Quantum Embedded Superstates

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Optical supercavity modes (superstates), i.e., hybrid modes emerging from the strong coupling of two modes of an open cavity, can support ultranarrow lines in their scattering spectra associated with quasi bound states in the continuum (quasi-BIC). These modes are of great interest for sensing applications as they enable compact systems with unprecedented sensitivity. However, classical quasi-BIC sensors are fundamentally limited by the shot-noise limit, which may be overcome in quantum sensors. Here, it is shown that a three-level quantum system (e.g., atom, quantum dot, superconducting qubit) can be tailored to support the quantum analog of superstates with an unboundedly narrow emission line. Remarkably, it is demonstrated that the coupling of such a system with a cavity (e.g., plasmonic or dielectric nanoparticle, microcavity, microwave resonator) enables sensing properties with excellent statistical features. The results can be applied to a plethora of quantum platforms, from superconducting circuits to cold atoms and quantum dots, opening exciting opportunities for quantum sensing and computing.

1. Introduction

Elastic light scattering lies at the heart of the vast majority of sensors and detectors. Examples include tiny plasmonic nanosensors that enable detection down to the single-molecule level[1,2] and massive systems such as LIGO used to detect gravitational waves.[3] Recent studies exploring unusual scattering phenomena, such as parity-time (PT) symmetry, exceptional points, topologically nontrivial phases, have unveiled sensing systems with exceptional properties.[4] Among the most exciting phenomena in this context bound states in the continuum (BICs), also known as embedded eigenstates, offer unique opportunities to tailor the scattering features to extreme levels.[5–10] Von Neumann and Wigner have originally introduced such eigenstates as a mathematical anomaly in quantum mechanics.[11] More recently, this concept has been generalized to different areas of wave physics, including acoustics, hydrodynamics, and photonics.[5,10,12–18] BICs are eigenstates with an unboundedly large Q-factors, and correspondingly vanishing linewidths, corresponding to the coalescence of a scattering pole and zero at the same real frequency.[4] True BICs can arise only in structures either infinite in at least one dimension or employing lossless permittivity (permeability) materials with extreme values.[5] As a result, in realistic structures BICs manifest themselves in the form of a narrow Fano resonance (quasi-BIC) with spectral width limited by the proximity to the ideal requirements.

Among various scenarios supporting quasi-BICs, optical supercavity modes are of particular interest for this work. These modes, according to the terminology generally accepted in the literature,[19,20] emerge when an open cavity (Figure 1a) supports two modes |\psi^1⟩ and |\psi^2⟩ that hybridize into dressed states for a certain coupling strength, usually defined by the cavity geometry and material properties. With a suitable design, one of these states can become dark, giving rise to Friedrich–Wintgen BIC,[21,22] Figure 1b, stemming from destructive interference. This dark supercavity mode manifests itself as a narrow Fano resonance (solid red curve in Figure 1c),[20] which can be of great interest for harmonic generation,[23,24] quantum-entangled photons emission,[25] nanolasers,[26] and sensing, as demonstrated in a series of recent studies.[14–16] These quasi-BIC sensors are of course bound to obey the classical shot-noise limit, hindering some sensor applications of these concepts, and motivating the exploration of analogous topics in the quantum limit for nonclassical states of light and matter. We also stress that BICs are fundamentally different from low-scattering states, such as anapoles. An anapole state is sustained by a non-radiating combination of oscillating polarization currents supported by an external plane wave excitation, whereas the BIC is truly an eigenmode of the system and, as such, does not require external excitation.[27] The key phenomenon underlying the physics of quasi-BIC supercavity modes is the coherent destructive interference of two non-Hermitian cavity modes. In this work, we extend this concept to a quantum system and show that a three-level quantum system (e.g., atom, quantum dot, superconducting qubit) can
Figure 1. Supercavity mode versus quantum embedded superstate. a–c) Illustration of an optical supercavity mode: a dielectric nanocavity supports two strongly coupled nonorthogonal modes $|\psi_1\rangle$ and $|\psi_2\rangle$ (a). Hybridization of the bare modes leads to the formation of two branches of hybrid (dressed) modes. One can become dark (Friedrich–Wintgen BIC), while another becomes bright (b). Such a supercavity quasi bound states in the continuum (quasi-BIC) state manifests itself as a narrow Fano resonance (c). d,e) Illustration of a quantum embedded superstate. d) Sketch of a 3-level V-type quantum system (atom) interacting with a cavity mode, excited by an external monochromatic wave. A wisely tailored atom-field interaction leads to a narrow emission line and ultra-high sensitivity (e).

support the quantum analog of an embedded superstate (QES) with an unboundedly narrow emission line (Figure 1d). This effect requires the simultaneous fulfillment of two conditions: the presence of quantum interference (QI) between multiple atomic transition pathways (between the ground state $|0\rangle$ and excited states $|1\rangle$ and $|2\rangle$, Figure 1d), and strong coupling of the excited states with an external field (freely propagating wave or standing cavity mode). Landau has first underlined the importance of QI in such a system dynamics,[28] and its effects can be introduced in a quantum treatment using generalized damping terms, which are not typically included in semiclassical damping theories. The QI plays a vital role in a plethora of quantum phenomena, including quantum sensing,[29] localization of atoms, spontaneous emission,[30] induced transparency, bistability, and gain without population inversion[31–36] and, as we show in the following, plays a crucial role in our proposed quantum sensors.

We explore QESs using the Lindblad master equation formalism (see Section 4.1), where the presence of QI and the interaction with the reservoir degrees of freedom are rigorously taken into account. First, we show that a QES can be supported by a solitary standing quantum emitter (atom, quantum dot (QD)). The crucial importance of QI on a V-type atom emission in Bloch formalism has been explored in this context.[37] In the following, we demonstrate that the coupling of such a three-level V-type quantum system with an optical cavity (e.g., plasmonic or dielectric nanoparticle) can provide, under suitable conditions, unboundedly narrow emission spectra (Figure 1), and it lifts the requirement for strong excitation, enabling the operation in the low-intensity quantum regime, overcoming the conventional noise limitations of classical sensing systems. A two-level quantum emitter (atom) coupled to a cavity is a popular system in nanophotonics and quantum optics.[38–43] On the other hand, in our work we investigate a three-level atom coupled to a cavity, a system that has been comparatively less explored. Experimentally this class of systems can be realized for instance with a V-type quantum dot embedded in a high-Q cavity, such as a microdisk cavity or a micropillar[44,45] or with superconducting qubits coupled to a detection line. To validate our results, we compare them with Bloch–Redfield equation approach without the secular approximation and rotating wave approximation (RWA) (see Section 4.2).

2. Results and Discussion

2.1. Theoretical Description of a V-Type Atom Interacting with a Cavity

We consider the case of a generalized three-level V-type atom (herein denoted as “atom”), interacting with a resonant cavity, see Figure 1d. To analyze this system, we employ the Lindblad master equation formalism (see Section 4.1 and Supporting Information), which rigorously describes an extensive class of quantum systems (e.g., atoms, quantum dots, and superconducting qubits).[46] The Hamiltonian of such a system reads

$$\hat{H}_S = \hbar \omega_{\text{cav}} \hat{a}^\dagger \hat{a} + \hbar \omega_{10} \hat{\sigma}_+^\dagger \hat{\sigma}_- + \hbar \omega_{20} \hat{\sigma}_+^\dagger \hat{\sigma}_- + \hat{V}$$

(1)

where $\omega_{\text{cav}}$ is the frequency of the cavity mode, $\hat{a}$ and $\hat{a}^\dagger$ are annihilation and creation operators of a quantum in the mode. The
transition frequencies from the ground state $|0\rangle$ to states $|1\rangle$ and $|2\rangle$ are $\omega_{01} = \omega_{02}$, respectively (Figure 1d). The transition operators between the atom states are $\hat{\sigma}^+ = |i\rangle\langle j|$, where $i, j = 0, 1, 2$. The interaction operator $\hat{V}$ accounts for the system interaction with the external electromagnetic field and subsystems with one another, which, using the Jaynes–Cummings model for the cavity-mode interaction and the RWA, \(^{46,61}\) reads

$$\hat{V} = \hbar \sum_\xi \left[ \hat{\Omega}^{\xi}_R (\hat{a}^+ \hat{\sigma}_\xi + \hat{\sigma}_\xi^+ \hat{a}) + \hat{\Omega}_c (\hat{\sigma}_\xi^+ \hat{a}^+ + \hat{a} \hat{\sigma}_\xi) \right] + \hbar \Omega_e (\hat{a}^+ + \hat{a}) \quad (2)$$

The interaction constant of the atom and the cavity is the Rabi constant $\Omega_e = -E_{ext} d_i / \hbar$, where $d_i = i |\sigma| j$ is the matrix element of the dipole moment of the $i \rightarrow j$ transition. The variable $E_{ext}$ is the electric near field of the cavity mode per one quantum, which can be found using the general relation $\frac{1}{\hbar} \int d\Omega \hat{V}_{\text{field}}^2 = \hbar \omega_{cav}$, for a dispersive non-Hermitian system with complex permittivity $\varepsilon$ (see Refs. [47–50]). The constants $\Omega_c = -d_i E_{\text{field}} exp(-i \Omega \varepsilon) + \hbar \Omega_e$ correspond to the atom’s $k$-th transition ($1 \rightarrow 0, 2 \rightarrow 1$ and $2 \rightarrow 0$) and the cavity with the external field $E_{\text{field}} exp(-i \Omega \varepsilon)$, where, for example, $d_{cav} = \sqrt{(3\hbar R^3)/(\varepsilon R_{\text{cav}})}$ for a metallic nanoparticle. Note that the difference between these frequencies is much smaller than the frequencies themselves, $\Delta \omega \ll \omega_{cav}, \omega_0, \omega$, implying that the RWA is accurate to order $\Delta \omega / \omega.$

In the rotating frame (see details in the Methods Section), the system Hamiltonian (1) has the form

$$\hat{H}_{\text{rot}} = \hbar \omega_{cav} \hat{a}^+ \hat{a} + \hbar \bar{\omega}_{10} \hat{\sigma}_{10}^+ \hat{\sigma}_{10} + \hbar \Delta \hat{20} \hat{\sigma}_{20}^+ \hat{\sigma}_{20} + \hbar \Omega_{20}^R (\hat{a}^+ \hat{\sigma}_{10} + \hat{\sigma}_{10}^+ \hat{a})$$

$$+ \hbar \Omega_{10}^R (\hat{a}^+ \hat{\sigma}_{10} + \hat{\sigma}_{10}^+ \hat{a})$$

$$+ \hbar \Omega_{20} (\hat{\sigma}_{20} + \hat{\sigma}_{20}^+)$$

$$+ \hbar \Omega_{10} (\hat{\sigma}_{10} + \hat{\sigma}_{10}^+)$$

(3)

where $\Delta \omega = \omega_{cav} = \bar{\omega}$ is the detuning between the cavity mode frequency and the frequency of the external wave; $\omega_{01} = \omega_{02} = \omega$ is the detuning of the $i$-th dipole transition frequency relative to the external wave frequency. The Hamiltonian (3) is Hermitian, since it describes a closed system. To take into account the relaxation processes, we should introduce the reservoir degrees of freedom, see the Methods Section for details. The system interaction with reservoirs and interaction between subsystems is considered small compared to the cavity mode and dipole transitions frequencies in the atom. After eliminating the reservoir degrees of freedom in Born–Markov approximation,\(^{51}\) we arrive at the local master equation in Lindblad form,\(^{52,53}\) which describes the evolution of the system interacting with thermal baths

$$\frac{d}{dt} \hat{\rho} = -i \left[ \hat{H}_{\text{rot}}, \hat{\rho} \right] + \mathcal{L}(\hat{\rho}) \quad (4)$$

where $\mathcal{L}(\hat{\rho}) = \frac{L(\hat{\rho})}{\hbar} L(\hat{\rho}) + \mathcal{L}(\hat{\rho})$, and $L(\hat{\rho})$ is the Lindblad operator

$$\mathcal{L}(\hat{\rho}) = \sum \left[ \Gamma_i \left( \hat{a}_i^+ \hat{a}_i \hat{\rho} - \hat{\rho} \hat{a}_i^+ \hat{a}_i + \hat{a}_i^+ \hat{a}_i \hat{\rho} + \hat{\rho} \hat{a}_i \hat{a}_i^+ \right) + \frac{\Gamma_i}{2} \left( \hat{a}_i^+ \hat{a}_i \hat{a}_i^+ \hat{a}_i - 2 \hat{a}_i^+ \hat{a}_i \hat{a}_i^+ \hat{a}_i + \hat{a}_i \hat{a}_i^+ \hat{a}_i \hat{a}_i^+ \right) \right]$$

with $\Gamma_i = \frac{4 \Delta_i}{\omega_{cav}^2} (1 + \frac{\Delta_i^2}{\omega_{cav}^2})$, where $\Delta_i = \omega_{cav} - \omega_i$. The Lindblad operator $\mathcal{L}(\hat{\rho})$ is a sum of all possible processes for all cavity modes $\hat{a}_i$, $\hat{a}_i^+$.

2.2. Quantum Embedded Superstate of a Solitary Atom

We start by analyzing a solitary three-level V-type quantum system excited by an external monochromatic field. We obtain its Hamiltonian by excluding the cavity terms from Equation (3): $\hat{H}_{\text{rot}} = \hbar \omega_{01} \hat{\sigma}_{10}^+ \hat{\sigma}_{10} + \hbar \Delta \hat{20} \hat{\sigma}_{20}^+ \hat{\sigma}_{20} + \hbar \Omega_{10} \hat{\sigma}_{10}^+ \hat{\sigma}_{10} + \hbar \Omega_{20} \hat{\sigma}_{20} + \hat{\sigma}_{20}^+ \hat{\sigma}_{20}$.

In the symmetric case, when the decay rates in the atom are equal and the frequency of the external field precisely falls in the center between frequencies of the dipole transitions $\gamma_1 = \gamma_2 = \gamma$, and $(\omega_{10} + \omega_{02}) / 2 = \omega_0$, the fluorescence can be quenched if the detunings meet the condition $\Delta \gamma \Omega_0^R + \Delta \omega \Omega_0^R = 0$ and the dipole moments of the atom are parallel, $\Gamma = \sqrt{\Delta \Omega_0^R / \gamma}.$

The fluorescence spectrum consists of coherent and incoherent parts. The coherent Rayleigh part emerges due to elastic scattering of the driving field. However, in our analysis we focus on the incoherent part of the fluorescence spectrum. The incoherent part can be calculated as (see Supporting Information)

$$S(\omega) = \mathcal{R} \int_0^\infty \frac{d\tau}{\tau} \text{lim}_{t \to \infty} |\Delta \hat{D}(t + \tau) \Delta \hat{D}(t)| e^{-i\omega \tau} d\tau \quad (5)$$

where $\Delta \hat{D}(t) = d_{10} \hat{\sigma}_{10}^+ + d_{20} \hat{\sigma}_{20}^+ + d_{02} \hat{\sigma}_{20}$ is the dipole polarization operator of the atom and the cavity mode. We find the two-time averages $\langle \Delta \hat{D}(t + \tau) \Delta \hat{D}(t) \rangle$ using the quantum regression formula, where $\Delta \hat{D}(t) = \bar{D}(t) - \langle \bar{D}(t) \rangle$, i.e., the difference of the dipole polarization operator and its averaged stationary value.

We simulated the fluorescence spectrum of a solitary atom for different values of $\Gamma$ in Figure 2a. Both the amplitude and width of the spectral line do not change significantly until $\Gamma / \gamma \approx 0.98$, when the dipole moments of the $1 \leftrightarrow 0$ and $2 \leftrightarrow 0$ transitions
Figure 2. Solitary three-level V-type atom in the external field. a) Amplitude of the central peak of the resonance fluorescence spectrum of the atom as a function of $\Gamma/\gamma$ (red curve); full width at half maximum of the central peak versus $\Gamma/\gamma$ (blue curve). b–d) Fluorescence spectra for $\Gamma/\gamma = 0.92$ (b); $\Gamma/\gamma = 0.998$ (c), and $\Gamma/\gamma = 1$ (quantum embedded superstate, QES) (d). The other parameters: $\Omega = 5\gamma$, $\delta = \gamma/2$.

2.3. Embedded Superstate in an Atom-Cavity System

It is important to note that this suppression is possible when the external field amplitude is very large, i.e., $\Omega \gg \gamma, \delta$. Although these fields are achievable in experiments, they are not convenient for the practical implementation of this effect. To overcome the limitation of large pumping fields in the solitary atom case, we place the atom near a resonant cavity. We study the variation of resonant fluorescence spectrum as a function of the cavity frequency and demonstrate very high sensitivity at low pump power under the condition that the dipole moments $d_{10}$ and $d_{20}$ are parallel.

The cavity mode is excited by an external optical wave, which is not in resonance with the cavity. We suppose that the pumping is small, such that the interaction constant between the external field and the mode is much less than the decay rates in the atom, $\Omega_i \ll \gamma_1, \gamma_2$, and is of the same order as the decay rate of the mode, $\Omega_i \sim \gamma_c$. Quenching follows $\delta_1(\Omega_{10}^2 + \Omega_{20}^2)^2 = 0$, and we assume strong coupling regime between cavity and atom, $\Omega_i^2 > \gamma_1, \gamma_2$ and equal spontaneous decay rates of the dipole transitions, $\gamma_1 = \gamma_2 = \gamma$.

Figure 4 shows the fluorescence spectra in two limiting scenarios, $\Gamma = 0$ and $\Gamma = \gamma$, i.e., without (a) and with (b) quantum coherence. In Figure 4a, the spectrum weakly depends on $\delta_{\text{cav}} = \omega_{\text{cav}} - \omega$ and almost does not change in the chosen frequency range for $\omega_f$, which is much smaller than $\Omega_i^2$. The spectra profiles on the cross-sections along corresponding dashed lines are presented by blue curves in Figure 4c,d. At $\omega_{\text{cav}} = \omega$, the spectrum without QI has the form of a triplet with side peaks at $\pm \sqrt{\delta^2 + 2\Omega_i^2}$.

In remarkable contrast to the non-QI case, the QI scenario demonstrates an extremely narrow spectral line (Figure 4b), revealing a strong dip at the resonance of the cavity mode and the external field. Although the dip in the center of Figure 4b is profound, there is no total suppression of fluorescence, as in the case of a solitary atom (Figure 2). This behavior is due to the contribution of the cavity to the emission and the finite dephasing rates.
Figure 4. Fluorescence spectra with and without quantum interference (QI). a) Spectrum of resonance fluorescence without QI, $\Gamma = 0$, in a plane of fluorescence frequency in the rotating frame, $\omega_f$, and the detuning between the frequency of the cavity mode and the external wave frequency, $\delta_{cav} = \omega_{cav} - \omega$. b) Spectrum with QI, $\Gamma = \gamma$. c,d) Spectra profiles on the cross-sections along corresponding dashed lines. The other parameters: $\gamma_a/\gamma = 10^{-2}$, $|\omega_{20} - \omega_{10}| = 2\gamma$, $\Omega_{10} = \Omega_{20} = \Omega = 3\gamma$, $\Omega_a/\gamma = 2 \times 10^{-2}$.

Out of resonance, the spectrum sharpens and its amplitude increases [Figure 4d, red curve]. One of the side peaks disappears, whereas the second peak starts degenerating when $\omega_{cav}$ moves away from the resonance. Note that in this QI scenario, i.e., $\Gamma = \gamma$, the eigenfrequencies of the Liouvillian superoperator with corresponding real parts approach the real axis (see Figure S2, Supporting Information). Their behavior is similar to the case of the atom without a cavity (Figure 3).

2.4. Quantum Embedded Superstate for Sensing Applications

We have shown how the QI significantly modifies the fluorescence spectra, sharpening them by three orders of magnitude. This effect significantly enhances the sensitivity to the cavity properties, ideally suited for quantum sensing applications. Here, we analyze the change in the fluorescence spectrum due to variations of the cavity frequency $\omega_{cav}$. This variation can be caused, for example, by a small permittivity change in the cavity.16,SS5–SS8 Accordingly, we define the sensitivity of our QES-based system as the figure of merit (FOM)

$$FOM = \frac{\omega}{\max(S)} \left| \frac{\partial S}{\partial \omega_{cav}} \right|$$

(6)

where $S$ is the fluorescence spectrum amplitude. Figure 5 shows the calculated FOM of our system for the chosen parameters. We can see that the FOM is maximized near the spectrum peaks, as expected. At zero fluorescence frequency in the rotating frame, $\omega_f$, the spectrum profile has two distinguishable peaks (Figure 5a). These peaks relate to the splitting of the mode levels due to the interaction with the external field. The spectral line here is very sharp, and it provides an excellent platform for sensor applications. The FOM reaches $\approx 5000$ along this cross-section. The
intensity of the external wave can be comparatively small, which allows using this approach in low-intensity quantum applications. Figure 5b studies the influence on the FOM of atom dipole moments not being exactly parallel. Although the maximum sensitivity can be achieved at the parallel condition, satisfactory sensitivity with FOM ≈100 is achievable up to Γ = 0.99γ. Thus, the effect of spectral line narrowing is pronounced only when the QI conditions are met. However, it stays robust for slight variations of the interference parameter.

Finally, we consider the fluorescent emission’s statistical properties in the parameter space formed by the frequency detuning between cavity mode and external wave frequency δ_cav and the collective dissipation rate Γ. Although the average number of quanta in the entire system is one, radiation’s resulting statistics can strongly differ from the single-photon regime. We study the second-order coherence function at zero time for the system radiation. As long as the dipole moment of the cavity mode is much larger than the dipole moments of the atom transitions, we can assume that almost the entire emission of the system originates from the mode. Thus, the coherence function has the form g^{(2)}(0) = ⟨â^+ââ⟩/⟨â^+â⟩^2 (see refs. [46,59]). The deviation of the correlation function g^{(2)}(0) from 1 gives the noise level in the system. Below, we show that QI in our system brings the statistical properties of the interacting atom and cavity close to the properties of a solitary cavity mode, i.e., g^{(2)}(0) tends to 2.

Figure 6 shows the dependence of the second-order coherence function g^{(2)}(0) on δ_cav and Γ. One can see substantial bunching of emitting photons with g^{(2)}(0) ≫ 10 near resonance in the region of nonparallel dipole moments. Remarkably, the coherence function decreases considerably when approaching the condition of full interference (Γ/γ = 1), see the zoomed area around Γ/γ ≈ 1 in Figure 6b. The coherence function has a two-peak shape as we vary the detuning. These peaks are connected with the frequency difference between the atom transitions. Strictly at resonance near the full interference, g^{(2)}(0) has a significant dip with a value of 2.4, see Figure 6d. When moving away from resonance, g^{(2)}(0) tends to 1 as the detuning grows. Nevertheless, in the full interference case, the value of g^{(2)}(0) demonstrates the fastest
convergence to 1. Thus, the system formed by a three-level atom strongly interacting with the cavity mode manifests bunching behavior in the whole parameter space in which the sensor operates. At resonance and for \( \Gamma/\gamma \approx 1 \), the \( g^{(2)}(0) \) function takes its minimum value \( \approx 2 \), corresponding to the case in which either the first or the second dipole transition delivers a photon to the cavity mode, which then reradiates the photons with its usual statistics close to thermal emission \( g^{(2)}(0) = 2 \). This result shows that QI and trapping in quantum embedded superstates significantly reduce the noise in the system, beyond the limits of classical sensors.

It is used to define the performance of various sensors via their sensitivity \( S_s \), that is a spectral shift of a resonant line in nm caused by a small change of some parameter. For example, in the refractive index (RI) sensors, the parameter is the RI and the sensitivity is defined per unit RI (RIU), \( S_s \) [nm RIU\(^{-1}\)]\(^{[60,61]} \). However, if the resonant line of a sensor is wide (small Q-factor), then even a relatively large shift of the line can be undetectable and hence it is more convenient to normalize \( S_s \) by the line spectral width, \( \Delta \lambda \) [nm]. The resulting dimensionless value is called a FOM ranging from units \(^{[62-64]} \) up to several hundred \(^{[65]} \) for the state-of-the-art photonic sensors. In our case, FOM [Equation (6)] is also a dimensionless quantity defined as the ratio of the change of the emission intensity over the causing change of the normalized parameter (the cavity resonant frequency). Our FOM reaches \( 5 \times 10^3 \) in the optimized operation regime. However, we note that it is not fair to compare these two FOMs directly because they characterize a different system response, namely resonant pick shift (classical definition) and change of the emission intensity (in our work). Nevertheless, such an enormous value of FOM of our system allows judging on the extremely high sensitivity of our system.

3. Conclusions

In this work, using the Lindblad formalism we have extended to a quantum scenario the concept of embedded superstates based on a three-level quantum system, e.g., atom, quantum dot,
and superconducting qubit. The critical phenomenon beyond the demonstrated quantum supercavity modes is the coherent destructive interference of two quantum states. These unique quantum states are demonstrated to support unboundedly narrow emission lines with significantly improved second-order photon emission statistics. We have also shown that the coupling of this tailored three-level atom with an optical cavity (e.g., plasmonic or dielectric nanoparticle) paves the way to quantum sensing with remarkable performance. Namely, the proposed system provides a figure of merit of 5 × 10^4 in the low-quantum regime. The effect of QI and trapping in the quantum embedded superstate significantly reduces the system noise.

Our concept requires high-quality cavities, \( r_s \leq r \), in order to significantly narrow the fluorescent line. One of the possible systems where it can be implemented is a V-type quantum dot in a microdisk cavity or in a micropillar cavity, where the Q factors exceed 10^8.\(^{[16,17]}\) One can also use superconducting qubits coupled to a detection line.\(^{[46]}\) Our work unveils quantum embedded superstates, a novel state of quantum light-matter interaction, and suggests their use in advanced quantum-enhanced sensors with superior noise performance.

4. Methods

4.1. Lindblad Master Equation Approach

The interacting atom and the cavity mode system is an open quantum system, which interacts with reservoirs. It was assumed that the system is in a low-temperature environment and interacts only with the reservoirs of the EM modes of the free space. Thus, the case was considered when the system’s energy dissipates only through the radiation into free space. The Hamiltonian of the EM modes of free space and the interaction Hamiltonian of the system with the reservoir has the form:

\[
\hat{H}_R + \hat{H}_{SR} = \sum_{k,i} \hbar \omega \hat{a}_{k,i}^{\dagger} \hat{a}_{k,i} + \sum_{k,i} \hbar \omega \hat{b}_{k,i}^{\dagger} \hat{b}_{k,i} - \sum_{k,i} \sqrt{\frac{2\pi \hbar \omega}{V}} e_k \hat{\Delta}_k \left( \hat{a}_{k,i} + \hat{a}_{k,i}^{\dagger} \right) - \sum_{k,i} \sqrt{\frac{2\pi \hbar \omega}{V}} e_k \hat{\Delta}_{k,uv} \left( \hat{b}_{k,i} + \hat{b}_{k,i}^{\dagger} \right)
\]  

where \( i = 10, 20 \). Note that two uncorrelated reservoirs for the atom and the cavity were considered. The system Hamiltonian \( \hat{H}(\tau) \) depends on time. Therefore, to get rid of this time dependence, the unitary transformation \( \hat{U}(\tau) = \exp\left[ -\frac{i}{\hbar} \hat{H}_0 \tau \right] \) (\( \omega \) is the frequency of the external field) of the Hamiltonian \( \hat{H}_R + \hat{H}_S + \hat{H}_{SR} \) should be performed and moved to the rotating frame:

\[
\hat{H}_S = \hat{U}^\dagger \hat{H}_R \hat{U} - i \frac{\partial \hat{U}}{\partial \tau}
\]

\[
= \hbar \delta_{\text{cav}} \hat{a}^{\dagger} \hat{a} + \hbar \delta_{\text{SR}} \hat{a}^{\dagger} \hat{a}_{10} + \hbar \delta_{\text{20}} \hat{a}^{\dagger} \hat{a}_{20} + \hbar \Omega_x^{(2)} \left( \hat{a}^{\dagger} \hat{a}_{10} + \hat{a}_{10}^{\dagger} \hat{a} \right) + \hbar \Omega_x^{(2)} \left( \hat{a}^{\dagger} \hat{a}_{20} + \hat{a}_{20}^{\dagger} \hat{a} \right) + \hbar \Omega_x^{(2)} \left( \hat{a}^{\dagger} \hat{a}_{10} + \hat{a}_{10}^{\dagger} \hat{a} \right)
\]

The expressions for decay rates see, e.g., in Ref.\(^{[68]}\) In Equations (M5) and (M6), it was assumed that \( T \ll \delta_{\text{cav}}, \delta_{10}, \delta_{20} \), such that \( \eta(\omega) \ll 1 \). Note that the last two terms are responsible for the QI between 1 ↔ 0 and 2 ↔ 0 dipole transitions. It should be noted that our approach only works within the Born–Markov approximation, which is based on perturbation theory with respect to the system–reservoir coupling and assumes short environmental correlation times, and does not work at ultrastong coupling regime, when Rabi constants are comparable with the transition frequencies in the system, \( \Omega \sim \omega_0 \). In the first case, one should consider non-Markovian dynamics of the system + reservoir\(^{[49]}\) in the second case, one should use the global master equation by decomposing the system’s operators on the eigenbasis of system’s Hamiltonian.\(^{[70,71]}\)

4.2. Bloch–Redfield Approach

Of course, the approach used in the main text of the manuscript could not be considered absolutely complete and accurate, and
the applicability of this local Lindblad master equation was discussed. However, in this section, Bloch–Redfield equation is derived with non-secular terms and the RWA is not used. It is shown that the main results remain the same.

In this section, the obtained results of the narrow spectral line at the regime of QI are verified using the rigorous approach of Bloch–Redfield equation. The Hamiltonian from Equations (1) and (2) for the system and Hamiltonian (M1) for reservoirs and the interaction between the system and reservoirs are considered. Here RWA is not used. The Hamiltonian of system–reservoir interaction is written in the form of Hamiltonian (M1) for reservoirs and the interaction between the system and reservoirs is in a thermal equilibrium. Further, the Fourier transform of system–reservoir interaction is written in the form of Hamiltonian (M1) for reservoirs and the interaction between the system and reservoirs is discussed. However, in this section, Bloch–Redfield equation is derived with non-secular terms and the RWA is not used. It is shown that the main results remain the same.

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Figure 7. Spectra profiles in a case of quantum interference (QI): a) at resonance, \( \delta_{\text{cav}} = 0 \), b) \( \delta_{\text{cav}} = 5 \times 10^{-4} \omega \). The other parameters: \( \gamma_a / \gamma = 10^{-2}, |\omega_{20} - \omega_{10}| = 2 \gamma, \Omega_R^{20} = \Omega_R = 3 \gamma, \Omega_a / \gamma = 2 \times 10^{-2}, T = 300 \text{ K} \).

The standard procedure of excluding the reservoir variables, Born–Markov approximation is followed (in the previous sections, the limitations of this approach were discussed), and finally Bloch–Redfield master equation is obtained for the system’s density matrix written in the eigenbasis of the system’s Hamiltonian, \( \hat{H}_S \) in the matrix form: \( [67,72] \)

\[
\frac{\partial}{\partial t} \hat{\rho}_S(t) = -i \omega_{j\beta} \hat{\rho}_S(t) - \hbar^{-2} \sum_{a,b} \sum_{m,n} \int_0^\infty d\tau \left\{ F_{ab}(\tau) \right. \\
\left. \times \left[ \delta_{nm} \sum_n S_{jn}^{\alpha} S_{nk}^{\beta} e^{i\omega_{jn} \tau} - S_{jn}^{\alpha} S_{nk}^{\beta} e^{i\omega_{jn} \tau} \right] \\
+ F_{ab}(-\tau) \left[ \delta_{jn} \sum_n S_{mn}^{\alpha} S_{nk}^{\beta} e^{i\omega_{mn} \tau} - S_{mn}^{\alpha} S_{nk}^{\beta} e^{i\omega_{mn} \tau} \right] \right\} \hat{\rho}_S(t)
\]

(M7)

where \( S_{mn}^{\alpha} = \langle n | \hat{S}_m^{\alpha} | m \rangle \), \( \omega_{mn} = \omega_m - \omega_n \), and \( \hat{H}_S | m \rangle = \hbar \omega_m | m \rangle \).

The results for the spectra profiles in a case of QI: a) at resonance, \( \delta_{\text{cav}} = 0 \), b) \( \delta_{\text{cav}} = 5 \times 10^{-4} \omega \) are presented in Figure 7. The behavior of the fluorescence spectrum remains the same as in Figure 4c,d and is quantitatively close to the results obtained using the local Lindblad approach. Thus, our assumptions made for the derivation of the local Lindblad master equation are reasonable and this approach can be used in the system and parameter range considered.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.
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