Directional launching of surface plasmons by polariton superradiance

Yu-Xiang Zhang, Yuan Zhang and Klaus Mølmer
Department of Physics and Astronomy, Aarhus University, DK-8000 Aarhus C, Denmark
(Dated: July 11, 2018)

The condition of phase matching prohibits the transfer of excitation from free-space photons to surface plasmon polaritons (SPP). We propose and analyze a scheme that optically prepares an ensemble of emitters in a collective timed-Dicke state, which is phase matched with the SPP by its position dependent excitation amplitudes. By a collective enhancement the ensemble, hence, emits an SPP in a well defined direction. Our theory, applicable also for other surface polaritons, incorporates the dispersive and dissipative properties of the plasmon modes to evaluate the non-Markovian emission by the ensemble.

Surface plasmon polaritons (SPP) are electromagnetic modes confined at metal-dielectric interfaces or near two-dimensional materials such as graphene. SPPs have shorter wavelength compared with free-space photons at the same frequency. This fact, occurring also for phonon polaritons, exciton polaritons and surface polaritons in heterostructures, leads to a wave number mismatch and prevents their effective production by conversion from free-space photons.

Conventional methods to close the mismatch either use prisms within the Otto configuration or the Kretschmann configuration to shorten the photon wavelength, or they lengthen the SPP wavelength or equip the SPP dispersion relation with band structure using grating couplers. In these cases, the launching of SPPs is directional because they inherit their wave vectors from the incident photons. Alternatively, a single, localized quantum emitter may absorb an optical photon and subsequently emit an SPP by spontaneous emission. The mismatch of wave number is thus circumvented but at the cost of efficiency and directionality since the SPP is emitted as a circular wave.

In this Letter, we shall show that emitter-mediated directional SPP launching, with its rich potential for applications, is possible via the mechanism of single-photon superradiance. While we shall focus on SPP, the theory will be applicable also to other surface polaritons.

When a photon is absorbed by a single emitter, the information of its wave vector is lost and has no impact on following emission processes. However, if the photon is uniformly absorbed by an ensemble of \( N \) emitters, its wave vector \( \mathbf{k} \) can be recorded by the emitters in the phases of the so-called timed-Dicke state,

\[
|\psi_k\rangle = \frac{1}{\sqrt{N}} \sum_{i=1}^{N} e^{i \mathbf{k} \cdot \mathbf{r}_i} |s_i\rangle \bigotimes_{j \neq i} |g_j\rangle, \tag{1}
\]

where \( |g\rangle \) is the emitter ground state, \( |s\rangle \) is an excited state, and \( \mathbf{r}_i \) is the position of the \( i^{th} \) emitter. In analogy with single-photon superradiance, spontaneous emission from \( |\psi_k\rangle \) generates a polariton excitation with wave vector \( \approx \hbar \mathbf{k} \) and energy \( \hbar \omega_{sg} = E_s - E_g \).

Directional SPP launching based on this process is possible only if \( \omega_{sg} \) and \( \mathbf{k} \) match the SPP dispersion relation. Therefore, we need a scheme for the preparation of \( |\psi_{k_{sp}}\rangle \) with the appropriate SPP wave vector \( \mathbf{k}_{sp} \), such that the SPP frequency \( \omega_{k_{sp}} = \omega_{sg} \).

**Preparation of \( |\psi_{k_{sp}}\rangle \):** We consider the simple case where SPPs are confined to an infinite plane interface, above which a parallel thin layer of emitters is deposited. The preparation of \( |\psi_{k_{sp}}\rangle \) proceeds by two steps.

**Step 1:** A single quantum is uniformly absorbed by a Raman process \( |g\rangle \rightarrow |e\rangle \) transition, e.g., following the heralded scheme, proposed in Refs. \( 23, 24 \) (the case of more excitations is discussed below). Applying optical fields propagating perpendicular to the emitter layer, see Fig (1a), the collectively shared excitation has no phase variation across the ensemble.

![Figure 1]( image)

**Figure 1.** Scheme for preparation of an SPP phase matched timed-Dicke state. (a) Raman excitation of one of the emitters into the excited state \( |e\rangle \), using uniform optical illumination perpendicular to the surface. (b) Illumination by a train of \( \pi \)-pulses on the \( |s\rangle \leftrightarrow |e\rangle \) transition, driving the emitters to the target timed-Dicke state \( |\psi_{k_{sp}}\rangle \).
Step 2: A train of \((2n_p + 1)\)\(\pi\)-pulses resonant with the \(|e\rangle\rangle\langle s|\) transition bounces the collective population of the emitters back and forth between \(|e\rangle\) and \(|s\rangle\), while the in-plane wave-vectors \(k_1\) or \(k_2\), see Fig. 1b), cause accumulation of a wave vector that we design to satisfy the equality \(-(2n_p + 1)k_1 + n_p k_2 = k_{sp}.\]

The combination of these processes produces the desired timed-Dicke state \(|\psi_{tD}\rangle\). Now let us comment on the apparent contradictory requirements \[23, 25\] that to make the superradiant emission dominate the incoherent emissions, the ensemble should be optically thick for the emitted mode \[22, 27\], while the presumed uniform optical excitation requires the ensemble to be optically thin during the state preparation \[25\]. We can indeed satisfy both conditions simultaneously: The optical processes are either driven orthogonally to the thin emitter ensemble or they act on only a single emitter population (of states \(|e\rangle\) and \(|s\rangle\)), thus the system is optically thin. The emission modes here are SPPs modes propagating parallel to the emitter layer, and the SPP-emitter interaction is collectively enhanced by the large number of atoms in the final internal state \(|g\rangle\). Thus, the system may be optically thick upon emission.

The state \(|e\rangle\rangle\langle s|\) is coupled to \(|g\rangle\) by a two-photon Raman process, and the collectively shared excitation in \(|e\rangle\rangle\langle s|\) may hence be stable against spontaneous decay to the atomic ground state \(|G\rangle = |g_1, g_2, \ldots, g_N\rangle\). During Step 2, until we have completed the pulse train, the intermediate timed-Dicke states \(|\psi_{-(n+1)k_1+n_k_2}\rangle\) \((n < n_p)\) may be protected from decay by the wave number mismatch, provided that the intermediate state lifetime supplies enough time window for the \(\pi\)-pulses.

The length of the pulse train depends on the ratio between the wavelength of optical photons (\(\lambda_{sp}\)) and the SPP wavelength (\(\lambda_{sp}\)), \(2n_p + 1 \simeq \lambda_{sp}/\lambda_{sp}\). For the values of \(\lambda_{sp}\), graphene SPP may serve as an example. Graphene SPPs are distinguished by their tight confinement and long lifetime, and by their high tunability via electrostatic gating \[2, 29\]. For SPPs with frequency \(\hbar\omega < 2E_f\) \[30\] where \(E_f \leq 1\) eV is the Fermi energy, the graphene surface conductivity is approximated by the Drude conductivity: \(\sigma_D(\omega) \approx i\frac{e^2}{m\omega}\omega/F(\omega + i\tau_D^{-1})\), the value of \(\tau_D\) currently available in experiments is 0.5 ps \[10\], while it may intrinsically reach values of 10^2 ps \[31\]. Supposing for simplicity a vacuum below and above the graphene monolayer, the dispersion relation of the p-mode graphene SPP is \(\omega_{sp} = \sqrt{2\alpha E_f k_{sp}/\hbar}\), where \(\alpha \approx 1/137\) is the fine-structure constant. Supposing \(E_f = 0.5\) eV, then for \(\hbar\omega_{sp} \in [0.01\text{ eV}, 1\text{ eV}]\) \[31\] \(\lambda_{sp}\) ranges from 90 \(\mu\)m to 18 nm.

For optical pulses \(\lambda_{sp} \in [380\text{ nm}, 750\text{ nm}]\), the number of pulses \(2n_p + 1 \simeq \lambda_{sp}/\lambda_{sp} < 50\) and even a single pulse is sufficient for low-energy SPPs with \(\lambda_{sp} > \lambda_{sp}\). We can drive the optical \(\pi\)-pulses on the time scale of nanoseconds using pulse powers that are far from damage graphene \[32\] and other surface polaritons.

**Emitter-SPP Coupling:** To study the lifetime of the intermediate states and also the emission from the prepared state \(|\psi_{tD}\rangle\), we now return to the general analysis concerning the coupling to the dispersive and dissipative electric field quantized as \[33, 33\]

\[
E(r_i) = i\mu_0 \frac{\hbar c_0}{\pi} \int_0^\infty \int d\omega \int d^3r' \omega^2 \sqrt{\epsilon(r',\omega)} \times G(r_1, r', \omega) \cdot f(r', \omega) + h.c.,
\]

where \(\mu_0\) and \(c_0\) are the vacuum susceptibility and permittivity; \(\epsilon(r',\omega)\) is the imaginary part of the relative permittivity; \(G(r, r', \omega)\) is the dyadic Green’s tensor determined by Maxwell’s equations, and the field \(f(r', \omega)\) with three Cartesian operator components \(f_a\) obeys the bosonic commutator relations \(\{f_a, f_b\} = 0\), \(\{f_a, f^\dagger_b\} = 0\) and \(\{f_a(r_1, \omega_1), f^\dagger_b(r_2, \omega_2)\} = \delta_{\omega_1\omega_2}\delta(r_1 - r_2)\delta(\omega_1 - \omega_2)\).

The Hamiltonian is written as \(H = \sum_{v=1}^N \frac{\hbar^2}{2}\omega_{sp} \sigma_v^z d_i \cdot E(r_i) + \int d^3r' \int_0^\infty d\omega \hbar c_0 f(r', \omega) \hat{f}(r', \omega)\)

\[
\Psi = \sum_{i=1}^{N} \alpha_i |s_i, \phi\rangle \otimes |g_i\rangle + \int_{a, , \omega, r'} \beta_a(\omega, r') |G, 1_a, \omega, r'\rangle
\]

where \(|\phi\rangle\) is the field vacuum state, \(\int_{a, , \omega, r'} f_a(r', \omega)|\phi\rangle = 0\) and \(\int_{a, , \omega, r'} f_a\) is the short hand for \(\sum_{a} \int d\omega \int d^3r'\). The strength of the emitter-emitter coupling mediated by all environmental modes

\[
g_{ij}(\omega) = \frac{\mu_0}{\pi} \omega^2 d_i \cdot G(r_i, r_j, \omega) \cdot d_j
\]

has the symmetry \(g_{ij}(\omega) = g_{ji}(\omega)\). Due to the in-plane translation symmetry (we assume that the dipoles of the emitters are identical) \[31\], \(g_{ij}(\omega)\) can be expanded in the wave number representation

\[
g_{ij}(\omega) = \int \frac{d^2k}{(2\pi)^2} g_{z_i z_j}(\omega, k_\perp)e^{i k_\perp \cdot (r_i - r_j)},
\]

where the subindex \(z_i, z_j\) indicates the emitter heights above the interface. For a thin emitter layer, we approximate all the emitter \(z\)-coordinates by a single value \(z_{at}\), and we thus express \(g_{z_i z_j}(\omega, k_\perp)\) as \(g_{zat}(\omega, k_\perp)\).

Similarly, the excitation amplitudes of the individual emitters defined in Eq. \[3\] can also be transformed into wave number representation, i.e., \(\alpha_{k_\perp}(t) = \langle \psi_{k_\perp}, \phi |\Psi\rangle\), which follows the equation

\[
-\partial_t \alpha_{k_\perp}(t) = N \int \frac{d^2q_\perp}{(2\pi)^2} \int \frac{d\omega}{2\pi} g_{zat}(\omega, q_\perp)\zeta(k_\perp, q_\perp) \times \int_0^t d\tau \alpha_{k_\perp}(\tau)e^{-i(\omega - \omega_{sp})(t-\tau)}
\]
where \( \mathcal{G} \) denotes the imaginary part and

\[
\zeta(q, k) = \langle \psi_q | \psi_k \rangle \tag{7}
\]

is a geometry factor which quantifies the sharpness of the phase matching condition given the spatial distribution of the emitters. If \( N \gg 1 \) and the emitters are for example distributed independently according to a Gaussian distribution with width \( L \), \( \zeta(k_q, q) = e^{-L^2(k_q - q)^2}/2 \).

The factor of \( N \) in Eq. \( \text{(9)} \) demonstrates the collective enhancement of the single-SPP superradiance. In the literature on single-photon superradiance, the collective emission rate and Lamb shift are usually obtained with the Markov approximation and without the rotating-wave approximation \[4, 24, 36, 37\]. For completeness, we give the corresponding formulae in Ref. \[38\].

**Evolution of |\( \psi_{k_{sp}} \rangle\)** - To analyze the evolution described by Eq. \( \text{(9)} \), we shall start from the initial state \( |\psi_{k_{sp}, 0}\rangle \) and focus on the state amplitude \( \alpha_{k_{sp}}(t) \). \( |\psi_{k_{sp}}\rangle \) resonantly matches the SPP with frequency \( \omega_{sp} \) while matched photon modes are off-resonant. Thus we may consider only the coupling to a range of SPPs. We use \( \omega_k \) and \( \gamma_k \) to denote the frequency and damping rate of the SPP labeled by the in-plane momentum \( k \). They are determined by the position of the pole of \( g_{z, z'}(\tilde{\omega}, k) \) in the complex \( \tilde{\omega} \) plane \[1\]. Keeping only the contribution from the poles leads to a Lorentzian type expression

\[
\mathbb{G} g_{z, z'}(\tilde{\omega}, k) \approx \frac{\mathcal{A}_z z'(k)}{(\tilde{\omega} - \omega_{k_{sp}})^2 + \gamma_{k_{sp}}^2}, \tag{8}
\]

where \( \mathcal{A}_z z'(k) \) is fixed by the residue of \( g_{z, z'}(\tilde{\omega}, k) \) at the pole \( \tilde{\omega} = \omega_{k_{sp}} - i\gamma_{k_{sp}} \).

When \( \zeta(k_{sp}) \) peaks sharply at \( k = k_{sp} \), the distribution of the emitter excitation is centered at \( |\psi_{k_{sp}}\rangle \), so that \( \alpha_{k_{sp}}(t) \approx \langle \psi_{k_{sp}} | \psi_{k_{sp}, 0} \rangle |\psi_{k_{sp}, 0}\rangle = \alpha_{k_{sp}}(t) \zeta(k_{sp}) \).

This approximation makes it possible to obtain a closed equation of evolution for \( \alpha_{k_{sp}} \), which, with the Gaussian distribution of emitters and the corresponding geometry factor, is written as

\[
-\partial_t \alpha_{k_{sp}} = \omega_{sp}^2 \int_0^t d\tau \alpha_{k_{sp}}(\tau) e^{-\frac{\gamma_{sp}^2}{4}(t-\tau)^2 - \gamma_{sp}(t-\tau)}, \tag{9}
\]

This expression verifies our observations in Fig. \[2\], e.g., that larger \( \omega_{sp} \) and \( \gamma_{sp} \) result in faster decay. When \( \omega_{sp}/L \) can be neglected, \( \Gamma_c \approx \omega_{sp}/\gamma_{sp} < \gamma_{sp} \).

Both the damped oscillation and pure decay regimes are achievable in experiments. With realistic parameters \( \omega_{sp} = E_f = 0.5 \text{ eV} \), the SPP group velocity is roughly \( v_{sp} = 10^{-2}c \), so that for \( L \geq 1 \mu m \), \( v_{sp}/L \leq 10^{12} \text{ Hz} = \gamma_{sp} \). For emitter vacuum decay rate \( 10^2 \text{ MHz} \) (governed by the transition dipole moment), emitter-graphene distance \( z_{at} = 10 \text{ nm} \) and emitter number density \( n_{at} = N/L^2 = (0.1/\text{nm})^2 \), the damped oscillation regime is reached with \( \omega_{sp} \approx 10^{14} \text{ Hz} \gg \gamma_{sp}, v_{sp}/L \)[38]. The pure decay regime can be realized by larger distance \( z_{at} \), lower density \( n_{at} \), or a smaller transition dipole moment.

**Evolution of Intermediate States** - Now we turn to the intermediate states \( |\psi_{q_n}\rangle \) that may be populated for nanoseconds durations in Step 2, where \( q_n = -(n+1)k_1 + nk_2 (0 \leq n < n_p) \). Since \( q_n \simeq (2n+1)\omega_{cs}/c \) and \( \omega_{cs} > \omega_{sg}, \omega_{sg}/c < q_n < k_{sp} \) and the modes matching the wave vector are not resonant with \( \omega_{sg} \); the matched SPP mode has frequency \( \omega_{q_n} < \omega_{sg} \) while photon scattering modes with in-plane wave vector \( q_n \) have frequencies no less than \( c\Delta \) and are hence blue-detuned from \( \omega_{sg} \). We denote the two detunings as \( \Delta = \omega_{sg} - \omega_{q_n} \) and \( \Delta_0 = c\alpha - \omega_{sg} \), respectively. For \( \omega_{cs} \) in the optical regime
and graphene SPP frequency $\omega_{sp}$ at most in the near-infrared, $\Delta_0 \gg \Delta$ ($\Delta_0$ ranges from optical to ultraviolet regimes) thus we can omit the coupling to free-space photons. Then the equation of evolution for $\alpha_{q_n}(t)$ resembles Eq. (9) but with the replacement $\gamma_{sp} \rightarrow \gamma_{sp} + i\Delta$ and $\omega_{sp}$, $\gamma_{sp}$ should be defined by the wave vector $q_n$ (see Ref. [38] for the full formulae). In the damped oscillation regime, we illustrate the solution to $|\alpha_{q_n}|$ in Fig. 3. It shows that compared with the phase-matched case ($\Delta = 0$), the intermediate states have longer lifetime, especially when $\omega_{sp}$ is smaller.

In the pure decay regime, the Markov approximation yields the decay rate of $|\psi_{q_n}|$.

$$\Gamma_{q_n} = \frac{\omega_{sp}^2}{\gamma_{sp} + i\Delta}$$

where we have omitted terms of order $v_{sp}/L$ that are dominated by $\gamma_{sp}$ in a large ensemble. Since $\gamma_{sp}$ is the uncertainty of SPP frequency, in practice we would have $\Delta \gg \gamma_{sp}$. For graphene SPP with $\hbar\omega_{sp} = E_f = 0.5$ eV and wavelength $\lambda_{sp} = 36.2$ nm, supposing optical pulse wavelength $\lambda_{ex} = 500$ nm, 15 pulses are sufficient to prepare $|\psi_{k_{sp}}\rangle$ and $\Delta \gtrsim 0.035$ meV $\approx 10\gamma_{sp}$ for all intermediate states $|\psi_{q_n}\rangle$ with $n < n_p$, and the lower bound is obtained upon $|\psi_{q_n}\rangle$ with $n = n_p - 1$. Equation (11) implies that when $\omega_{sp} \approx 0.1\gamma_{sp}$, $|\psi_{q_n+1}\rangle$ has a lifetime $10^6\gamma_{sp}^{-1} \approx 10$ ns, sufficient for the remaining two $\pi$-pulses required in order to prepare $|\psi_{k_{sp}}\rangle$.

However, in the damped oscillation regime the lifetime of the intermediate states may be too short to facilitate the preparation of $|\psi_{k_{sp}}\rangle$. In this case, we may employ another metastable emitter level $|s'\rangle$ which disallows the direct $|s'|-|g\rangle$ transition. The SPP wave number is accumulated with the $|e\rangle-|s'\rangle$ transition and finally we shall use one more pulse to move the collective population from $|s'\rangle$ to $|s\rangle$ and obtain $|\psi_{k_{sp}}\rangle$. This process is possible if the lifetime of the metastable $|s'\rangle$ is longer than the duration of the wave number accumulating pulses.

**Directionality of the emitted SPP**—Now we return to the emission from $|\psi_{k_{sp}}\rangle$. The amplitudes $\beta_n(\omega, \mathbf{r})$ defined in Eq. (3) can be transformed into wave number representation $\beta_n(\omega, \mathbf{k}_{sp}, z')$. Although exact solutions are not accessible, we may assume a uniform decay ansatz $\alpha_n(t) = \alpha_n(0)e^{-\gamma t}$, with which the electromagnetic frequency-wave number excitation distribution $P(k_{sp}, \omega)$ is

$$P(k_{sp}, \omega) = \frac{N^3 g_{sg}(\mathbf{k}_{sp})|\zeta(k_{sp})|^2}{\gamma^2 + (\omega - \omega_{sp})^2}.$$ 

Both the pole structure of $g_{sg}(\mathbf{k}_{sp}, \mathbf{k}_{si})$ and the geometry factor $\zeta(k_{sp}, \mathbf{k}_{si})$ in this formula guarantee the emission to peak sharply at $k_{sp} = k_{sp}$. The ratio between $P(k_{sp}, \omega_{sp})$ and the peak value $P(k_{sp}, \omega_{sp})$ is depicted in Fig. 4. The figure confirms that the larger ensemble size leads to stronger directionality.

**Conclusions and Discussions**—We propose to prepare timened Dicke states that match the wave vector of surface plasmon polaritons (SPP) and hence directionally emit SPPs via polariton superradiance. We find a non-Markovian behavior of the emitter excitation. With the Drude model parameters of graphene SPP, we predict excellent directionality of launching. Our main general formalism also applies to other families of surface polaritons [4, 10].

The requirement of only a single excitation in the timened Dicke state can be released to the low-excitation regime. In this regime, the spin operators can be approximated with bosons $a_i^{-\dagger} \rightarrow a_i$. We can translate them into momentum representation by $a_{q_n} = \frac{1}{\sqrt{N}} \sum_{i=1}^{N} a_i e^{-i q_n \cdot \mathbf{r}_i}$, with $[a_{q_n}, a_{q_n}^{-\dagger}] = \zeta(q_n, \mathbf{k}_{sp}) \approx \delta_{q_n, 0}$. The weak coherent state amplitudes then obey similar equations as the single excitation amplitudes in Eq. (3), see also Ref. [40]. Further studies may incorporate the inhomogeneous broadening and dephasing of the emitters relevant, for example, for doped rare-earth ions in crystals. Their influence on phase coherence has been studied in other contexts [41] while the effect on SPP emission may be minor because SPP modes have broad bandwidth in the range of THz. Besides the applications of directional SPPs [17, 21], our results may also facilitate interfaces between photonic and plasmonic systems for quantum information processing [16, 42]. Other phenomena related to
single-photon superradiance, such as superradiance amplification \[^{43}\] and superradiance lattice \[^{44}\], may also be investigated in surface polariton systems based on our scheme for timed-Dicke states.

Acknowledgement—This work was supported by European Unions Horizon 2020 research and innovation program (No. 712721, NanoQtech) and the Villum Foundation.

SUPPLEMENTAL MATERIAL

In the Supplemental Material, we shall present the derivation of the collective emission rate and the Lamb shift, Eq. (9) of the main text, details of the analysis of mismatched timed-Dicke states dissipation, and relevant information about the example of graphene surface plasmon polaritons.

A. Collective Emission Rate and Lamb Shift

Our ansatz for the quantum state goes beyond the rotating-wave approximation and is more general than Eq. (3) of the main text. With additional terms in the three-excitations manifold, the ansatz is written as

\[ |\Psi\rangle = \sum_{i=1}^{N} \alpha_i |s_i, 0\rangle \bigotimes_j |g_j\rangle + \int d\omega r' \beta_\omega(r', \omega) |G, 1_{a, \omega, r'}\rangle + \sum_{(i, j)} \int d\omega r' \xi_{ij,a}(\omega, r') |s_i, s_j, 1_{a, \omega, r'}\rangle \bigotimes_k |g_k\rangle, \]  

(S.1)

Equations for the time-dependent amplitudes of the above ansatz are given as

\[ -i\partial_t \alpha_i = \int d\omega r' g_a(r_i, r', \omega) \beta_\omega(r', \omega) e^{-i(\omega - \omega_{i, sg})t} + \sum_{j \neq i} \int d\omega r' \xi_{ij,a}(r', \omega) g_a(r_j, r', \omega) e^{-i(\omega_{i, sg} + \omega) t}, \]  

(S.2)

\[ -i\partial_t \beta_\omega(r', \omega) = \sum_{i=1}^{N} \alpha_i g_a^*(r_i, r', \omega) e^{i(\omega - \omega_{i, sg})t}, \]  

(S.3)

and

\[ -i\partial_t \xi_{ij,a}(r', \omega) = \alpha_i g_a^*(r_i, r', \omega) e^{i(\omega + \omega_{i, sg})t} + \alpha_j g_a^*(r_j, r', \omega) e^{i(\omega + \omega_{i, sg})t}. \]  

(S.4)

In the above equations,

\[ g_a(r_i, r', \omega) = i\mu_0 \sqrt{\frac{\epsilon_0 \Delta \epsilon(r', \omega)}{\pi}} \omega^2 \sum_b (d_b)_b G_{ba}(r_i, r', \omega) \]

We can formally solve Eqs. (S.3) and (S.4) with vanishing initial values of \( \beta_\omega(r', \omega) \) and \( \xi_{ij,a}(r', \omega) \). The following relation will be used in the calculation:

\[ \int d\omega g_a(r_i, r', \omega) g_a^*(r_j, r', \omega) = \frac{\mu_0}{\pi} \omega^2 d_i \cdot \Im\{G(r_i, r_j, \omega)\} \cdot d_j \equiv \Im g_{ij}(\omega). \]

Within the Markov approximation, we obtain the equation for the amplitudes of the individual emitters:

\[ -\partial_t \alpha_i(t) = \alpha_i(t) \int d\omega \Im g_{ii}(\omega) \xi_i(\omega, -\omega_{i, sg}) + \alpha_i(t) \sum_{j \neq i} \int d\omega \Im g_{jj}(\omega) \xi_i(\omega, \omega_{j, sg}) + \sum_{j \neq i} \alpha_j(t) \int d\omega \Im g_{ij}(\omega) \xi_i(\omega, -\omega_{j, sg}), \]  

(S.5)

where

\[ \xi_i(\omega_1, \omega_2) = \frac{1 - e^{-i(\omega_1 + \omega_2)t}}{i(\omega_1 + \omega_2)}. \]

The first and the third lines in Eq. (S.5) come from the “rotating wave” terms of the Hamiltonian, while the second and the fourth lines are attributed to the “counter-rotating wave” terms. Assuming \( \omega_{i, sg} = \omega_{sg} \) and taking the long time limit

\[ \xi_i(\omega, -\omega_{sg}) \rightarrow \frac{-i}{\omega - \omega_{sg} - i\epsilon}, \]

\[ \xi_i(\omega, \omega_{sg}) \rightarrow \frac{-i}{\omega + \omega_{sg} - i\epsilon}. \]

Eq. (S.5) can be evaluated as

\[ -\partial_t \alpha_i(t) = \alpha_i \left[ \pi \Im g_{ii}(\omega_{sg}) - i \int d\omega \Im g_{ii}(\omega) P \frac{1}{\omega - \omega_{sg}} \right] - i(N - 1) \alpha_i \int d\omega \Im g_{jj}(\omega) P \frac{1}{\omega + \omega_{sg}} \]

\[ + \sum_{j \neq i} \alpha_j \left[ \pi \Im g_{ij}(\omega_{sg}) - i \int d\omega \Im g_{ij}(\omega) P \frac{1}{\omega - \omega_{sg}} \right] - i \sum_{j \neq i} \alpha_j \int d\omega \Im g_{jj}(\omega) P \frac{1}{\omega + \omega_{sg}}, \]  

(S.6)

where \( P \) denotes the principal value integral and we have assumed the translation symmetry, i.e., \( \Im g_{ij}(\omega) \) is identical for all \( j \).

Next, we shall use the Kramers-Kronig relation

\[ \Re g_{ij}(\omega_{sg}) \equiv \frac{\mu_0}{\pi} \omega^2 d_i \cdot \Re G(r_i, r_j, \omega) \cdot d_j = \frac{2}{\pi} P \int_0^{\infty} d\omega \frac{\omega}{\omega^2 - \omega_{sg}^2} \Im g_{ij}(\omega). \]
After organizing terms from all four lines, we have

\[ \partial_t \alpha_i(t) = i \pi \sum_{j=1}^{N} \alpha_j(t) g_{ij}(\omega_{sg}) \]
\[ + i (N - 2) \alpha_i(t) \int_{\omega} 3 g_{ii}(\omega) \frac{1}{\omega + \omega_{sg}}. \]

We translate the amplitudes into wave vector representation

\[ \alpha_{q_i} = \frac{1}{\sqrt{N}} \sum_{i=1}^{N} \alpha_i(t) e^{-i q_i \cdot r_i}. \]

Then we obtain the equation for \( \alpha_{q_i} \):

\[ -i \partial_t \alpha_{q_i}(t) = (N - 2) \alpha_{q_i}(t) \int_{0}^{\infty} d\omega \frac{3 g_{ii}(\omega)}{\omega + \omega_{sg}} \]
\[ + \pi N \int \frac{d^2 k}{(2\pi)^2} \alpha_{k}(t) \zeta(q_i, k_i) g_{sz}(\omega_{sg}, k_i). \]  
(8.7)

Now we focus on \( q_i = k_{sp} \). Substituting the approximation \( \alpha_{k_i} = \alpha_{k_{sp}} \zeta(k_i, k_{sp}) \) into the above equation yields

\[ -i \partial_t \alpha_{k_{sp}}(t) = (N - 2) \alpha_{k_{sp}}(t) \int_{0}^{\infty} d\omega \frac{3 g_{ii}(\omega)}{\omega + \omega_{sg}} \]
\[ + \pi N \alpha_{k_{sp}}(t) \int \frac{d^2 k}{(2\pi)^2} |\zeta(k_{sp}, k_i)|^2 g_{sz}(\omega_{sg}, k_i), \]  
(8.8)

The collective level shift \( \delta \omega_{s} \) and decay rate \( \gamma_c \) can be extracted from the above equation,

\[ \delta \omega_{s} = -(N - 2) \int_{0}^{\infty} d\omega \frac{3 g_{ii}(\omega)}{\omega + \omega_{sg}} \]
\[ - \pi N \int \frac{d^2 k}{(2\pi)^2} |\zeta(k_{sp}, k_i)|^2 g_{sz}(\omega_{sg}, k_i), \]  
(8.9a)

\[ \gamma_c = \pi N \int \frac{d^2 k}{(2\pi)^2} |\zeta(k_{sp}, k_i)|^2 g_{sz}(\omega_{sg}, k_i). \]  
(8.9b)

Then we focus on the collective level shift of the emitter ground state \( |G\rangle \). We assume the ansatz

\[ |\Phi\rangle = |\beta |G, 0\rangle + \int_{\omega, r'} \sum_{i=1}^{N} \alpha_{i,a}(\omega, r') |e_i, 1_{a, r'}, r'\rangle \otimes |g_i\rangle, \]

and let \( \beta(t = 0) = 1 \). With the Markov approximation, the equation of evolution for \( \beta(t) \) gives the collective energy shift of the atomic ground state

\[ \delta \omega_{sg} = -N \int_{\omega} 3 g_{ii}(\omega) \frac{1}{\omega + \omega_{sg}}. \]  
(8.11)

The frequency shift between \( |\psi_{k_{sp}}\rangle \) and \( |G\rangle \) is given as \( \delta \omega_{s} - \delta \omega_{sg} \).

For the collective Lamb shift of \( |k_{sp}\rangle \), we have to subtract the the Lamb shift \( \delta \omega_{sg} \) of the system with only a single emitter, which can be obtained from Eqs. (8.6) and (8.11) with \( N = 1 \):

\[ \delta \omega_{sg} = - \int_{\omega} 3 g_{ii}(\omega) P_{\omega, \omega_{sg}}, \]  
(8.12)

The collective Lamb shift of the single-SPP superradiance, \( \Delta \omega_{sg} \), is hence determined from Eqs. (8.10), (8.11) and (8.12) as

\[ \Delta \omega_{sg} = \delta \omega_{s} - \delta \omega_{sg}. \]  
(8.13)

B. Derivation of Eq. (9) of the Main Text

The equations of evolution for the amplitudes introduced in Eq. (3) of the main text are

\[ -i \partial_t \alpha_i = \int_{r', \omega} g_a(\omega, r, r', \omega) \beta_a(r', \omega) e^{-i(\omega - \omega_{sg}) t}, \]  
(9.14)

and

\[ -i \partial_t \beta_a(r', \omega) = \sum_{j=1}^{N} \alpha_j g^*_a(r_j, r', \omega) e^{i(\omega - \omega_{sg}) t}. \]  
(9.15)

Substituting Eq. (8.13) into Eq. (8.14) yields

\[ -i \partial_t \alpha_{k_{sp}}(t) = \sum_{j=1}^{N} \int_{0}^{\infty} d\omega \int_{0}^{t} d\tau \alpha_{k_{sp}}(\tau) e^{-i(\omega - \omega_{sg})(t - \tau)}, \]

Then we go to the wave number representation and assuming the Gaussian distribution of the emitters as presented in the main text:

\[ -i \partial_t \alpha_{k_{sp}}(t) = N \int \frac{d^2 q}{(2\pi)^2} \int_{0}^{\infty} d\omega \int_{0}^{t} d\tau \alpha_{k_{sp}}(\tau) e^{-i(\omega - \omega_{sg})(t - \tau)} \]
\[ \times \int_{0}^{t} d\tau \alpha_{k_{sp}}(\tau) e^{-i(\omega - \omega_{sg})(t - \tau)}. \]  
(9.16)

We substitute the approximation of \( g_{sz}(\omega, q_i) \) introduced in Eq. (8) of the main text so that the integral over \( \tilde{\omega} \) becomes

\[ \int_{0}^{\infty} d\tilde{\omega} \int_{0}^{t} d\tau A_{sz}(\tilde{\omega}, q_i) e^{-i(\tilde{\omega} - \omega_{sg})(t - \tau)} \]
\[ \approx \pi A_{sz}(q_i) e^{-i(\omega_{sg} - \omega_{sz})(t - \tau)}. \]  
(9.17)

Then we substitute \( \alpha_{k_{sp}} \approx \alpha_{k_{sp}} \zeta(q_i, k_{sp}) \) and perform the integral over the in-plane momentum \( q_i \). We expand the expression as function of the surface plasmon frequency around this peak and get the approximation that

\[ \int \frac{d^2 q}{(2\pi)^2} \pi A_{sz}(q_i) e^{-L^2(q_i - k_{sp})^2 - i(\omega_{sg} - \omega_{sz})(t - \tau)} \]
\[ \approx \pi A_{sz}(k_{sp}) \int \frac{d^2 P}{(2\pi)^2} e^{-L^2 P_z^2 - iP_x v_{sp} - i\gamma_{k_{sp}}(t - \tau)}, \]
where we have made the substitution \( \mathbf{q}_i = \mathbf{p}_i - \mathbf{k}_{sp} \), and the surface plasmon frequency is expanded at \( \mathbf{q}_i = \mathbf{k}_{sp} \). The integral over \( \mathbf{p}_i \) can be written as

\[
\int \frac{p_i \, dp_i \, d\theta}{(2\pi)^2} \, e^{-L_z p_i^2 \parallel \mathbf{q}_i \mathbf{p}_i \cos \theta + \gamma_{sp} (t-\tau)} = \frac{1}{4\pi L^2} \, e^{-\frac{\gamma_{sp}^2}{4\pi^2} (t-\tau)^2 - \gamma_{sp} (t-\tau)},
\]

where we have assumed a constant SPP loss rate \( \gamma_{sp} \). Then we get Eq. (10) of the main text.

**C. Dissipation of Mismatched Timed-Dicke States**

For the initial emitter state \( |\psi_{\mathbf{k}_0}\rangle \) with \( \mathbf{k}_0 \neq \mathbf{k}_{sp} \), the SPP channel may or may not dominate the emission into photon free-space photon modes. The coupling strength to the free-space photon modes is [18]

\[
g^0(\tilde{\omega}, \mathbf{q}_i) = \frac{d^2}{\pi\epsilon_0} \frac{i}{2q_z} \mathbf{q}_i^2,
\]

where \( q_z = \sqrt{\tilde{\omega}^2/c^2 - \mathbf{q}_i^2} \), and we require that \( \Im q_z \geq 0 \). The subsequent calculation follows the outline of the previous section.

\[
\int_0^\infty d\tilde{\omega} \, \Im g^0(\tilde{\omega}, \mathbf{q}_i) e^{-i(\tilde{\omega} - \omega_{sp})(t-\tau)} = -\frac{i}{4\pi} d^2 \mathbf{q}_i^2 \Re \tilde{H}_0^{(2)}(\mathbf{q}_i, (t-\tau)) e^{i\omega_{sp}(t-\tau)},
\]

where \( \tilde{H}_0^{(2)} \) is the zero-order Hankel function of the second kind. To implement the integral over \( \mathbf{q}_i \), we write \( \tilde{H}_0^{(2)}(x) = \tilde{H}_0^{(2)}(x) e^{-ix} \) making use of the asymptotic behavior of Hankel functions. We fix the slowly varying part \( \tilde{H}_0^{(2)}(x) \) by its value at \( \mathbf{q}_i = \mathbf{k}_0 \) and integrate only the fast oscillating phase factor \( e^{-ix} \). This yields

\[
\int \frac{d^2 \mathbf{q}_i}{(2\pi)^2} e^{-L_z^2 (\mathbf{q}_i - \mathbf{k}_0)^2} \mathbf{q}_i^2 \Re \tilde{H}_0^{(2)}(\mathbf{q}_i, (t-\tau)) e^{i\omega_{sp}(t-\tau)} \approx \frac{k_0^2}{4\pi L^2} \Re \tilde{H}_0^{(2)}(\mathbf{k}_0, (t-\tau)) e^{-i\Delta_0(t-\tau) - \frac{\gamma_{sp}^2}{4\pi^2} (t-\tau)^2}.
\]

where \( \Delta_0 = \mathbf{k}_0 \cdot \omega_{sp} \). Meanwhile, the contribution from SPPs, Eq. (S.18), will acquire an additional off-resonant factor,

\[
\text{Eq. (S.18)} \rightarrow \frac{1}{4\pi L^2} e^{-\frac{\gamma_{sp}^2}{4\pi^2} (t-\tau)^2 - (\gamma_{sp} - i\Delta)(t-\tau)}.
\]

where \( \Delta = \omega_{sp} - \omega_{k_0} \) is the detuning between the SPP with momentum \( \mathbf{k}_0 \) and the emitter excitation. The final equation for the amplitude \( \alpha_{\mathbf{k}_0} \) is

\[
-i\partial_t \alpha_{\mathbf{k}_0}(t) = -i\omega_0^2 \int_0^t dt_{\tau} \alpha_{\mathbf{k}_0}(\tau) \tilde{H}_0^{(2)}(\mathbf{k}_0(t - \tau)) e^{-i\Delta_0(t-\tau) - \frac{\gamma_{sp}^2}{4\pi^2} (t-\tau)^2 - (\gamma_{sp} - i\Delta)(t-\tau)},
\]

where \( \omega_0^2 = \frac{N_c d^2 k_0^2}{(4\pi)^2 \epsilon_0 L^2} \).

**D. Graphene Surface Plasmon Polariton**

As already presented in the main text, the surface conductivity of the graphene monolayer given by the Drude model is

\[
\sigma(E_f, \omega) = \frac{e^2 E_f}{\pi\hbar^2} \frac{i}{\omega + i\tau_D}.
\]

This expression is convenient for our qualitative analysis when the temperature is low and \( \hbar\omega < 2E_f \).

For the graphene layer, the Fresnel coefficient of reflection of the p-modes is

\[
r_p(\tilde{\omega}, \mathbf{q}_i) = \frac{\sigma(\tilde{\omega}) \mathbf{q}_z}{2\omega_0 + \sigma(\tilde{\omega}) \mathbf{q}_z},
\]

where \( \mathbf{q}_z = \sqrt{\tilde{\omega}^2/c^2 - \mathbf{q}_i^2} \) and for graphene SPPs \( \mathbf{q}_z \approx iq_z \).

In the above expression, we have assumed that the dielectrics above and below the graphene monolayer are vacuum.

When the emitters are polarized perpendicular to the graphene layer, only one element of the scattering part of the dyadic Green’s tensor is relevant, which yields the coupling strength

\[
g_{z,z'}(\mathbf{q}_i, \tilde{\omega}) = \frac{i}{2\pi \omega_0 q_z} |d^2 q_z^2 r_p(\tilde{\omega}, \mathbf{q}_i) e^{-i\omega_0(z+z')}|.
\]

The poles that define the SPP are given by the equation

\[
\tilde{\omega}(\tilde{\omega} + i\tau_D^{-1}) = 2\alpha \omega_0 q_z\sqrt{\gamma_{sp}}/\hbar,
\]

where \( \alpha \approx 1/137 \) is the fine structure constant. Solutions of the above equation imply that \( \omega_{q_z} = \sqrt{2\alpha E_f q_z}/\hbar \) and inset.

\( \gamma_{q_z} = 0.5 \tau_D^{-1} \). Indeed, if the condition \( \omega_{q_z} \gg \gamma_{q_z} \) is not satisfied, the Drude model conductivity Eq. (S.23) should be replaced with more advanced expressions to yield well-defined SPPs.

The residue of \( r_p \) at the pole \( \tilde{\omega} = \omega_{q_z} - i\gamma_{q_z} \) is 0.5 \( \omega_{q_z} \), and \( A_{z,z'}(\mathbf{q}_i) \) defined in Eq. (8) of the main text is given as

\[
A_{z,z'}(\mathbf{q}_i) = \frac{3\hbar \omega_0^3}{4\omega_{q_z}^3} e^3 \omega_q q_z e^{-q_z(z+z')}.
\]
where we have used the vacuum spontaneous emission rate $\gamma_0$ to express the transition dipole.

For $E_I = 0.5$ eV, the SPP wave number is $q_{sp} = 1.074 \text{nm}^{-1}$ when $\hbar \omega_{sp} = 0.5$ eV. Suppose that the distance between the emitter layer and the graphene layer is $z_{at} = 10 \text{nm}$. Then we obtain $A_{z_{at}} = 1.87 \times 10^{20} \gamma_0 (\text{nm})^2 / \text{s}$. For larger distance, e.g., $z_{at} = 20 \text{nm}$, $A_{z_{at}} = 5.73 \times 10^{18} \gamma_0 (\text{nm})^2 / \text{s}$.