Novel signatures of quantized coupling between quantum emitters and localized surface plasmons

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Confining light to scales beyond the diffraction limit, quantum plasmonics supplies an ideal platform to explore strong light-matter couplings. The light-induced localized surface plasmons (LSPs) on the metal-dielectric interface acting as a quantum bus has a wide prospect in quantum information processing. However, the loss nature of light in the metal hinders its applications. Here, we propose a mechanism to make the reversible energy exchange and the multipartite quantum correlation of the QEs mediated by the LSPs persistent. Via investigating the quantized interaction between a collection of QEs and the LSPs supported by a spherical metal nanoparticle, we find that the diverse signatures of the quantized QE-LSP coupling in the steady state, including the complete decay, population trapping, and persistent oscillation, are essentially determined by different number of bound states formed in the energy spectrum of the QE-LSP system. Enriching our understanding on the light-matter interactions in lossy medium, our result is instructive to design quantum devices using plasmonic nanostructures.

Introduction.—Hybrid system composed of metal nanoparticle (MNP) and quantum emitters (QEs) have triggered intensive attentions in physics, chemistry, material, and life science [1–4]. By confining light within regions far bellow the diffraction limit in modes of localized surface plasmons (LSPs), the strong light-matter interaction is realizable in the vicinity of the MNP [5–13]. Recently, a dramatic progress has been made to explore the modified radiative properties of QEs by the LSPs in quantum plasmonics. Fascinating effects, including the superradiance of an ensemble of dipoles [14], the surface plasmon amplification by stimulated emission of radiation [15], the quantum statistics control of photons [16], and the suppression of quantum fluctuations of light [17], have been found. Whatever, it is the LSP mediated light-matter coupling that underlies such fascinating effects.

However, the intrinsic dissipations of light in the metal restrict the practical applications of LSPs in designing quantum devices [18]. It has been found that a QE residing near the metal is quenched by its decay through the nonradiative electromagnetic modes in the metal [19–22]. Such quenching hampers the complete quantum control in plasmonic system, where a persistent quantum coherence is of utmost importance [22, 23]. In the systems of a collection of QEs, the collectively-enhanced effect makes the strong coupling between the QEs and the radiative mode dominate the metal absorption [24] and suppresses quenching to the QEs [25]. It endows the multiple-QE system coupled to metal nanostructures a promising route for quantum devices [26, 27]. Going beyond the weak-coupling description to QE-LSP interactions [28–30], people found that the LSPs can act as a quantum bus to mediate the coherent interactions and generate the entanglement among QEs [31–33]. However, such quantum coherence is dynamically transient and tends to vanish in long-time limit. From a practical application point of view, a persistent quantum coherence and entanglement of the QEs is desired.

In this letter, going beyond the widely used pseudomode method, we investigate the exact dissipative dynamics of a collective QEs interacting with a MNP. The diverse signatures of the strong QE-LSP couplings, including the complete decay, population trapping, and persistent oscillations, are found in the long-time steady state. Our analyses reveal that they are determined by the formation of different numbers of QE-LSP bound states. Our result also shows that, quite different from the results under the Born-Markovian approximation and pseudomode method, a persistent quantum correlation among the QEs can be mediated by the LSPs even when the loss of the nanostructure are fully taken into account. Opening an avenue to achieve persistent quantum coherence and entanglement among the QEs by the mediation role of the LSPs, it is helpful for the practical application of quantum plasmonics.

System and quantization.—The system is composed of a MNP surrounded by N QEs. The QEs labeled by $l$ are distributed along a big circle with radius $r_l$ on the
equator of the MNP (see Fig. 1). Each QE is modeled as a two-level system with frequency $\omega_l$ and dipole moment $\mu_l$. The MNP has a radius $R$ and a dielectric permittivity denoted by a complex Drude model $\varepsilon_m(\omega) = \varepsilon_\infty - \omega_p^2/|\omega + i\gamma_p|$, where $\omega_p$ is the bulk plasma frequency, $\varepsilon_\infty$ is the high-frequency limit of $\varepsilon_m(\omega)$, and $\gamma_p$ is the Ohmic loss of light in the MNP [34]. The whole system is embedded in a homogeneous medium with dielectric constant $\varepsilon_d$. We consider that both of the dielectric and the metal are nonmagnetic and thus their permeability $\mu_d = \mu_m = 1$.

Once the QE is excited, it induces three modes: the radiative mode into the dielectric, the nonradiative mode absorbed by the metal, and the confined mode called LSPs near the metal-dielectric interface [35]. The LSPs enable a confinement of light within the deep subwavelength areas on the interface, which supplies an ideal platform to explore the strong quantized light-matter coupling [36, 37]. A macroscopic quantization method of light in the absorbing medium has been proposed based on the dyadic Green’s function, where the absorption of the medium to light is described by a Langevin noise [38, 39]. Then the electric field reads

$$\tilde{E}(r, \omega) = \frac{i\varepsilon \omega_0 \varepsilon^2}{\sqrt{\pi \varepsilon_0 \varepsilon}} \int d^3r' \sqrt{\varepsilon_m(\omega)} \mathbf{G}(r, r', \omega) \cdot \mathbf{f}(r', \omega),$$

where $\varepsilon_0$ is the vacuum permittivity, $c$ is the speed of light, and $\mathbf{f}(r, \omega)$ satisfying $[\mathbf{f}(r, \omega), \mathbf{f}(r', \omega)] = \delta(r-r') \delta(\omega - \omega')$ is the annihilation operator of light. The Green’s function $\mathbf{G}(r, r', \omega)$ denotes the field in frequency $\omega$ evaluated at $r$ due to a point source at $r'$, and satisfies the Helmholtz equation $[\nabla \times \nabla \times -\omega^2 \varepsilon_m(\omega)]\mathbf{G}(r, r', \omega) = \mathbf{I}(r-r')$, where $\mathbf{I}$ is the identity matrix. The spatial distribution of all of the three excitation modes has been incorporated into $\mathbf{G}(r, r', \omega)$. It allows for a complete description of the quantized light-matter coupling by calculating $\mathbf{G}(r, r', \omega)$.

The Hamiltonian of the coupled QE-MNP system under the dipole and rotating-wave approximations reads

$$\hat{H} = \sum_{l=0}^{N-1} \hbar \omega_l \hat{\sigma}^l_+ \hat{\sigma}^l_1 + \int d^3r \int d\omega \hbar \omega \tilde{f}^l(\mathbf{r}, \omega) \cdot \mathbf{f}(\mathbf{r}, \omega) - \sum_{l=1}^{N-1} \int d\omega [\mu_l \cdot \hat{E}(\mathbf{r}, \omega) \hat{\sigma}^l_1 + \text{H.c.}],$$

where $\hat{\sigma}^l_1 = |g_l\rangle \langle e_l|$ is the transition operator from the excited state $|e_l\rangle$ to the ground state $|g_l\rangle$ of the $l$th QE. The validity of the rotating-wave approximation in a related system has been revealed in [30]. The dipole approximation works when the QE size is sufficiently small [40–42].

**Exact dynamics.** We can see that the total excitation number $N = \sum_{l=1}^{N} \hat{\sigma}^l_1 \hat{\sigma}^l_1 + \int d^3r \int d\omega \tilde{f}^l_\omega(\mathbf{r}, \omega) \cdot \mathbf{f}(\mathbf{r}, \omega)$ is conserved. In the single-excitation subspace, the time-evolved state can be expanded as $|\Phi(t)\rangle = \left[ \sum_l c_l(t) \hat{\sigma}^l_1 + \int d^3r \int d\omega \tilde{b}_\omega(t) \tilde{f}^l_\omega(\mathbf{r}, \omega) \right] |G; \{0_u\} \rangle$, where $|G\rangle$ denotes all the QE's in the ground state, and $|\{0_u\} \rangle$ is the vacuum state of the total modes. It can be derived that $c_l(t)$ obey

$$\dot{c}_l(t) + i\omega_l c_l(t) + \int_0^t dt' \int_0^\infty d\omega e^{-i(\omega(t-t')} \mathbf{J}(\omega) c_l(t') = 0,$$

where $\mathbf{c}(t) = (c_0(t), \cdots, c_{N-1}(t))^T$ is a column vector and $\mathbf{J}(\omega)$ is a matrix with the elements $J_{ij}(\omega) = \omega^2 \mu_i^j \ln[\mathbf{G}(\mathbf{r}_i, \mathbf{r}_j, \omega)] + \mu_j^i / (\pi \hbar c^2 \omega^2)$ being the spectral densities. Thus all the actions of the metal-dielectric structure on the QEs has been collected in $\mathbf{J}(\omega)$. We have chosen the QEs having identical frequency $\omega_l = \omega_0$ and used $\int d^3s \frac{\omega_p^2}{\varepsilon_0} \ln[\varepsilon_m(\omega)] \mathbf{G}(\mathbf{r}, \mathbf{s}, \omega) \mathbf{G}^*(\mathbf{r}', \mathbf{s}, \omega) = \ln[\mathbf{G}(\mathbf{r}, \mathbf{r}')]$ [43]. The convolution in Eqs. (2) renders the QE dynamics non-Markovian. The correlation of different $c_l(t)$ indicates that although direct interactions of QEs in Eq. (1) are absent, the coherent couplings among them can be effectively induced by the mediation role of the LSPs.

The solution of Eq. (2) can be analyzed by Laplace transform, which yields $\tilde{c}(s) = \mathbf{V} \tilde{c}(s) \mathbf{V}^{-1} \mathbf{c}(0)$ with $\tilde{c}(s) = \{s + \omega_0 + \int_0^\infty d\omega \ln[\mathbf{D}(\omega)] - 1\}^{-1}$. We have used the Jordan decomposition of $\mathbf{J}(\omega) = \mathbf{VD}(\omega)\mathbf{V}^{-1}$ with $\mathbf{V}$ and $\mathbf{D}(\omega) = \text{diag}[D_0(\omega), \cdots, D_{N-1}(\omega)]$ being its similarity matrix and Jordan canonical form, respectively. Then $\mathbf{c}(t)$ is obtainable by inverse Laplace transform to $\tilde{c}(s)$, which can be done by finding its poles from

$$y_l(\varpi) = \omega_0 - \int_0^\infty \frac{D_1(\omega)}{\varpi - \omega} d\omega = \varpi, \quad (\varpi = is).$$

It can be proven that the roots $\varpi$ multiplied by $\hbar$ are just the eigenenergies of the whole system in the single-excitation subspace (see Supplementary Material [44]). Since $y_l(\varpi)$ is a monotonically decreasing function when $\varpi < 0$, each one of Eqs. (3) has one discrete root $\varpi_l^b$ if $y_l(0) < 0$. We call the discrete eigenstates with eigenenergy $\hbar \varpi_l^b$ the bound state. While in the region $\varpi > 0$, it has an infinite number of roots, which form a continuous energy band. Determined by the system parameters, at most $N$ independent bounds state could be formed. Using Cauchy residue theorem, we readily have $\mathbf{c}(t) = \mathbf{V} \tilde{c}(s) \mathbf{V}^{-1} \mathbf{c}(0)$ with the elements of $\mathbf{c}(t)$ as

$$\tilde{c}_l(t) = Z_l e^{-i\varpi_l^b t} + \int_{-\infty}^{i\varpi_l^b} \frac{d\varpi}{2\pi} \tilde{c}_l(-i\varpi)e^{-i\varpi t},$$

where the first term with $Z_l = [1 + \int_0^\infty \frac{D_1(\omega)}{\varpi_l^b - \omega} d\omega]^{-1}$ comes from the bound state and the second term are from the energy band. Oscillating with time in continuously changing frequencies, the second term in Eq. (4) behaves as a decay and tends to zero due to out-of-phase interference. Thus, if the bound state is absent, then $\lim_{t \to \infty} \mathbf{c}(t) = 0$ characterizes a complete decay; while if the bound states are formed, then $\lim_{t \to \infty} \mathbf{c}(t) = \mathbf{V}(\mathbf{Ze}^{-i\omega_l t})^{-1} \mathbf{c}(0)$ with $\omega_l = \text{diag}(\omega_0^b, \omega_0^b, \cdots, \omega_{N-1}^b)$ implies a decoherence suppression. Such results indicate
that the dynamics of the QEs in long-time limit is intrinsically determined by the energy-spectrum characters of the whole QE-LSP system. Generally, solving $\mathbf{V}$ and $\mathbf{D}(\omega)$ needs numerical calculations. Here, for concreteness, we choose that all the QEs have identical dipole moments and uniform coordinates $\mathbf{r}_l = (r, \pi/2, 2\pi l/N)$ such that $\mathbf{J}(\omega)$ is a symmetric circulant matrix with $J_{ij}(\omega) = J_{mn}(\omega)$ for $|i-j|=|m-n|$ (see Supplementary Material [44]). Because $\mathbf{J}(\omega)$ is a symmetric circulant matrix, we readily have $D_l(\omega) = \sum_{j} J_{ij}(\omega)\lambda_l^{-j}$ and $\mathbf{V} = (v_0, \ldots, v_{N-1})$ with $v_l = \frac{1}{\sqrt{N}}(1, \lambda_l, \ldots, \lambda_l^{N-1})^T$ and $\lambda_l = \exp(-2\pi il/N)$ [45].

**Results and discussion.**—It was previously found that the reversible energy exchange between the QEs induced by a common surface plasmon tends to vanish in the long-time limit under the Born-Markovian approximation [46]. Different from that result, we will show that such mediated coherent coupling can induce a persistently reversible energy exchange between the QEs even in the steady state when the approximation is relaxed. We choose the metal as silver with $\hbar\omega_p = 9.01$ eV, $\varepsilon_{\infty} = 3.718$, and $\hbar\gamma_p = 0.096$ eV in the interested frequency range [47] and the QEs with $\hbar\gamma_0 = 0.1$ meV. We focus on the QE dynamics by studying the initial-state fidelity $P(t) = |\langle \Phi(0) | \Phi(t) \rangle|^2$.

First, taking $N = 2$, we consider that only one of the QEs is excited initially, i.e., $|\Phi(0)\rangle = \delta_0^1|G; \{0_c\}\rangle$. We can calculate that with time evolution the fidelity reads $P(t) = |\langle c_0(t) \rangle|^2$. Figure 2(a) shows the evolution of $P(t)$ in three characteristic values of $r$. As a result of the near-field-enhancement of the LSPs, a significant oscillation appears in the dynamics for all the three cases. Absent in the Born-Markovian approximate result, this is entirely the non-Markovian effect, which represents a reversible energy exchange and thus manifests the strong coupling between the QEs mediated by the LSPs [36]. It is interesting to see that the non-Markovian effect manifests its action on the QEs not only in its transient dynamics, but also in its steady state. When $r = 9.5$ nm, $P(t)$ tends to zero accompanying the QEs decay completely to the ground state, which is consistent with the previous results [48, 49]. However, a remarkable difference appears with further decreasing $r$. One can see that $P(t)$ tends to a nonzero value when $r = 9.0$ nm, which represents a stable population trapping in the system; while when $r = 6.5$ nm, $P(t)$ tends to a lossless oscillation with a constant frequency, which quite likes the Rabi oscillation [50] and represents a persistent energy exchange among QEs caused by the QE-LSP interaction. These diverse signatures can be explained by our bound-state analysis. From Eq. (4), we readily have

$$\lim_{t \to \infty} |P(t)|^2 = \begin{cases} 0, & M = 0 \\ Z_0^2/4, & M = 1, \\ [Z_0^2 + Z_1^2 + D(t)]/4, & M = 2 \end{cases}$$

where $M$ is the number of the formed bound states and $D(t) = 2Z_0Z_1\cos[(\omega_0^b - \omega_0^a)t]$ is the interference between the two bound states. This conclusion can be confirmed by the energy spectrum shown Fig. 2(b). The two branches of bound states formed in the band gap divide the spectrum into three regions: without bound state when $r \gtrsim 9.0$ nm, one bound state when $8.5 \lesssim r \lesssim 9.0$ nm, and two bound states when $r \lesssim 8.5$ nm. The regions match well with the ones where the $P(\infty)$ shows different behaviors [see Fig. 2(c)], i.e., complete decay, population trapping, and persistent oscillation, as expected by Eq. (5). Such bound-state-favored behaviors are constructive to generate entanglement between the QEs. Different from the asymptotic vanishing in the Born-Markovian approximation [46] and in the absence the bound state, the generated entanglement can be preserved as long as the bound states are formed [see Fig. 2(d)]. This is helpful for utilizing plasmonic nanostructures in designing quantum devices. Our results can be generalized to the case of large number of QEs. With more of the bound states are formed in the large-$N$ case, the persistent oscillations will be complicated, but the mechanism is the same as the present case. In Supplementary Material [44], the dynamics for $N = 4$ is provided. Note that the similar bound-state-induced decoherence suppression for the single-QE case has been found in Refs. [29, 37].

Next, we consider that the QEs are initially in a...
FIG. 3. (a) Evolution of $P(t)$ with $r = 9.5 \text{ nm}$ in different $N$ obtained by the exact dynamics. (b) Long-time values of $P(t)$ obtained by the exact dynamics (red dots) and the bound-state analysis (green ×), respectively. (c) Eigenenergy of the bound state determined dynamics of the system. (d) Spectral density $J_0(\omega)$ and frequency of the dipole mode of the LSPs (gray dashed line). Other parameters are chosen the same as Fig. 2.

$W$-class state $|\Phi(0)\rangle = \frac{1}{\sqrt{N}} \sum_{l=0}^{N-1} \sigma_+^l |G\rangle$, which is a multipartite entangled state and widely used in quantum information processing [51, 52]. The canonical transformation $V$ can convert Eqs. (2) into $\dot{c}(t) + i\omega c(t) + \int_0^t d\tau \int d\omega e^{i\omega(t-\tau)} D(\omega) \dot{c}(\tau) = 0$ with $\dot{c}(t) \equiv V^{-1} c(t)$. Its initial condition can be calculated as $\dot{c}(0) = (1, 0, \ldots, 0)$, under which only the $\sigma_0(t)$-component of this matrix equation has nonzero solution. Thus its dynamics has a same equation of motion as the one of a single QE coupled to the LSPs [37]. It indicates that the $N$ QEs collectively act as a super two-level atom to interact with the LSPs with the spectral density characterized by $D_0(\omega)$. This notion of superatom is a powerful concept in designing single-photon quantum sources [53–56].

We can calculate the initial-state fidelity $P(t) = |\sigma_0(t)|^2$.

In the same mechanism as the case of $N = 2$, the entanglement of the QEs can be preserved in the steady state due to the formation of the bound state. Figure 3(a) shows the evolution of $P(t)$ in different number $N$ of QEs. It shows that $P(t)$ tends to a finite value for large $N$, where the QEs keep entangled. It can be understood from the bound-state analysis. As discussed above, we readily obtain $\lim_{t \to \infty} \sigma_0(t) = Z_0 e^{-i\omega_0 t}$ when Eq. (3) with $l = 0$ has an isolate root in the region $\omega_0 < 0$. Figures 3(b) and 3(c) show that the region where $P(t)$ tends to a stable value matches well with the one where a bound state is formed in the energy spectrum of the whole system. It verifies again our conclusion that it is the formation of bound state that preserves the entanglement in the steady state. We also plot in Fig. 3(d) the spectral density $D_0(\omega)$, which measures the coupling strength of the QEs and the LSPs. We can see that the contribution of the resonant dipole mode $\omega_1 = 3.77 \text{ eV}$ is entirely cancelled, while the one of the quadrupole mode $\omega_2 = 3.94 \text{ eV}$ is enhanced by increasing $N$ (see Supplementary Material [44]). It is due to the destructive interference of the undistinguished coupling-channels between different QEs and the LSPs [24, 56].

It is noted that the hybrid system studied here is experimentally realizable. The QEs could be $J$ aggregates and its strong coupling to LSPs has been investigated [57–60]. Although only the case that the dipole moments of the QEs are polarized along the radial direction is considered, our result can be generalized to other cases. Some quantitative difference might occur, but the diverse signatures of the quantized QE-LSP coupling in the steady state induced by different number of bound states does not change.

Conclusion.— We have investigated the exact dynamics of $N$ QEs coupled to the LSPs supported by a MNP. Quite different from the previous approximate result that the energy exchange among the QEs mediated by the LSPs exclusively tends to vanish, the diverse dynamical signatures of the quantized QE-LSP interactions, i.e., the complete decay, the population trapping, and the persistent Rabi-like oscillation, have been found. Our analysis indicates that it is the formation of bound states in the energy spectrum of the QE-LSP system that governs these rich behaviors. Such bound-state-assisted behaviors are helpful to the generation of stable quantum entanglement of the QEs. The further study on the multipartite $W$-class state demonstrates the collective suppression of the resonant dipole mode and the enhancement of the quadrupole mode to the QE-LSP coupling. Within the present experimental state of the art, our finding supplies a guideline for experiment to design quantum devices using the plasmonic nanostructures.

Acknowledgments.— The work is supported by the Natural Science Foundation (Grant Nos. 11704103, 11474139, and 11875150) of China, by the Doctoral Scientific Research Foundation of Henan Normal University (5101029170296), and by the Fundamental Research Funds for the Central Universities of China.

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Supplemental material for “Novel signatures of quantized coupling between quantum emitters and localized surface plasmons”

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GREEN’S FUNCTION OF THE SPHERICAL METAL NANOPARTICLE

In this section, we give the detailed derivations of the Green’s function of the spherical metal nanoparticle (MNP) in calculating exact dynamics of quantum emitters (QEs) coupled with localized surface plasmons (LSPs).

Given a spherical MNP with permittivity \( \varepsilon_m(\omega) \) and radius \( R \) embedded in a homogeneous medium with dielectric constant \( \varepsilon_d \), the Green’s function contributed from the free-space radiation sources and from the MNP-QE interaction are given by [1–3]

\[
G^0(r, r', \omega) = \frac{-\hat{r} \hat{r}'(r - r')}{k_0^2} + \frac{i k_1}{4\pi} \sum_{e, o} \sum_{n=1}^{\infty} \sum_{m=0}^{n} (2 - \delta_{0m}) \frac{2n + 1}{n(n+1)} \frac{(n-m)!}{(n+m)!} \times \left\{ 
\begin{array}{l}
[M^{(1)}_{mn}(k_1 r) M^{(1)}_{mn'}(k_1 r')] + N^{(1)}_{mn}(k_1 r) N^{(1)}_{mn'}(k_1 r'), \\
[M^{(1)}_{mn}(k_1 r) M^{(1)}_{mn'}(k_1 r')] + N^{(1)}_{mn}(k_1 r) N^{(1)}_{mn'}(k_1 r'), 
\end{array}
\right. \\
+ \sum_{i} \sum_{o} \sum_{n=1}^{\infty} \sum_{m=0}^{n} (2 - \delta_{0m}) \frac{2n + 1}{n(n+1)} \frac{(n-m)!}{(n+m)!} \frac{[R^H M^{(1)}_{mn}(k_1 r) M^{(1)}_{mn}(k_1 r')] + R^V N^{(1)}_{mn}(k_1 r) N^{(1)}_{mn}(k_1 r')}{|r - r'|},
\tag{S1}
\]

The upper (lower) line in Eq. (S1) holds for \( \hat{r} > \hat{r}' \) (\( \hat{r} < \hat{r}' \)), \( R^H \) and \( R^V \) are the scattering coefficients corresponding to the transverse electric field \( M^{(1)}_{mn} \) and the transverse magnetic field \( N^{(1)}_{mn} \) with even and odd contributions. According to the boundary conditions at the surface, \( R^H \) and \( R^V \) are given by

\[
R^H = \frac{\tau_2 \partial \tau_1 - \tau_1 \partial \tau_2}{\kappa_1 \partial \tau_2 - \kappa_2 \partial \tau_1}, \quad R^V = \frac{k_0^2 \tau_1 \partial \tau_1 - k_0^2 \tau_2 \partial \tau_2}{k_0^2 \kappa_1 \partial \kappa_1 - k_0^2 \kappa_2 \partial \kappa_2}, \tag{S3}
\]

where \( \tau_i = j_n(k_i R), \) \( \kappa_i = h_n^{(1)}(k_i R), \) \( \partial \tau_i = \partial_j\{\rho_{j_n}(\rho)\}_{j_n=k_i R} \) and \( \partial \kappa_i = \partial_j\{\rho_{h_n^{(1)}}(\rho)\}_{h_n^{(1)}=k_i R} \). Here, \( j_n(x) \) and \( h_n^{(1)}(x) \) are the spherical Bessel functions and the Hankel functions of the first kind, respectively, with \( k_1 = \omega \sqrt{\varepsilon_d}/c, \) \( k_2 = \omega \sqrt{\varepsilon_m(\omega)}/c \) being the wave vectors in the dielectric and the metal. The vector functions in spherical coordinate are defined as

\[
M^e_{mn}(kr) = -j_n(kr) \left[ \frac{m}{\sin \theta} P^m_n(\cos \theta) \sin m\varphi + \frac{d P^m_n(\cos \theta)}{d \theta} \cos m\varphi \right], \tag{S4}
\]

\[
M^o_{mn}(kr) = j_n(kr) \left[ \frac{m}{\sin \theta} P^m_n(\cos \theta) \cos m\varphi - \frac{d P^m_n(\cos \theta)}{d \theta} \sin m\varphi \right], \tag{S5}
\]

\[
N^e_{mn}(kr) = \frac{n(n+1)}{kr} - j_n(kr) P^m_n(\cos \theta) \cos m\varphi + \frac{1}{kr} \frac{d[r j_n(kr)]}{d r} \left[ \frac{d P^m_n(\cos \theta)}{d \theta} \cos m\varphi - \frac{m}{\sin \theta} P^m_n(\cos \theta) \sin m\varphi \right], \tag{S6}
\]

\[
N^o_{mn}(kr) = \frac{n(n+1)}{kr} j_n(kr) P^m_n(\cos \theta) \sin m\varphi + \frac{1}{kr} \frac{d[r j_n(kr)]}{d r} \left[ \frac{d P^m_n(\cos \theta)}{d \theta} \sin m\varphi + \frac{m}{\sin \theta} P^m_n(\cos \theta) \cos m\varphi \right]. \tag{S7}
\]

where \( P^m_n(x) \) are the associated Legendre polynomials.
In the case that the dipole moments of the QEs are polarized along the radial direction, only the \( rr \) component of the Green's function contributes to the light-matter interaction. In the studied structure, the QEs labeled by \( l \) are located at \( \mathbf{r}_l = (r, \pi/2, 2\pi l/N) \) with \( l = 0, \ldots, N - 1 \). We obtain

\[
\mathbf{G}_{rr}(\mathbf{r}_i, \mathbf{r}_j, \omega) = -\frac{\delta(\mathbf{r}_i - \mathbf{r}_j)}{k_1^2} + \frac{ik_1}{4\pi} \sum_{n=1}^{\infty} \sum_{m=0}^{n} c_{mn} \cos[2\pi m(l - j) + \mathcal{R}^V h_n^{(1)}(k_1 r)] / (k_1 r/P_n^m(0))^2,
\]

where \( c_{mn} = (2 - \delta_{0m})n(n+1)(2n+1)(n-m)!/(n+m)! \) and both of the contributions from the free-space field and the scattered field have been incorporated. From the definition, the spectral density characterizing the coupling strength between QEs and the LPSs can be calculated. They can be obtained as

\[
J_{ij}(\omega) = \frac{3\gamma_0 \omega^3 \sqrt{\varepsilon_d}}{4\pi \omega_0^2} \text{Re}\left[ \sum_{n=1}^{\infty} \sum_{m=0}^{n} c_{mn} \cos \left\{ \frac{2\pi m(l - j) + \mathcal{R}^V h_n^{(1)}(k_1 r)]}{(k_1 r/P_n^m(0))^2} \right\} \right],
\]

with the scattering coefficient in Eq. (S2) replaced by \( \mathcal{R}^V_n \). The poles of \( \mathcal{R}^V_n \) determines the resonance frequency of the LSPs. In this manner, the LSPs is expressed as a series of resonant modes labeled by \( n \) with eigenfrequency determined by

\[
n\varepsilon_m(\omega_n) + (n+1)\varepsilon_d = 0,
\]

from which the contributions of the different resonant modes of LSPs to the light-matter interaction can be studied. In the low-frequency condition, the resonant frequencies can be determined by \( \text{Re}[\varepsilon_m(\omega_n)] = -(n+1)\varepsilon_d/n \) due to \( \text{Re}[\varepsilon_m(\omega)] \gg \text{Im}[\varepsilon_m(\omega)] \). The first resonant mode is called the dipole mode and the second one is quadrupole mode [5]. Using the parameters in our system, we can calculated the frequencies of the dipole and quadrupole modes \( \omega_1 = 3.77 \text{eV} \) and \( \omega_2 = 3.94 \text{eV} \).

**EIGENENERGIES OF THE SYSTEM**

In this section, we give the derivation of the energy spectrum of the whole system in the single-excitation subspace and the proof that they are exactly the same as the poles in the evolution equation under the Laplace transform in the maintext.

The eigenstate \( \Phi_E \) of the QE-LSP system in the single-excitation subspace can be expanded as \( \Phi_E = \sum_{l=0}^{N-1} c_l \phi_{2l}^{(1)} + \int d^2 \mathbf{r} d\omega \phi_{F}(\mathbf{r}, \omega)|G; \{0_{\omega}\} \). According to the stationary Schrödinger equation \( \hat{H}(\Phi_E) = E(\Phi_E) \) with \( E \) being the eigenenergies, we have

\[\int d^2 \mathbf{r} d\omega \phi_{F}(\mathbf{r}, \omega)|G; \{0_{\omega}\} \]
\[ E_{c_l} = \hbar \omega_0 c_l - i \hbar \int d\omega \int d^3 r' \frac{e^{-2\omega^2}}{\hbar^{\frac{3}{2}}} \sqrt{\pi \varepsilon_0 / \hbar} \text{Im}[\varepsilon_m(\omega)] \mu_{ij}^* G_{ji}(r_l, r', \omega)c_{l}, \] (S17)

\[ E_{c_\omega} = \hbar \omega c_\omega + i \hbar \sum_{j=0}^{N-1} \frac{e^{-2\omega^2}}{\hbar^{\frac{3}{2}}} \sqrt{\pi \varepsilon_0 / \hbar} \text{Im}[\varepsilon_m(\omega)] \mu_{jk} G_{kl}(r_l, r, \omega)c_l, \] (S18)

with \( l, j = 0, \cdots, N-1 \) and \( \hat{i}, \hat{j}, \hat{k} = x, y, z \). Solving \( c_\omega \) and substituting it into Eq. (S17), it is easy to obtain

\[ (E - \hbar \omega_0)c_l - \hbar^2 \sum_{j=0}^{N-1} \int d\omega \frac{J_{ij}(\omega)}{E - \hbar \omega} c_j = 0, \] (S19)

or

\[ (E - \hbar \omega_0)c - \hbar^2 \int \frac{\textbf{J}(\omega)d\omega}{E - \hbar \omega} c = 0, \] (S20)

expressed in a matrix form. Using Jordan decomposition of \( \textbf{J}(\omega) = \textbf{VD}(\omega)\textbf{V}^{-1} \) with \( \textbf{V} \) and \( \textbf{D}(\omega) = \text{diag}[D_0(\omega), \cdots, D_{N-1}(\omega)] \) being its similarity matrix and Jordan canonical form, Eq. (S20) can be expressed as

\[ [E - \hbar \omega_0 - \hbar^2 \int \frac{\textbf{D}(\omega)}{E - \hbar \omega} d\omega] \bar{c} = 0 \] (S21)

where \( \bar{c} = \textbf{V}^{-1}c \). The equations have non-trivial solutions if and only if the determinant of the coefficient matrix is zero. Therefore, the eigenvalues of the QE-LSP system in single-excitation subspace are determined by

\[ E_l = \hbar \omega_0 + \hbar^2 \int \frac{D_l(\omega)}{E_l - \hbar \omega} d\omega. \] (S22)

Equation (S22) takes the same form as the equation to determine the bound state obtained in the maintext. This clearly demonstrates that the dynamics of QEs essentially depends on the energy-spectrum character of the whole QE-LSP system.

**EXACT DYNAMICS FOR \( N = 4 \)**

The spectral density matrix for \( N = 4 \) reads

\[ \textbf{J}(\omega) = \begin{bmatrix} J_0(\omega) & J_1(\omega) & J_2(\omega) & J_1(\omega) \\ J_1(\omega) & J_0(\omega) & J_2(\omega) & J_1(\omega) \\ J_2(\omega) & J_1(\omega) & J_0(\omega) & J_1(\omega) \\ J_1(\omega) & J_2(\omega) & J_1(\omega) & J_0(\omega) \end{bmatrix}, \] (S23)

where the periodic condition \( J_l(\omega) = J_{N-l}(\omega) \) has been used. As a symmetric and circulant matrix, \( \textbf{J}(\omega) = \textbf{VD}(\omega)\textbf{V}^{-1} \), where \( \textbf{D}(\omega) = \text{diag}[D_0(\omega), 2J_1(\omega) + J_2(\omega), J_0(\omega) - J_2(\omega), J_0(\omega) - 2J_1(\omega) + J_2(\omega), J_0(\omega) - J_2(\omega)] \) and \( \textbf{V} = \frac{1}{2} \begin{bmatrix} 1 & 1 & 1 & 1 \\ 1 & i & -1 & -i \\ 1 & -1 & 1 & -1 \\ 1 & -i & -1 & i \end{bmatrix} \) [6]. Note that

\[ \textbf{J}(\omega) = \begin{bmatrix} 1 & 1 & 1 & 1 \\ 1 & i & -1 & -i \\ 1 & -1 & 1 & -1 \\ 1 & -i & -1 & i \end{bmatrix} \]

and \( \textbf{V}(0) = \frac{1}{2} [1, 1, 1, 0]^T \) under the initial condition \( |\Phi(0)\rangle = \beta_0^\dagger |G_l\rangle \), where \( c_1(t) = c_3(t) \) has been used. Equation (S24) is analytically solvable by the Laplace transform. As shown in the maintext, its solution in long-time limit reads

\[ \lim_{t \to \infty} \bar{c}_l(t) = \begin{cases} \langle Z e^{-\gamma t} \rangle \bar{c}_l(0), & y_l(0) < 0 \\ 0, & y_l(0) > 0 \end{cases} \] (S26)

![Figure S1](image-url) (a) Evolution dynamics of \( P(t) \) in different \( r \). (b) Details of the dynamics in long-time. (c) Energy spectrum of the whole system. (d) Long-time values of \( P(t) \) with various values of \( r \). The orange region in (d) covers the values of \( P(\infty) \) that can be taken in its evolution. The parameters are chosen the same as Fig. 2 in the maintext, but with \( N = 4 \).
It clearly shows that the dynamics of the system in the long-time limit is determined by the formation of bound of the whole system. It is not easy to find that the initial-state fidelity equals to \( P(t) = |c_0(t)|^2 \) with 
\[
c_0(t) = \frac{1}{4}[\bar{c}_0(t) + 2\bar{c}_1(t) + \bar{c}_2(t)].
\]

Figure S1(a) plots the evolution of \( P(t) \) in different \( r \). The different behaviors, i.e., complete decay, population trapping, and persistent oscillation, are present depending on the value of \( r \). Details on the long-time behaviors are shown in Fig. S1(b). Such phenomena are associated with the formation of bound state of the QE-LSP system. Figure S1 (c) shows the energy spectrum of the whole system. If no bound state is formed, then \( P(t) \) tends to zero, which characterizes the complete decoherence. If one bound state is formed, then \( P(t) \) tends to a finite value, which describes the population trapping. If two or more bound states are formed, then \( tP(t) \) tends to the Rabi-like persistent oscillations in long-time limit. Such behaviors coincides with our analytical analysis.

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