(t).
\]

(23)

(24)

In the weak coupling regime, the population of the plasmon states remain small so that they can be adiabatically eliminated, that is \( C_n(t) \approx 0 \). We obtain

\[
C_e(t) = \left[ \frac{\gamma_0}{2}C_e(t) + \sum_{n=1}^{N} \frac{\gamma_n^2}{\gamma_n^2 + \Gamma_n/2} \right] C_e(t),
\]

\[
C_e(t) = C_e(0)e^{-i\delta\omega t}e^{-\gamma_{\text{tot}}t},
\]

(25)

where we recognize the Lamb shift and the total decay rate

\[
\delta\omega = \sum_{n=1}^{N} \frac{\gamma_n^2(\omega_0 - \omega_n)}{\left(\omega_0 - \omega_n\right)^2 + (\Gamma_n/2)^2},
\]

\[
\gamma_{\text{tot}} = \gamma_0 + \sum_{n=1}^{N} \frac{\gamma_n^2 \Gamma_n}{\left(\omega_0 - \omega_n\right)^2 + (\Gamma_n/2)^2}.
\]

(26)

(27)

We can define the Purcell factor \( F_p^n \) for each mode such that

\[
\frac{\gamma_{\text{tot}}}{\gamma_0} = 1 + \sum_{n=1}^{N} F_p^n \frac{1}{1 + 4\Omega_n^2 \left(\frac{\omega_0 - \omega_n}{\omega_n}\right)^2}
\]

(28)
along the TLS dipole moment (considered in the following). The normalized Fermi golden rule

\[ \gamma_n = \frac{g_n^2}{\gamma_0} \]

where \(Q_n = \omega_n/\Gamma_n\) is the LSP\(_n\) quality factor. Note that inserting the definition of the coupling strength (equations 7 and 9)) into the total decay rate (equation (27)), we recover the Fermi golden rule result [11]

\[ \frac{\gamma_{\text{tot}}}{\gamma_0} = 1 + \frac{2k_b^2}{h\gamma_0} \sum_{n=1}^{N} \text{Im}[\mathbf{d}_{eg} \cdot \mathbf{G}_{\omega,n}(\mathbf{r}_d, \mathbf{r}_f) \cdot \mathbf{d}_{eg}^\dagger] \]

\[ = 1 + \frac{6\pi}{k_b} \text{Im}G_{\omega,n}^{uu}(\mathbf{r}_d, \mathbf{r}_f), \]

where \(\eta\) is the intrinsic quantum yield \(\eta = \gamma_{\text{rad}}/\gamma_0\) and \(k_b = n_b k_0\). \(G_{\omega,n}^{uu} = \mathbf{u} \cdot \mathbf{G}_{\omega,n}^{uu} \cdot \mathbf{u}\) with \(\mathbf{u}\) an unitary vector along the TLS dipole moment (\(\mathbf{d}_{eg} = d_{eg}\mathbf{u}\)). Similarly, if we assume that the Green tensor follows a first order resonance (see e.g. Equation (B.25) in appendix B), the Lamb shift (equation (31)) can be rewritten as

\[ \frac{\omega_{LSP}}{\gamma_0} = -\frac{3\pi}{k_b} \text{Re}G_{\omega,n}^{uu}(\mathbf{r}_d, \mathbf{r}_f). \]

This Lamb shift is generally negligible and will not be considered in the following. The normalized Fermi golden rule (Equation (30)) and Lamb shift (equation (31)) are in full agreement with the result obtained from classical approach considering an oscillating dipole [49].

We consider in figure 7 a typical TLS emitting at \(\lambda = 670\) nm with \(\tau_0 = 1/\gamma_0 = 50\) ns and presenting an intrinsic quantum yield \(\eta = 90\%\). This corresponds to a dipole transition moment \(d_{eg} = 3.4\) D. The dynamics of the excited state close to the MNP is described in figure 7(a). In this weak coupling regime, we observe an exponential decay with a fluorescence lifetime \(\tau = 1.7\) ns in agreement with Fermi’s golden rule (\(\gamma_{\text{tot}}/\gamma_0 = \gamma_0/\tau = 30\) at \(d = 5\) nm, see figure 7(b)).

Actually, the TLS emission is not limited to a single value \(\omega_0\) but rather follows a Lorentzian profile. The contribution to the decay rate originating from the coupling to the LSP\(_n\) is therefore

\[ \gamma_n = \frac{g_n^2}{\gamma_0} \frac{\omega_0 / \gamma_0}{(\omega - \omega_0)^2 + (\Gamma_n/2)^2}, \]

\[ \mathcal{L}(\omega) = \frac{\gamma_0/2\pi}{(\omega - \omega_0)^2 + (\gamma_0/2)^2}, \]

where \(\mathcal{L}(\omega)\) is the free-space normalized emission spectrum of the TLS. This is the analog to the cQED description. Following the work of van Exter and coworkers [50], we can solve the integration over \(\omega\) as \(\gamma_n = 2\pi g_n^2 C_{LL}(0)\), where \(C_{LL}(u) = \int_{-\infty}^{\infty} \mathcal{L}(\omega)\mathcal{L}_n(u - \omega)\, d\omega\) is the convolution product with a normalized Lorentzian profile peaked at \(-\omega_0\) with an FWHM of \(\Gamma_n\). The convolution of two normalized Lorentzians is also a normalized Lorentzian and it yields

\[ \gamma_n = \frac{g_n^2(\gamma_0 + \Gamma_n)}{(\omega_0 - \omega_0)^2 + [(\gamma_0 + \Gamma_n)/2]^2}. \]

Even at ambient temperature, \(\gamma_0 \ll \Gamma_n\) so that expressions (27, 30) remain valid without working at cryogenic temperatures as for cQED. As expected, because of the strong subwavelength mode confinement, quantum plasmonics permits transposing cQED behavior to ambient temperature [51].

5. Leaky modes and LSP Fano states

For large MNP, the LSP modes can become strongly leaky, deforming the coupling strength spectrum, as shown in figure 8. Indeed, the dipolar plasmon LSP\(_1\) becomes strongly radiative so that \(|\kappa_i|^2\) does not follow a Lorentzian profile anymore [11]. In the following, we discuss how to modify the effective Hamiltonian to include this behavior. At this point, it is necessary to recall that the contribution of the free-space Hamiltonian does not follow a Lorentzian profile anymore [11]. In the following, we discuss how to modify the effective Hamiltonian to include this behavior. At this point, it is necessary to recall that the contribution of the free-space Hamiltonian does not follow a Lorentzian profile anymore [11].
5.1. Fano states for lossless TLS and MNP

5.1.1. Heuristic presentation of Fano states. Let us consider the ideal situation without absorption (TLS intrinsic quantum yield $\eta = 100\%$ and lossless metal $\Gamma_p = 0$). Radiation into the far-field is the only available channel for energy dissipation. Moreover, all LSPs modes are dark, except the LSP$_1$ dipolar mode, for not too large particles. Therefore, we consider the open quantum system schematic in figure 9 where the emitter and LSP$_1$ are coupled to the same radiation bath and we restrict the discussion to a single mode (LSP$_1$) MNP for the sake of clarity. In analogy to the work of Knight et al.\[52, 53\] we define a Fano state $|F\rangle$ diagonalizing the LSP$_1$-bath states and construct a modified effective Hamiltonian (see also section C$_I$ in \[54\]) in the basis $\{|e\rangle, |\epsilon\rangle, |F\rangle, |g\rangle\}$

$$H_{\text{eff}} = \hbar \begin{bmatrix} \omega_0 - i\frac{\gamma_0^{\text{rad}}}{2} & g_1 - i\frac{\sqrt{\gamma_0^{\text{rad}} \Gamma_1^{\text{rad}}}}{2} \\ g_1^* - i\frac{\sqrt{\gamma_0^{\text{rad}} \Gamma_1^{\text{rad}}}}{2} & \omega_1 - i\frac{\Gamma_1^{\text{rad}}}{2} \end{bmatrix}$$ \hspace{1cm} (34)

\(\gamma_0^{\text{rad}}\) and \(\Gamma_1^{\text{rad}}\) define the radiative contribution to the decay rates since no absorption is considered here.

5.1.2. Microscopic derivation of LPS’ Fano states. Generalization of the effective Hamiltonian (34) to all LSPs is not straightforward since the emitter and all LSPs could couple the free-space continuum for large particle. Therefore, we go back the bath model inferred in figure 4 where each LSP mode is associated to a specific reservoir. We add the direct emitter-radiative bath coupling ($C_0$) so that we consider...
the following Hamiltonian (compare with equation (13)):  
\[
\hat{H}_{SE} = H_S + H_E + H'_f, \\
H'_f = i\hbar \int d\omega \sum_{n=1}^{\infty} \beta_n^f(\omega)(\hat{b}^+_\omega \hat{a}_n + \hat{a}^+_\omega \hat{b}^r_\omega) \\
+ i\hbar \int d\omega \sum_{n=1}^{\infty} \zeta_n(\omega)(\hat{b}^r_\omega \hat{\alpha}_n - \hat{\alpha}^r_n \hat{b}^r_\omega).
\]
(35)

For flat couplings \( \beta_n^f(\omega) \approx \sqrt{\Gamma_n/2\pi} \), and \( \zeta_n(\omega) \approx \sqrt{\tilde{\gamma}_0(\omega_0)/2\pi} \) it leads to the following Lindblad equation:  
\[
\frac{d\hat{\rho}}{dt} = \frac{1}{i\hbar} [\hat{H}_S, \hat{\rho}(t)] + \mathcal{D}_F[\hat{\rho}(t)],
\]
where the new dissipator is  
\[
\mathcal{D}_F[\hat{\rho}(t)] = -\frac{1}{2} \sum_{n=1}^{\infty} \left[ \hat{c}_n \hat{\alpha}_n \hat{\rho}(t) + \hat{\rho}(t) \hat{\alpha}^+_n \hat{c}^+_n - 2 \hat{c}_n \hat{\alpha}_n \hat{\rho}(t) \hat{\alpha}^+_n \right].
\]
(36)

\( \gamma_{0r} \) refers to the emitter relaxation into the radiation bath \( \hat{b}_\omega \). The total radiative decay rate of the emitter in free-space obeys  
\[
\gamma_0 = \frac{2\kappa^2}{\hbar c} \text{Im}[\mathcal{G}_0(\mathbf{r}_0, \mathbf{r}_1, \omega_0) \cdot \mathbf{d}^*_{\mathbf{r}_0}]
\]
\[
= n_\text{a} \frac{d^2_{\mathbf{r}_0} \omega^3}{3\pi \epsilon_0 \hbar c^3}
\]
(38)
so that \( \gamma_{0r} \) corresponds to the decomposition of this decay rate on the spherical harmonics and \( \gamma_0 = \sum_{n=1}^{\infty} \gamma_{0r}(\omega_0) \) (see appendix A).

Finally, the dissipator (equation (37)) can be written as the sum of three contributions:  
\[
\mathcal{D}_F[\hat{\rho}(t)] = \mathcal{D}_0[\hat{\rho}(t)] + \sum_{n=1}^{N} \mathcal{D}_{LSR}[\hat{\rho}(t)] \\
+ \sum_{n=1}^{N} \mathcal{D}_{0,LSR}[\hat{\rho}(t)].
\]
(37)

5.2. General non-hermitian effective Hamiltonian

We now take into account the absorption into the metal (\( \Gamma_p = 0 \)) but still consider TLS intrinsic quantum yield \( \eta = 100\% \). The LSP, coupled to an additional non-radiative bath as shown in the schematic in figure 10. The Hamiltonian is written as  
\[
\hat{H}'_S = H_S + H_E + H', \\
H' = \int d\omega \frac{\kappa_1}{\hbar} \sum_{n=1}^{\infty} \hat{b}^+_\omega \hat{b}^r_\omega + \int d\omega \frac{\kappa_2}{\hbar} \sum_{n=1}^{\infty} \hat{b}^r_\omega \hat{b}^+_\omega.
\]
(40)

This leads to the following Lindblad equation:  
\[
\frac{d\hat{\rho}(t)}{dt} = \frac{1}{i\hbar} [\hat{H}_S, \hat{\rho}(t)] + \mathcal{D}_F[\hat{\rho}(t)] + \mathcal{D}_{nr}[\hat{\rho}(t)]
\]
(43)
with the additional non-radiative dissipator  
\[
\mathcal{D}_{nr}[\hat{\rho}(t)] = -\frac{1}{2} \sum_{n=1}^{N} \Gamma_{nr}^n \hat{a}_n \hat{\alpha}_n \hat{\rho}(t)
\]
\[
+ \hat{\rho}(t) \hat{a}_n^+ \hat{a}_n - 2 \hat{a}_n \hat{\rho}(t) \hat{a}_n^+\]
(41)
leading to the effective Hamiltonian

$$H_{\text{eff}} = \hbar \begin{pmatrix} -\frac{\gamma_0}{2} & g_1 - i \frac{1}{2} \sqrt{\gamma_0} \Gamma_{\text{rad}} & \cdots & g_N - i \frac{1}{2} \sqrt{\gamma_0} \Gamma_{\text{rad}} \\ g_1 & -\frac{\gamma_1}{2} & 0 & \cdots & 0 \\ \vdots & \vdots & \ddots & \ddots & \ddots \\ g_N & 0 & \cdots & -\frac{\gamma_N}{2} & 0 \end{pmatrix}$$

(44)

with $\Gamma_n = \Gamma_n^{\text{rad}} + \Gamma_n^{\text{nr}}$ the total decay rate of the LSP$_n$. We observe that the non-radiative processes increases the LSPs losses but does not play a role on the emitter-LSP coupling (off-diagonal elements). Actually, the Lindblad equation (equation (14)) proposed for small (non-leaky but absorbing) MNP corresponds to LSPs coupled to the non-radiative bath only but the emitter coupled to the radiative bath, explaining why the emitter spontaneous emission ($D_{\text{SP}}$) was phenomenologically introduced whereas it naturally appears within the bath model of figure 10.

Finally, the effective Hamiltonian formally takes the form in the LSPs Fano state basis $\{|e>, \{g, F_1\}, \ldots, |g, F_N\}\$

where $\alpha_n$ is the ratio between the coupling to the radiative quasi-continuum $(\alpha_n(\omega_0)) g_n = \sqrt{\gamma_0} (\omega_0) \Gamma_{\text{rad}}$ and the coupling to the discrete LSP$_n$ state (given by $g_n$). It depends on the emission angular frequency $\omega_0$. $q_{F, n} = 2/\alpha_n$ is the Fano parameter for the $n$th mode. It is equivalent to the exact discrete form (equation (12)) in the limit $\alpha_n \rightarrow 0$ corresponding to a negligible coupling to the quasi-continuum.

5.3. Weak coupling regime

We now discuss how the excited emitter dynamics is modified considering the new effective Hamiltonian. Here, we again consider adiabatic elimination in the weak coupling regime. The population dynamics then follows

$$C_e(t) = C_e(0)e^{-i\Delta_0 t}e^{-\gamma_0 t},$$

(46)
such that the Fano parameter obeys $g_i = -6.91 \times 10^{-2}\text{ meV}$ ($h = 15$ nm) or $g_1 = -4.03 \times 10^{-2}\text{ meV}$ ($h = 30$ nm). Finally, one can write the Purcell factor independently of the distance to the MNP as $F_p = \frac{4g_i^2}{\gamma_0\Gamma_n^\text{rad}}$ for $h = 30\text{ nm}$ and the Purcell factor is obtained from $F_p^\text{rad} = \frac{4g_i^2}{\gamma_0\Gamma_n^\text{rad}}$.

We can again express the total decay rate such that

$$\frac{\gamma_n}{\gamma_0} = \frac{1}{\gamma_0} + \sum_{n=1}^{\infty} \frac{\gamma_n}{\gamma_0},$$

$$\frac{\gamma_n}{\gamma_0} = F^n_p(\omega_0) = \frac{1}{1 + 4Q^2\left(\frac{\omega_0 - \omega_n}{\omega_n}\right)^2}$$

$$= \left[ 1 - \frac{\alpha_n^2(\omega_0)}{4} + 2\alpha_n(\omega_0)Q_n(\omega_0 - \omega_n) \right].$$

$\gamma_n$ refers to the decay rate into LSP$_n$. The dependency of the parameters on the emission frequency $\omega_0$ is explicitly indicated.

Since $\alpha_n$ only slightly depends on the emission frequency $\omega_0$, the Fano shape of the decay rate (equation (49)) is similar (at the first order in $\alpha_n$) to the one obtained by Sauvan and coworkers [32] considering a fully classical treatment of the atom-leaky mode coupling. They generalized the effective volume of the mode by defining a complex mode volume $V_n$ such that the Fano parameter obeys $\frac{2}{q_n}\approx \alpha_n(\omega_0) = \text{Im}(V_n)/\text{Re}(V_n)$, that characterizes the TLS coupling branch ratio to the leaky (quasi-continuum) and the discrete contribution to LSP$_n$ modes. Specifically, they use a quasi-normal mode analysis that accurately describes the mode leakage. Since the QNM corresponds to the pole of the Green tensor, the two approaches are fully equivalent, creating a bridge between the cQED and classical approaches in the weak coupling regime.

Assuming that the decay rate still obeys to the classical Fermi golden rule (equation (30)), we plot in figure 11(a) the decay rate to LSP$_1$ for a large (leaky) non absorbing silver nanoparticle. We observe a Fano behavior in agreement with expression (49). There are only three fitting parameters, namely $\omega_i$, $\Gamma_i^\text{rad}$, and $g_i$. The Fano parameter obeys $q_p = 2\sqrt{\alpha_i(\omega_0)} = 2\sqrt{\gamma_0(\omega_0)\Gamma_i^\text{rad}} = -4.2$ at $\omega_0 = \omega_1$ for $h = 30\text{ nm}$ and the Purcell factor is obtained from $F_p^\text{rad} = \frac{4g_i^2}{\gamma_0\Gamma_n^\text{rad}}$.

In figure 11(b), we include metal absorption. We use the same $\omega_i$, $\Gamma_i^\text{rad}$ and $g_i$ as for lossless configuration and fit the decay rate spectral behavior with one additional parameter; the LSP non-radiative rate $\Gamma_n^\text{nr}$. Finally, one can write the Purcell factor in presence of the lossy MNP as

$$F_p^n = \frac{4g_i^2}{\gamma_0\Gamma_n^\text{rad}} = \frac{\Gamma_n^\text{nr}}{\Gamma_n^\text{rad} + \Gamma_n^\text{nr}F_p^n}. \quad (50)$$

The total Purcell factor is the LSP quantum yield times the lossless Purcell factor. LSP losses therefore decrease the Purcell factor independently of the distance (but the lossless Purcell factor depends on the distance to the MNP). We achieve $F_p^1 = 12.2$ for $h = 30\text{ nm}$ and $F_p^1 = 35.1$ for $h = 15\text{ nm}$.

6. Conclusion

We have built effective Hamiltonians describing the emitter-MNP interaction extending the cQED approach to quantum plasmonics. We extensively discuss the role of Joule and radiative losses in the coupling process and their effect on the Hamiltonian’s structure. Because the effective Hamiltonian of the hybrid nanosource is time independent, we can introduce true energy levels (so-called dressed states) and discuss the effect of the atom–plasmon interaction on the wavefunction of the coupled system. We also discussed the link between near-field and far-field spectra with the population of the emitter and radiative dipolar plasmon, respectively. Moreover, with the quantized plasmon field, we can clearly identify...
the elementary process of spontaneous emission and we define a Purcell factor for each LSP. For large particles, we observe a Fano profile, fully explained considering a modified effective Hamiltonian, inspired from cQED considerations. We also derive Lindblad equations for each situation and introduce a collective dissipator to describe the Fano behavior. This clarify the role of radiative leakages (spontaneous emission) and overcome the difficulty of their phenomenological introduction that misses this collective dissipator. Finally, we stress that our formalism directly transposes cQED concepts to the nanoscale and therefore constitutes a powerful tool to propose and design ultrafast nanophotonics devices, taking advantage of the mode subwavelength confinement.

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Appendix A. Spherical particle—Mie expansion of the Green dyad

The Green dyad associated with the spherical particle is expressed using the Mie expansion [56, 57]

\[
G_0(\mathbf{r}, \mathbf{r}_d) = \frac{i k_b}{4 \pi} \sum_{e, o} \sum_{n=1}^{\infty} \sum_{m=0}^{n} (2 - \delta_{om}) \times \frac{2n + 1}{n(n + 1)} (n - m)! \times \frac{n(n + 1)(2n + 1)!}{n(n + 1)} \left[ A_n M_{am}^{(e)}(\mathbf{r}) \otimes M_{an}^{(o)}(\mathbf{r}_d) + B_n N_{am}^{(e)}(\mathbf{r}) \otimes N_{an}^{(o)}(\mathbf{r}_d) \right].
\]  

(A.1)

The formula of the spherical vector wavefunctions \( \mathbf{M}, \mathbf{N} \) can be found in [57]. The Mie coefficients are

\[
A_n = \frac{j_n(k_m R) \psi_n(z)}{h_n(1)k_m R \psi_n^{(0)}(k_m R)} - j_n(k_m R) \psi_n^{(0)}(k_m R),
\]

\[
B_n = \frac{k_m^2 j_n(k_m R) \psi_n(z)}{k_m^2 j_n(k_m R) \psi_n^{(0)}(k_m R)} - \frac{k_m^2 j_n(k_m R) \psi_n^{(0)}(z)}{k_m^2 j_n(k_m R) \psi_n^{(0)}(k_m R)}.
\]

(A.2)

(A.3)

where \( j_n \) and \( h_n^{(1)} \) are the spherical Bessel and Hankel functions and \( \psi_n(z) = z j_n(z), \psi_n(z) = z h_n^{(1)}(z) \) are the Ricatti–Bessel functions.

It can be also useful to expand the free-space Green tensor on the spherical harmonics.

\[
G_0(\mathbf{r}, \mathbf{r}_d) = \frac{\delta(\mathbf{r} - \mathbf{r}_d) e_r \otimes e_r + i k_b}{k_b^2} \sum_{p=e, o} \sum_{n=0}^{\infty} \sum_{m=0}^{n} \frac{(2 - \delta_{0m})(2n + 1)(n - m)!}{n(n + 1)(n + m)!} \times \left\{ \begin{array}{ll}
M_{am}^{(e)}(\mathbf{r}) \otimes M_{an}^{(o)}(\mathbf{r}_d) + N_{am}^{(e)}(\mathbf{r}) \otimes N_{an}^{(o)}(\mathbf{r}_d) & r > r_d \\
M_{am}^{(e)}(\mathbf{r}) \otimes M_{an}^{(o)}(\mathbf{r}_d) + N_{am}^{(e)}(\mathbf{r}) \otimes N_{an}^{(o)}(\mathbf{r}_d) & r \leq r_d
\end{array} \right.
\]

This expansion was used to calculate \( \gamma_{0m} \) in section 5.

Appendix B. Near-field coupling rate in the quasi-static approximation

In the following, we consider a dipolar emitter with radial orientation since it corresponds to the most efficient coupling. Then, we get

\[
G_s''(\mathbf{r}_d, \mathbf{r}_d) = \frac{i k_b}{4 \pi} \sum_{n=1}^{\infty} n(n + 1)(2n + 1) B_n \left[ \frac{h_n^{(1)}(k_b r_d)}{k_b r_d} \right]^2.
\]

(B.1)

B.1. Quasi-static approximation

We assume that the sphere radius is very small compared to the wavelength, i.e. \( k_b R \ll 1, |k_m R| \ll 1 \). Then, the Mie coefficient \( B_n \) can be approximated to (with \( u_b = k_b R, u_m = k_m R \))

\[
B_n \approx \frac{1}{i(2n - 1)!!(2n + 1)!!} \frac{(n + 1)u_m^n u_m^2(k_m^2 - k_m^2)}{[nk_m^2 + (n + 1)k_m^2]u_m^n u_m^{n + 1}}
\]

\[
\approx k_b^{2n+1} \frac{i(n + 1)}{(2n - 1)!!(2n + 1)!!} \frac{(\epsilon_s - \epsilon_B)^{2n+1}}{n \epsilon_s + (n + 1) \epsilon_B},
\]

(B.2)

(B.3)

where we have used the limiting values [58]

\[
\begin{align*}
\tilde{j}_n(z) &\sim \frac{z^n}{(2n + 1)!!}, \\
\tilde{\psi}_n(z) &\sim \frac{(n + 1)z^n}{(2n + 1)!!}, \\
\tilde{h}_n^{(1)}(z) &\sim \frac{z^{n + 1}}{(2n + 1)!!},
\end{align*}
\]

(B.4)

(B.5)

(B.6)

(B.7)

Finally, the Mie coefficient depends on the quasi-static polarisability for small particle size. Indeed, in the quasi-static regime, the optical response of the particle can be described using a multipolar expansion. If the particle excited with an incident field \( \mathbf{E}_0 \), the \( n \)th multipole tensor moment is given by

\[
p^{(n)} = \frac{4\pi \epsilon_0 c}{(2n - 1)!!} \alpha_n \nabla^{n-1} \mathbf{E}_0.
\]

(B.8)
so that the approximate form of the Mie coefficient (equation (B.6)) can be rewritten as

\[ B_n \approx i \frac{(n + 1) k_b^{2n+1}}{n(2n - 1)!!(2n + 1)!!} \alpha_n. \]  

(B.10)

### B.1.1. Resonance profile

For the sake of clarity, we assume that the surrounding medium is air, \( \epsilon_b = 1 \) (see [59] for \( \epsilon_b \neq 1 \)). Considering a Drude metal

\[ \epsilon_m = 1 - \frac{\omega_p^2}{\omega^2 + i \Gamma_\rho \omega} \]  

(B.11)

the \( n \)th resonance occurs for \( \epsilon_n(\omega_n) = -\frac{n}{n + 1} \epsilon_b \) that is for

\[ \omega_n = \omega_p \sqrt{n \frac{2n + 1}{2n + 1}} \]  

(B.12)

so that the \( n \)th polarizability becomes

\[ \alpha_n = \frac{n(\epsilon_s - 1)}{n \epsilon_s + (n + 1)} R^{2n+1}, \]  

(B.13)

\[ = \frac{-n \omega_p^2}{(2n + 1)(\omega^2 + i \Gamma_\rho \omega) - n \omega_p^2} R^{2n+1}. \]  

(B.14)

We now use \( n \omega_p^2 = (2n + 1) \omega_n^2 \) to write

\[ \alpha_n = \frac{-\omega_n^2}{\omega^2 - \omega_n^2 + i \Gamma_\rho \omega} R^{2n+1}, \]  

(B.15)

\[ = \frac{-\omega_n^2}{(\omega - \omega_n)(\omega + \omega_n) + i \Gamma_\rho \omega} R^{2n+1}. \]  

(B.16)

Finally, near a resonance, \( \omega \approx \omega_n \); we obtain

\[ \alpha_n \approx \frac{-\omega_n^2}{2(\omega - \omega_n) + i \Gamma_\rho \omega} R^{2n+1}, \]  

(B.17)

\[ \approx \frac{-\omega_n^2}{2(\omega - \omega_n) + i \Gamma_\rho \omega} R^{2n+1}. \]  

(B.18)

Thus \( |\alpha_n|^2 \) follows a Lorentzian profile peaked on the \( n \)th resonance \( \omega_n \) and with an FWHM of \( \Gamma_\rho \) associated with Joule losses in the metal.

### B.1.2. Radiative losses

In the previous section, only Joule losses appear although radiative losses are expected, at least for the dipolar \((n = 1)\) mode. This difficulty comes from the approximation that the electric field (or its \( n \)th order gradient for the next modes) is assumed constant over the particle size. Taking into account the variation of the electric field over the particle (or, more easily, applying the optical theorem to ensure the energy conservation), it is possible to show that the above expressions are improved using the effective polarizabilities [59, 60]

\[ \alpha_n^\text{eff} \approx \left[ 1 - i \frac{(n + 1) k_b^{2n+1}}{n(2n - 1)!!(2n + 1)!!} \right]^{-1} \alpha_n. \]  

(B.19)

that behave near a resonance as

\[ \alpha_n^\text{eff} \approx \frac{\omega_n}{2(\omega_n - \omega)} - i \frac{\omega_n R^{2n+1}}{2n \epsilon_s + (n + 1)} \]  

so that the approximate form of the Mie coefficient (equation (B.6)) can be rewritten as

\[ G_\text{eff}(r_i, r_d) \approx \frac{1}{4\pi \epsilon_0} \sum_{n=1}^{\infty} \frac{(n + 1)^2 R^{2n+1}}{r_d^{2n+4}} \frac{\omega_n}{2(\omega_n - \omega) - i \Gamma_n} \]  

(B.21)

so that the near-field coupling rate to LSP\(_n\) (equation (7)) is approximated by

\[ |\alpha_{\omega_n}(r)|^2 = \frac{k_0^2}{\hbar \pi \epsilon_0} \frac{d_{eg}^2}{\omega_n} \text{Im}[G_\text{eff}(r_i, r_d)] \]  

(B.22)

\[ \approx \frac{d_{eg}^2}{4\pi \epsilon_0 \epsilon_b} \frac{(n + 1)^2 R^{2n+1}}{r_d^{2n+4}} \frac{\omega_n/2}{(\omega_n - \omega)^2 + (\Gamma_n/2)^2} \frac{\Gamma_n}{2}. \]  

(B.23)

Identification with equation (9) leads to the following expression for the coupling strength:

\[ g_n \approx \sqrt{\frac{\omega_n/2}{\hbar \epsilon_0}} \frac{(n + 1) R^{n+1/2}}{2n \epsilon_b}. \]  

(B.24)

Finally, from equation (B.21), we obtain that the Green tensor follows a first order resonance

\[ G_\text{eff}^{(1)}(r_i, r_d) \approx \frac{\hbar \epsilon_0}{k_0^2 d_{eg}^2} \sum_{n=1}^{\infty} \frac{g_n^2(r_d)}{(\omega_n - \omega)^2 + (\Gamma_n/2)^2} \times \left[ -\omega - \omega_n + i \frac{\Gamma_n}{2} \right]. \]  

(B.25)

### Appendix C. Far-field spectrum

The light spectrum at position \( r_d \) is related to the polarization spectrum by [57]

\[ S(r, \omega) = \frac{1}{2\pi} \left| k_0 \mathbf{G}(r, r_d, \omega) \cdot \mathbf{d}_{eg} \right|^2 P(\omega). \]  

(C.1)
$k_0^2 \mathbf{G}(\mathbf{r}, \mathbf{r}_f, \omega) \cdot \mathbf{d}_{\mathbf{r}_f} / \epsilon_0$ describes the electric field scattered at the point $\mathbf{r}$ by a dipolar source located at $\mathbf{r}_f$ so that the far-field spectrum clearly appears as the signal propagating from the hybrid source to the detector position (including both scattering by the MNP and direct free-space propagation). The far-field signal is presented in figure C1(a) when scanning the TLS emission frequency $\omega_0$ [16]. We observe again a Rabi splitting of 144 meV for $\omega_0 = 2.94$ eV that is a reminiscence of the strong coupling regime observed in the polarization spectrum. However, the main contribution comes from the bright LSP$_1$ scattering near $\omega = 2.79$ eV (see also figure 6). Qualitative understanding of the far-field behavior can be achieved considering the dipolar LSP$_1$ mode population. Indeed, one can infer the total radiated power

$$P_{\text{rad}}(\omega) = \int S(\mathbf{r}, \omega) \, d^3 \mathbf{r}$$

and using far-field asymptotic expansion for the Green’s tensor, one obtains

$$P_{\text{rad}}(\omega) = \frac{1}{2\pi} \gamma_{\text{rad}}(\omega) P(\omega),$$

where $\gamma_{\text{rad}}$ is the radiative decay rate (at the angular frequency $\omega$) in presence of the MNP, that depends on the dipole moment orientation [61]. For small particle size, the far-field Green’s function is peaked at the dipolar LSP$_1$ frequency and the radiative rate is approximated by

$$\gamma_{\text{rad}}(\omega) \approx n_b \frac{d_{\text{eg}}^2 \omega^3}{3\pi \epsilon_0 \hbar c^3} \left[ 1 + \frac{4}{d^6} |\alpha_1(\omega)|^2 \right]$$

for a radial emitter close to the MNP.

Since the far-field emission should be governed by the LSP mode radiation, it is of strong interest to express the far-field spectra as a function of the LSP$_n$ population. The dynamics of the coupled system is governed by the effective Hamiltonian (12), $i\hbar \partial_t |\psi_{\text{eff}}(t)\rangle = \hat{H}_{\text{eff}}|\psi_{\text{eff}}(t)\rangle$. In particular,

$$\dot{C}_n(t) = -\frac{\gamma_0}{2} C_n(t) + \sum_{n=1}^{N} g_{n} C_n(t),$$

so that

$$C_n(\omega) = \frac{C_n(0) - \sum_{n=1}^{N} g_{n} C_n(\omega)}{i(\omega - \omega) + \gamma_0 / 2},$$

$$C_0(\omega) = \int_0^\infty dt e^{i(\omega - \omega_0)t} C_0(t) \, dt$$

and for an emitter initially in its excited state, the far-field radiated signal can be written as

$$P_{\text{rad}} = \frac{1}{2\pi} \gamma_{\text{rad}} P(\omega)$$

$$\approx n_b \frac{d_{\text{eg}}^2 \omega^3}{3\pi \epsilon_0 \hbar c^3} \left[ 1 + \frac{4}{d^6} |\alpha_1(\omega)|^2 \right] \frac{1 - \sum_{n=1}^{N} g_{n} C_n(\omega)}{i(\omega - \omega) + \gamma_0 / 2},$$

$$\approx n_b \frac{d_{\text{eg}}^2 \omega^3}{3\pi \epsilon_0 \hbar c^3} \left[ 1 + \frac{4}{d^6} |\alpha_1(\omega)|^2 \right] \frac{|1 - g_1 C_1(\omega)|^2}{(\omega - \omega_0)^2 + (\gamma_0 / 2)^2}$$

since the radiative decay rate selects the dipolar emission near $\omega \approx \omega_1$ for small particles. Finally, one can infer that the radiated power is proportional to the LSP$_1$ population in a rough approximation, $P_{\text{rad}} \sim |C_1(\omega)|^2$. We compare in figure C1 (see also figure 6 in the main text) the far-field emission and the bright dipolar mode population. We obtain very good agreement, justifying that far-field emission is governed by the dipolar LSP$_1$ mode scattering. We attribute the discrepancy to the fact that $P_{\text{rad}}$ describes the scattering in the whole space rather than in a specific direction as $S(\mathbf{r}, \omega)$.

**Appendix D. Biorthogonal basis of eigenvectors**

The aim is to construct the dual basis or left eigenvectors of a non-hermitian Hamiltonian $H$ defined by

$$\langle \Pi^R_m | H | \Pi^R_m \rangle = \lambda_m | \Pi^R_m \rangle$$

from the right eigenvectors

$$H|\Pi^R_m \rangle = \lambda_m |\Pi^R_m \rangle.$$

![Figure C1. Far-field emission. (a) Spectrum calculated at the detector position (equation (C.1)) varying the TLS emission angular frequency $\omega_0$. (b) LSP$_1$ population $|G_1(\omega)|^2$. All figures are normalized with respect to their maximum.](image)
We show below that these two sets of vectors satisfy the bi-orthogonality relation
\[ \langle \Pi^L_m | \Pi^R_n \rangle = \delta_{m,n}', \]  
(D.3)

(D.2) defines the non-unitary transformation \( T_R \) that diagonalizes \( H \)
\[ T_R^{-1} H T_R = D \]  
(D.4)
for which the right eigenvectors are on the column
\[ T_R = [|\Pi^R_1 \rangle \ |\Pi^R_2 \rangle \ ... \ |\Pi^R_M \rangle \]. \]

The dual basis can be defined from the matrix \( T_R \) as
\[ T^L_1 = T_R^{-1}, \]
(D.6)
where the left eigenvectors are on the row of the matrix \( T^L_1 \)
\[ T^L_1 = \left( \begin{array}{c} |\Pi^L_1 \rangle \\ |\Pi^L_2 \rangle \\ \vdots \\ |\Pi^L_M \rangle \end{array} \right). \]

According to the relation D.6, the bi-orthogonality relation D.3 is automatically satisfied since
\[ T^L_1 T_R = T_R^{-1} T_R = 1. \]
with
\[ T^L_1 H T_R = D. \]
(D.9)
We consider now the particular case where the Hamiltonian takes the form
\[ H = \begin{pmatrix} 0 & |g_1|e^{i\theta_1} & |g_2|e^{i\theta_2} & \cdots & |g_N|e^{i\theta_N} \\ |g_1|e^{-i\theta_1} & \Delta_1 - i\frac{\gamma_1}{2} & 0 & \cdots & 0 \\ |g_2|e^{-i\theta_2} & 0 & \Delta_2 - i\frac{\gamma_2}{2} & \cdots & \vdots \\ \vdots & \vdots & \ddots & \ddots & \vdots \\ |g_N|e^{-i\theta_N} & 0 & \cdots & 0 & \Delta_N - i\frac{\gamma_N}{2} \end{pmatrix}. \]
(D.10)

One can define a symmetric Hamiltonian from the unitary transformation
\[ H_S = S^\dagger H S, \]
(D.11)
where \( S \) is a diagonal matrix of the form
\[ S = \begin{pmatrix} 1 & 0 & 0 & \cdots & 0 \\ 0 & e^{-i\theta_1} & 0 & \cdots & 0 \\ 0 & 0 & e^{-i\theta_2} & \vdots & \vdots \\ \vdots & \vdots & \ddots & \ddots & \vdots \\ 0 & 0 & \cdots & 0 & e^{-i\theta_N} \end{pmatrix}, \]
(D.12)
with \( \kappa \) an arbitrary phase and \( S'S = I \). The new Hamiltonian reads as
\[ H_S = \begin{pmatrix} 0 & |g_1| & |g_2| & \cdots & |g_N| \\ |g_1| \Delta_1 - i\frac{\gamma_1}{2} & 0 & \cdots & 0 \\ |g_2| & 0 & \Delta_2 - i\frac{\gamma_2}{2} & \cdots & \vdots \\ \vdots & \vdots & \ddots & \ddots & \vdots \\ |g_N| & 0 & \cdots & 0 & \Delta_N - i\frac{\gamma_N}{2} \end{pmatrix}. \]
(D.13)

Indeed we can write
\[ H_S^\dagger |\psi^L_m \rangle = \lambda^*_m |\psi^L_m \rangle, \]
(D.15)
\[ H_S^\dagger |\psi^L_m \rangle = \lambda_m |\psi^L_m \rangle, \]
(D.16)
and make the link with the definition of the right eigenvectors
\[ H_S |\psi^R_m \rangle = \lambda_m |\psi^R_m \rangle. \]
(D.18)
Using the relation (D.14) and the transformation \( S \) allowing to express the right and left eigenvectors of \( H \) in terms of the right and left eigenvectors of \( H_S \)
\[ |\psi^R_m \rangle = S^\dagger |\Pi^R_m \rangle, \]
(D.19)
\[ |\psi^L_m \rangle = S^\dagger |\Pi^L_m \rangle, \]
(D.20)
we obtain the expression of the dual basis in terms of the right eigenvectors of \( H \)
\[ |\Pi^L_m \rangle = SS^\dagger |\Pi^R_m \rangle. \]
(D.21)
According to the matrix form of \( S \) (see equation (D.12)), we can write
\[ |\Pi^L_m \rangle = e^{2\kappa} \begin{pmatrix} 1 & 0 & 0 & \cdots & 0 \\ 0 & e^{-2i\theta_1} & 0 & \cdots & 0 \\ 0 & 0 & e^{-2i\theta_2} & \cdots & \vdots \\ \vdots & \vdots & \ddots & \ddots & \vdots \\ 0 & 0 & \cdots & 0 & e^{-2i\theta_N} \end{pmatrix} |\Pi^R_m \rangle. \]
(D.22)
The effective Hamiltonian studied in the article is defined with \( \theta_i = \frac{\pi}{2} \forall i \). The matrix form of \( SS^\dagger \) can then be written as
\[ SS^\dagger = \begin{pmatrix} -1 & 0 & 0 & \cdots & 0 \\ 0 & 1 & 0 & \cdots & 0 \\ 0 & 0 & 1 & \vdots & \vdots \\ \vdots & \vdots & \ddots & \ddots & \vdots \\ 0 & 0 & \cdots & 0 & 1 \end{pmatrix}. \]
(D.23)
where we have chosen the phase \( \kappa = \frac{\pi}{2} \). This completes the proof of equation (19).
**References**

[1] Nomura M, Iwamoto S, Kumagai N and Arakawa Y 2008 Ultra-low threshold photonic crystal nanocavity laser *Physica E* 40 1800–3

[2] Greif P (ed) 2016 *Nonlinear Optical Cavity Dynamics* (New York: Wiley)

[3] Laurent S, Varoutsis S, Le Gratiet L, Lemaitre A, Sagnes I, Raineri F, Levenson A, Robert-Philip I and Abram I 2005 Indistinguishable single photons from a single quantum-dot in a two-dimensional photonic crystal *Appl. Phys. Lett.* 87 163107

[4] Faroq A, Fushman I, Englund D, Stoltz N, Petroff P and Vuckovic J 2008 Coherent generation of non-classical light on a chip via photon-induced tunnelling and blockade *Nat. Phys.* 4 859–63

[5] Savvidis P 2014 A practical polariton laser *Nat. Photon.* 8 588

[6] Yannopapas V, Pasalakis E and Vitanov N V 2009 Plasmon-induced enhancement of quantum interference near metallic nanostructures *Phys. Rev. Lett.* 103 063602

[7] Cuche A, Mollet O, Drezet A and Huant S 2010 *Agio M 2012 Optical antennas as nanoscale resonators NanoScale* 4 692–706

[8] Tame M S, McEnery K R, Ozdemir S K, Lee J, Maier S A and Kociak M 2018 Strong enhancement of forbidden atomic transitions using plasmonic nanostructures *Phys. Rev. A* 022501 022501

[9] Lodahl P, Mahmoudian S and Stobbe S 2015 Interface singling of single photons with single quantum dots with photonic nanostructures *Rev. Mod. Phys.* 87 347

[10] Colas des Francs G, Barthes J, Bouhelier A, Weeber J-C, Dureux A, Cuche A and Girard C 2016 Plasmonic purcell factor and coupling efficiency to surface plasmons. Implications for addressing and controlling optical nanosources *J. Opt.* 18 094005

[11] Vaska P and Lienau C 2017 ‘Deterministic’ quantum plasmonics *Nano Lett.* 17 4566–70

[12] Trügler A and Hohenester U 2008 Strong coupling between a metallic nanoparticle and a single molecule *Phys. Rev. B* 77 115403

[13] Aberra-Guebrou S, Symonds C, Homeyer E, Plenet J C, Gartstein Yu N, Agranovich V M and Bellessa J 2012 Coherent emission from a disordered organic semiconductor induced by strong coupling with surface plasmons *Phys. Rev. Lett.* 108 066401

[14] Delga A, Feist J, Bravo-Abad J and Garcia-Vidal F J 2014 Quantum emitters near a metal nanoparticle: strong coupling and quenching *Phys. Rev. Lett.* 112 253601

[15] Zengin G, Wersal M, Nilsson S, Antosiewicz T, Käll M and Shegai T 2015 Realizing strong light–matter interactions between single-nanoparticle plasmons and molecular excitons at ambient conditions *Phys. Rev. Lett.* 114 157401

[16] Smolyaninov I, Zyayats A, Gungor A and Davis C 2002 Single-photon tunneling via localized surface plasmons *Phys. Rev. Lett.* 88 187402

[17] Alpeggiani F, D’Agostino S, Sanvitto D and Gerace D 2016 Visible quantum plasmonics from metallic nanodimers *Sci. Rep.* 6 34772

[18] Rousseaux B, Dzotsojan D, Colas des Francs G, Jaunlin H R, Couteau C and Guérin S 2016 Adiabatic passage mediated by plasmons: a route towards a decoherence-free quantum plasmonic platform *Phys. Rev. B* 93 045422

[19] Rousseaux B, Dzotsojan D, Colas des Francs G, Jaunlin H R, Couteau C and Guérin S 2016 *Phys. Rev. B* 94 199902

[20] Sacz-Blazquez R, Feist J, Fernandez-Dominguez A I and Garcia-Vidal F J 2017 Enhancing photon correlations through plasmonic strong coupling *Optica* 4 1363–7

[21] H Varguet, Rousseaux B, Dzotsojan D, Jaunlin H, Guérin S and Colas des Francs G 2016 Dressed states of a quantum
emitter strongly coupled to a metal nanoparticle Opt. Lett. 41 4480–3

[41] Leung P T 1990 Decay of molecules at spherical surfaces: nonlocal effects Phys. Rev. B 42 7622

[42] Castanie E, Boffety M and Carminati R 2010 Fluorescence quenching by a metal nanoparticle in the extreme near-field regime Opt. Lett. 35 291–3

[43] Girard C, Cuche A, Dujardin E, Arbouet A and Mayah A 2015 Molecular decay rate near nonlocal plasmonic particles Opt. Lett. 40 2116–9

[44] Hümmer T, García-Vidal F J, Martín-Moreno L and Zaeco D 2013 Weak and strong coupling regimes in plasmonic QED Phys. Rev. B 87 115419

[45] Rousseaux B, Baranov D, Käll M, Shegai T and Johansson G 2018 Quantum description and emergence of nonlinearities in strongly coupled single-emitter nanoantenna systems Phys. Rev. B 98 045435

[46] Hughes S, Richter M and Knorr A 2018 Quantized pseudomodes for plasmonic cavity QED Opt. Lett. 43 1834–7

[47] Varguet H, Guérin S, Jauslin H and Colas des Francs G 2018 Cooperative emission by quantum plasmonic superradiance arxiv:1810.11315

[48] Dridi D, Guérin S, Jauslin H R, Viennot D and Jolicard G 2010 Adiabatic approximation for quantum dissipative systems: formulation, topology, and superadiabatic tracking Phys. Rev. A 82 022109

[49] Metiu H 1984 Surface enhanced spectroscopy Prog. Surf. Sci. 17 153–320

[50] van Exter M P, Nienhuis G and Woerdman J P 1996 Two simple expressions for the spontaneous emission factor β Phys. Rev. A 54 3553

[51] Chikkaraddy R, de Nijs B, Benz F, Barrow S, Scherman O, Rosta E, Demetriadou A, Fox P, Hess O and Baumberg J 2016 Single-molecule strong coupling at room temperature in plasmonic nanocavities Nature 7 127–30

[52] Knight P, Lauder M and Dalton B 1990 Laser-induced continuum structure Phys. Rep. 190 1–61

[53] Durand P, Paidarova I and Gadea F 2001 Theory of Fano profiles J. Phys. B: At. Mol. Opt. Phys. 34 1953–66

[54] Cohen-Tannoudji C, Dupont-Roc J and Grynberg G 1996 Processus d’interaction entre photons et atomes (Paris: CNRS Editions)

[55] Prasanna Venkatesh B, Juan M and Romero-Isart O 2018 Cooperative effects in closely packed quantum emitters with collective dephasing Phys. Rev. Lett. 120 033602

[56] Chew H 1987 Transitions rates of atoms near spherical surfaces J. Chem. Phys. 87 1355–60

[57] Hakami J, Wang L and Zubairy M 2014 Spectral properties of a strongly coupled quantum-dot-metal-nanoparticle system Phys. Rev. A 89 053835

[58] Abramowitz M and Stegun I 1972 Handbook of Mathematical Functions (New York: Dover)

[59] Colas des Francs G, Derom S, Vincent R, Bouhelier A and Dereux A 2012 Mie plasmons: modes volumes, quality factors and coupling strengths (Purcell factor) to a dipolar emitter Int. J. Opt. 2012 175162

[60] Colas des Francs G 2009 Molecule non-radiative coupling to a metallic nanoparticle: an optical theorem treatment Int. J. Mol. Sci. 10 3931–6

[61] Colas des Francs G, Bouhelier A, Finot E, Weeber J-C, Dereux A, Girard C and Dujardin E 2008 Fluorescence relaxation in the near-field of a mesoscopic metallic particle: distance dependence and role of plasmon modes Opt. Express 16 17654–66