Research article

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Quantum exceptional chamber induced by large nondipole effect of a quantum dot coupled to a nano-plasmonic resonator

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Abstract: Exceptional points (EPs) are the singularities of a non-Hermitian system where the eigenenergies and eigenstates simultaneously coalesce, a topological property that gives rise to a plethora of exotic phenomena. Probing the EPs and associated effects requires the system to go through the EPs. However, the ultrahigh sensitivity of an isolated EP to the external disturbances makes accessing the EPs difficult. To overcome this limit, many approaches have been presented to form the exceptional line/ring and surface. Here, we demonstrate that a quantum exceptional chamber, which is a three-dimensional collection of the EPs, can be constructed in the coupled plasmon-quantum dot (QD) systems by the nondipole effect of the QD. For an asymmetric QD adjacent to a plasmonic nanoparticle, it is found that the contributions of multipole transitions to the coupling strength can be larger than that of dipole transition. The orientation-dependent quantum interference between the dipole and multipole transitions can lead to controllable switch between the weak and strong coupling, and provides an extra degree of freedom to form a high-dimension EP space. Our approach provides a

robust platform for accessing the quantum EPs and related applications.

Keywords: exceptional point; nondipole effect; plasmon–exciton interaction.

1 Introduction

A realistic physical system with an energy-nonconserving process, such as the energy exchange with environments, can be described by an effective non-Hermitian Hamiltonian. An intriguing property of non-Hermitian Hamiltonians is the existence of singularities in Hilbert space that are called as exceptional points (EPs), where the eigenenergies and corresponding eigenstates simultaneously coalesce. At these degenerate points, the state space of system loses one or more dimensions [1–7]. This exotic property gives rise to a plethora of interesting phenomena and important functionalities in classical domain, including unidirectional transmission and reflection [8], loss-induced revival of lasing [9], single-mode laser [10] and ultrasensitive sensing [11–15]. Although the isolated EPs allow researchers to investigate certain important aspects of non-Hermitian systems and gain insight into their behaviors, they are very sensitive to the external disturbances, such as the unavoidable fabrication errors or the experimental uncertainty. This hinders the access to the EPs and the observation of the related phenomena. Extending the isolated EPs to higher dimension is beneficial to access the EPs and related applications [5, 14]. Furthermore, high-dimension EP enables intriguing physical phenomena [14, 16–18], but only exceptional line and surface have been reported.

The EPs also exist in the coupled quantum systems [19–27], and we called them as the quantum EPs to distinguish from the conventional EPs in the above-mentioned systems of classical wave. The research of the quantum EPs and corresponding applications is an emerging field, demonstrating the potential in developing advanced quantum devices [26, 27]. For two coupled quantum systems, the

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quantum EPs are the critical points between the weak and strong coupling. While their associated applications, such as the ultrasensitive quantum sensing, are based upon measuring the change of the eigenenergy splitting around the EPs. This requires the coupled system to be operated in the strong-coupling regime. Furthermore, to experimentally identify the quantum EPs, the system needs to switch between the strong and weak coupling to encircle the EPs [28–30]. However, it is difficult to dynamically fine tune the coupling strength in the solid-state cavity-QED systems. Therefore, it is of practical importance to find a flexible approach for effectively manipulating the coupling between two quantum systems.

Among various nanophotonic structures, the metallic nanoparticles that are able to confine the electromagnetic fields down to a few nm³ have been demonstrated as a good platform for the strong light-matter interaction [31]. Strong coupling between plasmon modes and single molecular aggregates [32, 33] or QDs [34–36] has been achieved at room temperature. However, it is still hard to achieve the strong plasmon-QD coupling in the near-infrared wave band due to the large plasmon decaying and low photon energy. Compared to atomic and molecular emitters, a unique feature of mesoscopic QDs is the spatially extended exciton wave function [37]. Due to the imperfect shape or the structural inhomogeneities of QDs, their exciton wave functions are generally asymmetrical, resulting in non-vanishing multipole transition moments [37, 38]. Together with the rapid varying of electromagnetic field inside the QDs, the multipole transitions can break down the usual dipole approximation (DA). The resulting nondipole effect has been experimentally observed even when the QD is in tens of nanometers away from the plasmonic structures, and there is no counterpart of similar phenomenon in atom physics [39–41]. Although many works reveal the significant nondipole effect in weak light-matter interaction, there are rare reports in the strong-coupling regime except for the recent works that studied this effect of single QD in a photonic crystal dielectric cavity [42], as well as single molecule in the plasmonic cavities with subnanometric gap [43–45].

Here, we report the nondipole effect in a strongly coupled plasmon-QD system and apply it to construct a high-dimension collection of the quantum EPs. We show that by rotating the QD, the quantum interference between the dipole and multipole transitions can be flexibly manipulated. This provides an effective method of tuning the coupling strength between the QD and plasmon to access the quantum EPs. Furthermore, the nondipole effect as an extra degree of freedom can extend an isolated

EP to the exceptional lines, surfaces and even a chamber, offering a robust platform for accessing the quantum EPs and their applications.

In this work, we consider the self-assembled InAs/GaAs QDs that own extended exciton wave function [37]. The broken parity symmetry of their electron and hole wave functions along the growth direction results in non-vanishing multipole moments [38]. Therefore, such self-assembled QDs have a prominent nondipole effect, and can be characterized by the usual dipole transition and a set of additional high-order transitions with suitable truncation.

2 Theoretical model of nondipole effect

Different from the dipole transition that is dependent on the electric field intensity, the high-order transitions are coupled to the corresponding order of electric field gradient. Therefore, we expand the electric field around the center-of-mass $\mathbf{r}_0$ of QD, $\hat{E}_i(\mathbf{r}, \phi) = \hat{E}_i(\mathbf{r}_0) + r_j(\phi) \times \partial_j \hat{E}_i(\mathbf{r})|_{\mathbf{r}=\mathbf{r}_0} + \frac{1}{2} r_k(\phi) r_j(\phi) \times \partial_k \partial_j \hat{E}_i(\mathbf{r})|_{\mathbf{r}=\mathbf{r}_0} + \dots$, where the electric field operator is expressed via the classical Green's function $\hat{\mathbf{E}}(\mathbf{r}) = i \sqrt{\frac{\hbar}{\pi \epsilon_0}} \int_0^\infty d\omega \int d^3 \mathbf{r}' \frac{\omega^2}{c^2} \sqrt{\text{Im}[\epsilon(\mathbf{r}', \omega)]} \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) \hat{f}(\mathbf{r}', \omega)$ [46]. $\hat{f}(\mathbf{r}', \omega)$ and $\mathbf{G}(\mathbf{r}, \mathbf{r}', \omega)$ are the annihilation operators of the quantized electromagnetic field and the dyadic Green's tensor, respectively. $r_q = (\mathbf{r} - \mathbf{r}_0)_q$, where $q = x, y, z$ stands for the coordinate components. ϕ represents the rotation angle of QD along its dipole moment, as illustrated in Figure 1(b). With operator $\hat{E}_i(\mathbf{r}, \phi)$, the interaction Hamiltonian of plasmon–exciton composite reads [47]

$$H_{\text{int}} = - \sum_{i,j,k} \sigma_- \left(\mu_i \hat{E}_i^\dagger(\mathbf{r}_0) + \Lambda_{ji}(\phi) \partial_j \hat{E}_i^\dagger(\mathbf{r}_0) + \Omega_{kji}(\phi) \partial_k \partial_j \hat{E}_i^\dagger(\mathbf{r}_0) \right) + \text{H.c.} \quad (1)$$

where μ_i , $\Lambda_{ji}(\phi)$ and $\Omega_{kji}(\phi)$ are the zero-to second-order mesoscopic moments of QD. In Ref. [47], we show that the contributions of magnetic multipoles are much smaller than that of the corresponding electric multipoles, and thus in the following study we call μ_i , $\Lambda_{ji}(\phi)$ and $\Omega_{kji}(\phi)$ as the equivalent electric dipole, quadrupole and octupole transition moments, respectively. The magnitude of multipole moments under QD rotation can be found by applying the unitary transformation $\hat{U}_{r_x}(\phi) = e^{-(i/\hbar) \hat{L}_{r_x} \phi}$. With the interaction Hamiltonian (1), we can obtain the local coupling strength (LCS) up to the second-order expansion of $\mathbf{G}(\mathbf{r}_0, \mathbf{r}_0, \omega)$, which governs the light-matter interaction

Figure 1: Physical models of the coupled plasmon-QD system and the QD. (a) Schematic diagram of an InA/GaA QD coupled to a silver nanoshell. The QD has spatially extended electron and hole wave functions. (b) QD rotation along the dipole orientation (x axis). The cross section of QD is trapezoid, with height h and radius R . (c) Vector map and electric field intensity distribution for plasmonic field generated by an x -oriented dipole source (blue dot and arrow) located 2 nm from nanoshell.

and QD dynamics [47]

$$\Gamma(\mathbf{r}_0, \phi) = \Gamma^{(d)}(\mathbf{r}_0) + \Gamma^{(Q)}(\mathbf{r}_0, \phi) + \Gamma^{(dQ)}(\mathbf{r}_0, \phi) + \Gamma^{(dO)}(\mathbf{r}_0, \phi) \quad (2)$$

with

$$\Gamma^{(d)}(\mathbf{r}_0) = A|\mu|^2 \text{Im} G_{xx}(\mathbf{r}_0, \mathbf{r}_0, \omega) \quad (3)$$

$$\Gamma^{(Q)}(\mathbf{r}_0, \phi) = -A \cos^2(\phi) |\Lambda_{xz}|^2 \partial_x \partial_x \text{Im} G_{zz}(\mathbf{r}_0, \mathbf{r}_0, \omega) \quad (4)$$

$$\Gamma^{(dQ)}(\mathbf{r}_0, \phi) = 2A \text{Re} [\cos(\phi) \Lambda_{xz} \mu_x^*] \partial_x \text{Im} G_{zx}(\mathbf{r}_0, \mathbf{r}_0, \omega) \quad (5)$$

$$\begin{aligned} \Gamma^{(dO)}(\mathbf{r}_0, \phi) = & A \{ 2 \text{Re} [\cos^2(\phi) \Omega_{xzz} \mu_x^*] \partial_x \partial_z \\ & \times \text{Im} G_{zx}(\mathbf{r}_0, \mathbf{r}_0, \omega) + \text{Re} [\Omega_{xxx} \mu_x^*] \partial_x \partial_x \text{Im} G_{xx}(\mathbf{r}_0, \mathbf{r}_0, \omega) \} \\ & + \{ \text{Re} [(\cos^2(\phi) \Omega_{yyx} + \sin^2(\phi) \Omega_{zzx}) \mu_x^*] \partial_y \partial_y \\ & + \text{Re} [(\cos^2(\phi) \Omega_{zzx} + \sin^2(\phi) \Omega_{yyx}) \mu_x^*] \partial_z \partial_z \} \\ & \times \text{Im} G_{xx}(\mathbf{r}_0, \mathbf{r}_0, \omega) \end{aligned} \quad (6)$$

where $A = 2\omega^2 / \hbar \epsilon_0 c^2$. $\Gamma^{(d)}(\mathbf{r}_0)$ and $\Gamma^{(Q)}(\mathbf{r}_0, \phi)$ are the parts of the LCS contributed by the dipole and the quadrupole, while $\Gamma^{(dQ)}(\mathbf{r}_0, \phi)$ and $\Gamma^{(dO)}(\mathbf{r}_0, \phi)$ are the contributions of dipole-quadrupole and dipole-octupole transition interferences, respectively. We can see that the QD rotation is trivial for a point-like dipole emitter, while leads to the orientation-dependent LCS in appearance of the nondipole effect. Therefore, the nondipole effect is an extra degree-of-freedom that can tune the plasmon-QD interaction.

To demonstrate the manipulation of strong coupling by the nondipole effect, we couple the QD to a silver nanoshell with external radius $r_1 = 15$ nm and thickness $t = 1$ nm, as shown in Figure 1(a). The dielectric permittivity of silver is taken from Ref. [48], and the core of nanoshell is chosen as GaAs with refractive index $n_c = 3.4$. QD transition energy is taken as $\hbar\omega_0 = 1.213$ eV, with a dipole moment of $\mu_x = 60$ D ($1.24e$ nm) [37]. The hybrid system is embedded in homogeneous GaAs background. With the aforementioned parameters, one LSPR of silver nanoshell matches with the QD transition energy [47]. The tightly confined electromagnetic field near the metallic interface produces large electric field gradient around the QD in the XZ plane, as Figure 1(c) displays.

Figure 2(a) and (b) show how the nondipole effect modifies the LCS as the QD-nanoshell separation s varies from 2 to 12 nm for $\phi = 0$ and π , respectively. For the range of parameters (s, R) we choose, the LCS shows an approximately five-fold enhancement at (2 nm, 10 nm) for $\phi = 0$. While for the opposite QD orientation, the suppression of the LCS is observed because $\Gamma^{(dQ)}(\mathbf{r}_0, \phi)$ changes its sign (see the red lines in Figure 2(c) and (d)). Compared to previous works of the nondipole effect in weak coupling, two remarkable phenomena appear as the QD-nanoshell separation is down to a few nanometers and enters the strong-coupling regime. First, with a fixed s , the LCS is monotonically increasing with larger QD radius for $\phi = 0$; however, there is a non-monotonic change of LCS as the QD radius increases from 4 to 10 nm for $\phi = \pi$, as

Figure 2: Modification of the spontaneous emission by the nondipole effect. (a) and (b) show the enhancement of LCS $\Gamma(\mathbf{r}_0, \phi) / \Gamma^{(d)}(\mathbf{r}_0)$ as a function of QD radius R and distance s for $\phi = 0$ and π , respectively. The dashed gray line in (b) labels the minimal $\Gamma(\mathbf{r}_0) / \Gamma^{(d)}(\mathbf{r}_0, \phi)$ for a fixed s . (c) and (d) show $\Gamma(\mathbf{r}_0, \phi) / \Gamma^{(d)}(\mathbf{r}_0)$ and the decomposed contribution of $\Gamma^{(Q)}(\mathbf{r}_0, \phi)$, $\Gamma^{(dQ)}(\mathbf{r}_0, \phi)$ and $\Gamma^{(dO)}(\mathbf{r}_0, \phi)$ versus QD radius for $s = 2$ nm (c) and 4 nm, respectively. The pink circle labels the minimal $\Gamma(\mathbf{r}_0, \phi) / \Gamma^{(d)}(\mathbf{r}_0)$ for $\phi = \pi$. The annotation $(\mathbf{r}_0, \phi)$ in all figures are dropped for simplicity. $\Gamma^{(d)}(\mathbf{r}_0)$ is evaluated as 32.42 meV (0.13 ns) and 5.47 meV (0.76 ns) for $s = 2$ and 4 nm, respectively, and decreases linearly from $s = 2$ to 8 nm.

Figure 2(b) shows. We can see that the LCS has a minimum at around $R = 8$ nm, while it increases as the QD radius enlarges. Second, according to the trend of LCS versus R , we can expect that for a large QD with $R > 20$ nm, the nondipole effect will not suppress the LCS even for $\phi = \pi$.

Figure 2(c) and (d) plot the LCS and its multipole decomposition as a function of QD radius for $s = 2$ nm and $s = 4$ nm, respectively, normalized by $\Gamma^{(d)}(\mathbf{r}_0)$. We can see that the non-monotonic change of LCS shown in Figure 2(b) results from the quantum interference between the dipole and multipole transitions. The transition interferences $\Gamma^{(dQ)}(\mathbf{r}_0, \phi)$ and $\Gamma^{(dO)}(\mathbf{r}_0, \phi)$ dominate over the quadrupole transitions $\Gamma^{(Q)}(\mathbf{r}_0, \phi)$ for small QD radius R , but the latter grows faster when $R > 7$ nm. As a result, the LCS shows a non-monotonic change for $\phi = \pi$. The reason

why $\Gamma^{(Q)}(\mathbf{r}_0, \phi)$ drastically increases can be found from (4)–(6). We can see that both $\Gamma^{(Q)}(\mathbf{r}_0, \phi)$ and $\Gamma^{(dQ)}(\mathbf{r}_0, \phi)$ are related to the quadrupole transition moment Λ_{xz} , which is induced by the strain effect and scales with QD radius. However, $\Gamma^{(Q)}(\mathbf{r}_0, \phi)$ is square to Λ_{xz} and increases quadratically compared to $\Gamma^{(dQ)}(\mathbf{r}_0, \phi)$. On the other hand, according to the selection rules, the octupole moments exist if the dipole transition is allowed, but these octupole moments do not have the same relation with QD radius and height as those induced by the strain effect (Λ_{xz} and Ω_{xzz}). Consequently, we can see that the increasing of $\Gamma^{(dO)}(\mathbf{r}_0, \phi)$ is slower than $\Gamma^{(Q)}(\mathbf{r}_0, \phi)$ as QD radius enlarges, and becomes negative due to the transition interferences between the dipole and different octupoles.

3 Tuning the strong plasmon-QD coupling

In the following we study the impact of the nondipole effect on the vacuum Rabi splitting (VRS), which is the fingerprint of strong light-matter interaction. In Figure 3(a), we plot the effective cooperativity parameter $C_{\text{eff}} = \Delta^2 / \gamma_1 \gamma_2$ for $\phi = 0$, where Δ is the eigenenergy splitting, γ_1 and γ_2 are the linewidth of two eigenmodes [47]. $C_{\text{eff}} \geq 1$ indicates the preservation of coherence in light-matter interaction and is a qualitative criterion for strong coupling [49], and

C_{eff}^{-1} is also the number of quantum emitters needed to significantly disturb the cavity to exhibit the quantum nonlinearities [50]. Figure 3(a) shows that C_{eff} reaches 50 for $(s, R) = (2 \text{ nm}, 10 \text{ nm})$ with the nondipole effect, demonstrating a four-fold enhancement compared to a point dipole. Consequently, the probabilities of upper level $P_e(t) = |C_e(t)|^2$ for the initially excited QDs with the nondipole effect present faster and greater Rabi oscillations than that of a point dipole (see Figure 3(b)), indicating the stronger coherent energy exchange in plasmon-matter interaction.

Figure 3(c) presents the spontaneous emission (SE) spectra of QDs with various radii and different orientations

Figure 3: Tuning the plasmon-QD strong coupling by the nondipole effect. (a) Cooperativity parameter C_{eff} as the function of QD radius and s for $\phi = 0$. (b) Temporal dynamics of spontaneous emission (SE) for various QDs, with parameters $s = 2 \text{ nm}$ and $\phi = 0$. (c) SE spectra versus QD energy for $s = 2 \text{ nm}$ (left) and $s = 4 \text{ nm}$ (right) with $\phi = 0$ (upper panel) and $\phi = \pi$ (lower panel) for various QD radii. (d) Sketch of an experimental realization of QD rotation along its dipole orientation (i), where a QD is embedded in the center of a cylindrical medium and separated from the nanoshell at a distance of R . The corresponding SE spectra and $g - \phi$ relation for a QD with $s = R = 4 \text{ nm}$ are plotted in (ii) and (iii), respectively. The results of DA are obtained by the point dipole with a dipole moment of 60D. $n_c = 1.46$.

for $s = 2$ and 4 nm. The definition and calculation of the SE spectra can be found in Ref. [47]. With $\phi = 0$, we can see that the VRS enlarges as QD radius increases. The VRS of a QD with 10 nm radius is ~ 38 meV, while the VRS for a point dipole is 20 meV. For $\phi = \pi$, no spectral splitting can be resolved for $R = 6$ and 8 nm, while the peak width of an 8 nm-radius QD is narrower than that of a 6 nm-radius QD. However, the VRS emerges again as R increases. This behavior is similar to the LCS shown in Figure 2(c) and (d), indicating a relatively weak coupling for a QD with 8 nm radius. The right column of Figure 3(c) plots the SE spectra for $s = 4$ nm, where the VRS in DA is hard to recognize. Under this circumstance, the enhanced plasmon–exciton coupling is found for $\phi = 0$, while with the opposite orientation ($\phi = \pi$), the VRS for various QDs radii disappears. Figure 3(c) indicates that the nondipole effect offers strong tunability to the plasmon–exciton interaction.

To flexibly utilize the nondipole effect of QDs, in Figure 3(d), we propose an experimental model that can continuously tune the QD orientation ϕ , and thus the coupling strength g of plasmon–QD interaction can be directly manipulated. The lower panels in Figure 3(d) show the SE spectra and corresponding g as ϕ varies from 0 to 2π for $s = R = 5$ nm. We can see that by tuning the QD orientation, the VRS in SE spectra smoothly evolves into a single sharp peak at $\phi = \pi$ due to the suppression of plasmon–QD coupling. In Ref. [51], a similar spectral feature has also been found in the power spectrum of two coupled atoms in quasichiral regime, but results from the subradiant state of two atoms. We can also see that the VRS, and thus the coupling strength g , has π -symmetry and decreases as ϕ approaches to π . It implies that the dipole–quadrupole interference dominates over the dipole–octupole interference and the quadrupole transitions, because the former has π -symmetry while the latter shows no obvious symmetry with respect to ϕ (see (3)–(6)).

4 High-dimension EP space

An EP is the coalescence of two polariton branches [4], which is a degenerate point in the underlying Hilbert space and a critical point between the weak and strong coupling. Therefore, the EP corresponds to a certain coupling strength g_{EP} . For a point dipole with fixed dipole orientation, the coupling strength of plasmon–exciton interaction depends on the relative location of the dipole and plasmonic structure. Therefore, there are at most three degrees of freedom (x, y, z) to form an exceptional surface (ES). As shown in Figure 3(c) and (d), the nondipole effect

establishes an extra degree of freedom ϕ for the coupled plasmon–QD system, then its EPs can be extended into a three-dimensional exceptional space by utilizing all four degrees of freedom (x, y, z, ϕ). Although exceptional ring [1, 6] and surface [16] have been created in classical optical systems, the formation of three-dimensional exceptional space in two coupled quantum systems far below the diffraction limit is for the first time predicted.

Figure 4(a) demonstrates the exceptional space of our system for $x, y > 0$, where s is set to be equal to the QD radius R . We can see that each value of ϕ corresponds to an ES and their collection constructs a three-dimensional EP chamber. As a comparison, a point dipole can only form an ES inside the chamber. Figure 4(b) shows the slices of the EP chamber for different values of s , where we can see that the collection of EPs has a ring shape for small R , because the strong nondipole effect can bring the plasmon–QD interaction into the weak-coupling regime. For a middle s , the shape of EP slice is a circle but with a smaller radius than the EP ring. In Figure 4(c), we plot a one-dimensional exceptional line (EL) at $x = y = 0$ for $R = 5$ nm. It shows that accessing the quantum EPs can be realized in the range of QD-nanoshell separation $5.5 \text{ nm} < s < 6.4 \text{ nm}$ with proper QD orientation, while the EP of a point dipole is fixed at $s = 5.48 \text{ nm}$ (dashed gray line). Furthermore, note that rotating the plasmonic nanoparticles has a similar effect as adjusting the QD orientation, and can be easily done by using a metallic tip as scanning probe [34, 36]. Therefore, the nondipole effect provides an effective method for experimentally demonstrating the non-Hermitian quantum physics at nanoscale.

The quantum EP can be discerned by dynamically measuring the power spectra [28–30, 52], and has potential applications in tuning the photon statistics [53, 54] and ultrasensitive quantum sensing [26, 27, 55]. For the coupled plasmon–QD system operated around EP, it is sensitive to the perturbative variation of coupling strength. The eigenenergy splitting Δ around the EP is approximately expressed as $\Delta_{\text{EP}} = 2\sqrt{2g_{\text{EP}}\epsilon}$ and approaches 2ϵ far from the EP [47]. This implies that the VRS can demonstrate the typical feature of second-order EP [2]. As shown in (i) of Figure 4(d), the SE spectra of our system show larger spectral splitting compared to the system without or working far away from EP when subjected to the same perturbation. For a 4 nm-radius QD, the VRS observed in the SE spectra is $\sim 50\%$ larger than a linear-response system with the same coupling strength g . Notice that g and ϕ show approximately linear dependence in the parameter range of $0.5\pi < \phi < 0.9\pi$ (see (ii) in Figure 4(d)), which

Figure 4: A high-dimensional quantum exceptional space. (a) Visualization of the exceptional space for QDs with the nondipole effect. The exceptional surface for a point dipole is also shown. (b) Exceptional surfaces for $s = R = 4$ nm (upper panel) and 5 nm (lower panel) in (a). No EP exists in the gray area. (c) Exceptional line as a function of s and ϕ for $x = y = 0$ nm and $R = 5$ nm. The gray dashed line labels the QD radius for eigenenergy splitting $\Delta = 0$ in the DA. The blue area is the parameter region of weak coupling with the nondipole effect, while the black solid lines mark the EL between the strong and weak coupling. (d) Comparison of the eigenenergy splitting Δ (blue line) and the VRS (purple hollow circle) when varying g via rotating a 4 nm-radius QD from $\phi = 0.5\pi$ to 0.9π (i), where g and ϕ have a well linear relation (ii). The VRS is extracted from the SE spectra of QD (iii). The system configuration is the same as that in Figure 3(d). See the text for the expressions of “EP” and “Linearity” response in (i) of (d).

makes our system as an ultrasensitive quantum sensor with respect to the rotation angle ϕ .

5 Discussion and conclusion

At last, we carry a deep discussion about the theoretical model. The remarkable nondipole effect of InAs/GaAs QDs is beneficial from the large lattice distortion. For a longitudinal-coupled QD with deformation in dipole orientation, the broken symmetry at the level of Bloch function gives rise to quadrupole moments, and the resulting

nondipole effect is comparable to the InAs/GaAs QDs [47]. Therefore, the nondipole effect studied here can also be applied to other kinds of QD, even when their origins are fundamentally different. In addition, in this study the QD is modeled as a two-state quantum system, the validity of this approximation has shown to break down in the Coulomb gauge when the cavity QED system enters the ultrastrong-coupling regime [56, 57], i.e., $g/\omega_0 > 0.1$. In our system, the critical coupling strength of the EP is smaller than the QD transition energy by about three orders of magnitude. Therefore, the results obtained in this study depend

explicitly on the choice of gauge, but remain consistent under different gauges. Based on the same reason, the single-mode approximation is adequate to describe the plasmon-QD interaction in this study [58].

The EPs shown in Figure 4 are the degeneracies of non-Hermitian Hamiltonians (NHH) of Jaynes–Cummings model (JCM), corresponding to the coherent nonunitary evolution of QD-photon interaction without quantum jumps. A recent study has shown that in the quantum limit, the EPs of NHH are essentially different from the degeneracies of Liouvillian superoperator (LS) due to the existence of quantum jumps [59]. For the SE dynamics and SE spectra studied here, there is at most one photon in plasmon, thus we can restrict the state space into the single-excitation subspace. In this special case, the EP of JCM is the same as two coupled bosonic modes, which according to the result of Ref. [59], its EPs of NHH are equivalent to that of LS. Therefore, the existence of quantum jumps will not change the results shown in Figure 4. Besides, it is possible to form the higher-order quantum EPs in the coupled multi-QD-plasmon systems. For example, one can construct the three-order EPs by coupling two QDs to nanoshell, with the critical coupling strengths $g_{EP}^{1,2}$ for two QDs [47]. Utilizing the nondipole effect and the model shown in Figure 3(d), the required coupling strengths $g_{EP}^{1,2}$ can be achieved by rotating two QDs separately, with no need for precisely adjusting the relative position of three subsystems.

In conclusion, we have theoretically studied the nondipole effect of QDs strongly coupled to a nanoshell. It is for the first time revealed that the plasmon-QD interaction can be flexibly tuned between the weak and strong coupling by the nondipole effect of QDs, and that it induces a quantum exceptional chamber in the parameter space. The nondipole effect is ubiquitous and provides an extra degree of freedom with great flexibility and robustness to access the EPs of non-Hermitian quantum systems. Our work not only enriches the knowledge of light-matter interaction at nanoscale, but also opens a path to probe the non-trivial quantum topological properties.

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