Rabi oscillation study of strong coupling in a plasmonic nanocavity

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Abstract

Strong interaction between emitters and plasmonic nanocavity has various applications in quantum fields at room temperature. As Rabi oscillation gives the direct proof to the energy exchange in strong coupling, it is more intuitive and necessary to analyze the interaction in time domain. In this paper, we give the Rabi oscillation in a high-dissipation plasmonic nanocavity by using full-quantum method and draw a new strong coupling criterion about mode volume which provides a significant guidance in plasmonic nanocavity's nanofabrication. Moreover, we reveal the relation between Rabi oscillation and Rabi splitting, which is beneficial for exploring emitter–plasmon hybrid systems time-domain property through frequency-domain response. An emitter–hexagon hybrid system with ultrasmall mode volume is designed to verify our theory. The numerical simulation shows good agreements with our theoretical results. Our work has applications in quantum information and quantum computing in the future.

1. Introduction

Low-Q plasmonic nanocavity with ultrasmall mode volume strongly coupled with emitters has been a hot topic now for its wide applications in many fields such as quantum information processing, modification of the rate of chemical reactions, and low threshold laser [1–6]. A light–matter hybridized mode exhibits when the exchange rate of energy between light and matter is faster than the dissipative rate, showing an observable Rabi splitting in the frequency domain. This strong coupling between excitons and surface plasmon (SP) can be used in manipulating quantum states, providing new ideas for quantum computing and quantum devices [7–9].

Previously, strong coupling is generally realized in high-Q traditional optical cavity [10–13], which has to be achieved in cryogenic environment. In the past decade, it has been shown that plasmon produced by metallic nanoparticle can act as a nanocavity to achieve strong coupling. Moreover, compared to traditional optical cavity, strong coupling between nanoparticle and excitons can be realized in room temperature, because of the subwavelength feature of plasmon. Surface plasmon, oscillating electron on the surface of metal, can beat the diffraction limit and confine the electromagnetic fields into a volume that is much smaller than the wavelength of incident light. Such strongly confined optical mode provides necessary condition for strong coupling at room temperature although metallic nanoparticles cause high dissipation.

Recently, people focus on strong coupling between excitons and SP from multiple systems to single system with different geometries [14–24], and various of excitons are studied such as J-aggregates, quantum dots (QDs), two-dimensional materials [14–31]. In theory, lots of works concentrate on the strong interaction in frequency domain or in a composite microcavity [32–36]. Though the strong coupling in these fields has been widely reported, there are few reports about Rabi oscillation in time domain for the emitter–plasmon hybrid system, which is more significant to reveal the strong coupling mechanism. Rabi
oscillation of J-aggregate and nanoslit array system has been observed experimentally [37]. However, there still exists a great challenge for observing Rabi oscillation in single emitter–plasmon hybrid system, because the output signal of such a hybrid system is too weak to collect signal efficiently. Although the output signal intensity can be enhanced by increasing the energy or repetition rate of incident femtosecond laser, the properties of emitters (molecular, quantum dot, J-aggregate and so on) will be destroyed.

In this paper, we obtain Rabi oscillation in time domain for such a single emitter–plasmon hybrid system by utilizing a full-quantum method and draw a new strong coupling criterion about mode volume, which is more intuitive and provides a significant guidance in nanofabrication for plasmonic nanocavity. We reveal the relation between Rabi oscillation and Rabi splitting and design a hybrid system of silver regular hexagon nanocavity strongly coupled with emitters. The numerical simulation in frequency domain is in a good agreement with our theoretical results. Finally, we find an abnormal relation between Rabi splitting and mode volume, which provides idea for realizing strong coupling in single plasmonic nanocavity with ultrasmall mode volume.

2. Exciton interacts with plasmon mode in a damping system

In this section, we consider the situation that both the emitter and plasmonic nanocavity have a decay term in Jaynes–Cummings model [38], which describes the hybrid system of a two-level system (TLS) interacting with a quantized mode of an optical field. It can be written as

\[ H = \frac{1}{2} \hbar \omega_0 \hat{\sigma}_z - i \hbar \frac{\gamma_0}{2} \hat{\sigma}_{ee} + \hbar \omega_a \hat{a}^\dagger \hat{a} - i \hbar \frac{\gamma_p}{2} \hat{a}^\dagger \hat{a} + h g ( \hat{a} \hat{\sigma}_+ + \hat{a}^\dagger \hat{\sigma}_- ). \]  

(1)

Here we suppose that the decay of two-level system only happens at the excited state. \( \hat{\sigma}_z = |e\rangle \langle e| - |g\rangle \langle g| \) is the emitter-population inversion and \( \hat{\sigma}_{ee} = |e\rangle \langle e| \) is the excited state population. \( \omega_0 \) is the TLS transition frequency. \( \gamma_0 \) and \( \gamma_p \) are the decay rate of the excited state and plasmon mode, respectively. \( \omega_p \) is the frequency of LSP optical field. \( \hat{a}^\dagger \) and \( \hat{a} \) are the creation operator and annihilation operator, respectively. \( g \) is the coupling coefficient and \( \hat{\sigma}_+ = |e\rangle \langle g| \) and \( \hat{\sigma}_- = |g\rangle \langle e| \) are the raising operator and lowering operator. \( \hat{a}^\dagger \hat{\sigma}_+ \) presents the process that TLS absorbs a photon and transfers to excited state. Conversely, \( \hat{a}^\dagger \hat{\sigma}_- \) presents the process that TLS returns to ground state and releases a photon.

The wave function of the hybrid system in the interaction picture can be written at time \( t \) as

\[ |\psi(t)\rangle = \sum_n [c_e(t)|e,n\rangle + c_g(t)|g,n+1\rangle]. \]  

(2)

It shows that wave function is a linear combination of the excited state and ground state during the interaction process. \( c_e(t) \) and \( c_g(t) \) are the probability amplitude of excited state and ground state, respectively. \( |e,n\rangle \) is the state where emitter is in the excited state, while \( |g,n+1\rangle \) is the state where emitter is in the ground state.

We use Schrödinger equation to solve equation (1) in interaction picture, and obtain the probability amplitude of excited state. The probability of excited state can be obtained by multiplying the probability amplitude and its conjugate. We assume the initial state of this system is \( |e,n\rangle \), and at resonance \( (\omega_0 = \omega_p) \), the probability of excited state \( P_e(t) \) can be written as

\[ P_e(t) = e^{-\frac{\Gamma}{2}} \left[ \cos \left( \frac{\Omega_n t}{2} \right) + \frac{\Gamma}{2\Omega_n} \sin \left( \frac{\Omega_n t}{2} \right) \right]^2, \]  

(3)

where \( \Gamma = \gamma_p - \gamma_0 \) and \( \delta = \omega_0 - \omega_p + i\Delta/2 \). \( \Omega_n = \sqrt{4g^2(n+1) + \Delta^2} \) can be simplified to \( \Omega_n = \sqrt{4g^2(n+1) - \Gamma^2/4} \) at resonance. Equation (3) demonstrates that the probability of excited state oscillates with exponential decay until it decreases to zero rather than a continuous oscillation without damping. It attributes to the damping term that we introduced to our method, which makes our method more practical. \( \Omega_n \) represents Rabi oscillation frequency in time domain and it can also be recognized as Rabi splitting for a single emitter in frequency domain.

In published works, there are usually two criteria of strong coupling \( 2g > (\gamma_p - \gamma_0)/2 \) and \( 2g > (\gamma_p + \gamma_0)/2 \) [32], where \( 2g \) represents the vacuum Rabi splitting simply in frequency domain. The former only representing the minimum of coupling strength where the energy level splits while an obvious Rabi splitting may not be observed in frequency-domain spectrums due to the damped mode. The latter gives the damped mode with linewidths characterized by \( (\gamma_p + \gamma_0)/2 \), and we can observe a clear Rabi splitting in spectrums on this condition. It may seem puzzling in \( 2g > (\gamma_p - \gamma_0)/2 \) that the two decays of emitter and nanocavity can cancel each other and produce a steady Rabi oscillation in time domain.
oscillation frequency $\Omega$ that optical field at least once showing in figure 1 (blue curve and red curve). Otherwise, energy is dissipated coupling is satisfied, Rabi oscillation appears and reversible energy exchange happens between TLS and equation (3) and sine term in equation (3) is modified to represent the density of excitons in unit mode volume where $\Gamma$ coefficient considered. However, in a high dissipative system, such as plasmonic nanocavity, decay $\Gamma$ coefficient on account of the line width (damped mode) of emitter and plasmon. Here we should emphasize that $\gamma_p$ is much larger than $\gamma_0$ in our hybrid system. Both criteria $2g > (\gamma_p - \gamma_0)/2$ and $2g > (\gamma_p + \gamma_0)/2$ can be recognized as the same, in other words the strong coupling phenomenon appears when the energy-level splitting happens in our hybrid system. Here we only consider the situation of photon number $(n = 0)$, which means there is only one photon in hybrid system. Thus excited state of TLS $|e, 0\rangle$ and ground state of TLS $|g, 1\rangle$ also represent the states of no photon in the field and one photon in the field, respectively. The sine term in equation (3) is a modification to $P_e(t)$. It can be ignored when a low dissipative system is considered. However, in a high dissipative system, such as plasmonic nanocavity, decay $\Gamma$ is in the same order of Rabi oscillation frequency $\Omega_e$ and sine term has an important influence to $P_e(t)$.

Rabi oscillation is given in figure 1 with different parameters of coupling coefficient $g$ and decay coefficient $\Gamma$. For convenience we nondimensionalize $g$, $\Gamma$ and $t$ by using $\gamma$ which is set to 1 eV. We notice that $g$ determines the oscillating frequency while $\Gamma$ limits the oscillating time. When the criterion of strong coupling is satisfied, Rabi oscillation appears and reversible energy exchange happens between TLS and optical field at least once showing in figure 1 (blue curve and red curve). Otherwise, energy is dissipated through spontaneous emission of TLS without exchange, which is irreversible. It is obvious that when Rabi oscillation frequency $\Omega_e > 0$, exciton–plasmon system exhibit coupled state. At the critical point $\Omega_e = 0$, equation (3) is simplified to $P_e(t) = e^{-\Gamma t/2}(1 + \Gamma t/4)^2$ shown in figure 1 (black curve).

Furthermore, we demonstrate the relation between coupling coefficient $g$ and decay coefficient $\Gamma$ and draw a new strong coupling criterion about mode volume in a single emitter–plasmon hybrid system. From equation (3) and $\Omega_e = \sqrt{4g^2(n + 1) + \delta^2}$, we know that increasing $g$ or decreasing $\Gamma$ can increase Rabi oscillation frequency $\Omega_e$. The physical essence behind this is that $\Omega_e$ determines the energy exchange rate in time domain. When the energy exchange rate is smaller than the decay, the hybrid system are considered reaching weak coupling regime. Stronger coupling and less decay will increase the interaction of light and matter leading to a transformation process from weak coupling to strong coupling. To find obvious anticross, here we use the criterion of strong coupling which is widely accepted in frequency domain $\Omega > (\gamma_0 + \gamma_p)/2$ [24], $\Omega$ is Rabi splitting in frequency domain. It should be noticed that we can also ignore $\gamma_0$ because $\gamma_0$ is much smaller than $\gamma_p$ in our hybrid system. At resonance ($\omega_p = \omega_0$) our criterion is given by $(n + 1)g^2 > (\gamma_0^2 + \gamma_p^2)/8$. This criterion indicates that increasing coupling coefficient or lowering decay is the requirement to reach strong coupling regime. For $N$ emitters system, the total coupling coefficient $g_{\text{tot}} = d\sqrt{\hbar\omega_0 N/2\varepsilon_0 V}$, the criterion can be given by

$$N/V > \frac{\varepsilon_0\omega_0(\gamma_0^2 + \gamma_p^2)}{4(n + 1)\omega_0 d^2}. \quad (4)$$

Here $d$ is the dipole moment. $\varepsilon_0$ is the vacuum permittivity and $\varepsilon$ is the relative permittivity. $N/V$ represents the density of excitons in unit mode volume where $N$ is the number of excitons and $V$ is the
mode volume of plasmon mode. From equation (4) we know that increasing the number of coupling excitons or confining the light into smaller volume can increase the strength of the coupling in hybrid system. However, it should be noted that Rabi oscillation frequency $\Omega_n$ is not changing with the number of coupling emitters $N$ in contrary to Rabi splitting $\Omega$ which is proportional to $\sqrt{N}$. For a exciton–plasmon hybrid system of single quantum emitter, we can obtain $V < 4(n + 1)\omega_0 d^2 / (\varepsilon_0(\gamma_0^2 + \gamma_p^2))$. This condition gives the maximal mode volume of electric field for realizing strong coupling in a plasmonic nanocavity with a single exciton, and we find that larger dipole moment and lower dissipation can liberalize the ultrasmall mode volume, which has an important influence in reaching strong coupling regime. It provides a guidance for designing a plasmonic nanocavity to reach strong coupling regime especially for single emitter–plasmon hybrid system.

Additionally, we notice that photon number $n$ also plays an critical role in strong coupling criterion. In this paper we focus on weak field, which means only the strong coupling involving zero to one photons will be considered. In the absence of photon, there is no coupling between TLS and plasmon mode. When there is only one photon in the energy exchange ($n = 0$), our conclusion $g^2 > (\gamma_0^2 + \gamma_p^2)/8$ is accorded with former works including theory work [34] and experimental work [24]. It indicates that our time-domain method has a good match with previous frequency-domain method. However, when the photon number ($n \geq 1$), the hybrid system has a discrete set of states and splittings given by photon number $n$ called the Jaynes–Cummings ladder, which gives rise to an intriguing optical effect of a photon blockade. This means that the presence of first photon’s excitation blocks absorption of the second photon of the same energy, where Rabi splitting can describe by $\Omega_{\text{ladder}} = \sqrt{n} \Omega$, enabling nonlinear response with a single photon.

Moreover, we reveal the relation between the property in time domain and the response in frequency domain for an emitter–plasmon hybrid system, which is beneficial for exploring the hybrid systems time-domain property through frequency domain that is observed more easily both in simulation and experiment [40, 41]. The transform is carried out by

$$P(\omega) = \left| \int_0^\infty dt c(t)e^{-i(w - \omega_p)t} \right|^2,$$

where $\omega_p = \omega_0 - i\gamma_p/2$. The results are given in figure 2. Figures 2(a), (c) and (e) give the probability of excited state $P_e(t)$ in time domain while figures 2(b), (d) and (f) give the scattering spectra $P(\omega)$ in frequency domain. By changing the ratio of $g$ and $\Gamma$, figure 2 exhibits three states of hybrid system’s coupling—no coupling, weak coupling, and strong coupling. Figures 2(a), (c) and (e) shows that the probability of excited state changes from exponential decay to reversible Rabi oscillation with damping. Figures 2(b), (d) and (f) shows that the scattering spectrum evolves from a Lorentzian to small splitting of weak coupling, and finally an obvious Rabi splitting appears reaching strong coupling regime. The
comparison of our method results and other frequency-domain work’s results [39] is shown in figures 2(b), (d) and (f). We notice that two methods show a good agreement, which proves our theory correct. Moreover, we calculate the two polaritonic modes of exciton–plasmon hybrid system in strong coupling regime, which is given by

\[
\Omega = \frac{\omega_p - \omega_0}{2} \pm \frac{\sqrt{4\gamma_p^2 + \delta^2}}{2},
\]

where \(\omega'_0 = \omega_0 - i\gamma_p/2, \omega'_p = \omega_p - i\gamma_p/2\) and \(\delta = \omega_0 - \omega_p + i\Gamma/2\). It shows two splitting levels of hybrid system, which can also be obtained by calculating the eigenvalue of coupled oscillation model in frequency domain. The term \(i(\gamma_p + \gamma_p)/4\) in equation (6) (see equation (19) in the supplementary material (http://stacks.iop.org/NJP/22/063053/mmedia)) represents the line width (decay rate) of each polaritonic mode. Although the exponential decay term in equation (3) is the difference of two decays, we can still obtain the form of sum of two decays in frequency domain which is in agreement with strong coupling criterion. Our result fits other experimental [24] and theoretical works [39] well. Rabi splitting can be obtained from equation (6) written as \(\Omega = \sqrt{4\gamma_p^2 + \delta^2}\), where \(\gamma_p = \sqrt{Ng}\). This frequency-domain result is proportional to the concentration of excitons representing the collective effect of coupling emitters. However, Rabi oscillation \(\Omega_p\) gives the single-emitter energy exchange rate which depends on the characteristic of emitter and plasmon rather than the number of emitters \(N\). When there is no damping in the hybrid system, at resonance, the relation of Rabi oscillation and Rabi splitting can be given by \(\Omega = \sqrt{\Omega_p}\). Nevertheless, when we take the damping into account, the detuning term \(\delta\) is no longer zero at resonance. Thus we can only obtain \(\Omega \propto \Omega_p\) and \(\Omega \propto \sqrt{N}\). Our method can describe strong coupling in the both domains and demonstrate the coupling mechanism between exciton and plasmon more comprehensively.

3. Strong coupling between excitons and a plasmonic nanocavity

3.1. Hexagon strong coupled with enormous excitons

In this section, we use simulation with Lorentzian model to obtain some specific parameters for accurate analysis in quantum model and verify our theoretical results. We design a silver regular hexagon with 40 nm length of each side and 10 nm thickness shown in figure 3(a). Obviously, there are six symmetry axes in geometry and can be classified into two different types. Along two axes (x axis and y axis), we simulate the LSP mode and scattering spectrum of our structure to study the relation between electric field mode and the polarization of incident light. Finite-difference time-domain (FDTD) method is used and the background refraction index is set to 1.33. In figure 3(a), the simulation results of LSP mode are given. We find that the optical field is confined in the four corners along x axis when the polarization of incident light parallels x axis. In contrast, when the polarization parallels y axis, the field localizes in two corners along y axis. This result indicates that the optical field of regular hexagon generally localizes in the angles that is perpendicular to the polarization of incident light, which is similar to triangle nanocavity [32] and rectangle nanocavity [24]. On the other side, it is unexpected and interesting that the scattering spectrums of two different LSP modes are almost the same. We further study the relation of polarization and scattering spectrum shown in figure 3(b). When polarization changes from 0 degree to 30 degree (from an angle to the midpoint of an adjacent edge), the whole situations of polarization are involved due to regular hexagons symmetry. We notice that scattering spectrum almost stay unchanged when the polarization angle changes from 0 degree to 30 degree at interval of 5 degree. It means that scattering spectrum has no dependency on various of LSP modes caused by different polarization and stays at 636 nm. Furthermore, LSP modes is sensitive to its environment reflecting in the scattering spectrum, which has potential application in molecule detection and modulation.

Then we add emitters to this plasmonic nanocavity by coating regular hexagon with a 2 nm thick layer of J-aggregates, which can be realized in experiment. In simulation, J-aggregate can be described by Lorentzian model which is written as \(\varepsilon(\omega) = \varepsilon_{\infty} + f\omega_0^2/(\omega_0^2 - i\gamma_0\omega - \omega^2)\). \(\varepsilon_{\infty}\) is the dielectric constant and set to 1.6 [24]. \(\omega_0\) is the resonance frequency of J-aggregate. \(f = 0.03\) is the density of the dipole moment in material, and \(\gamma_0\) sets to 25 meV representing the line width. These parameters are obtained from reference [24]. When \(\omega_p = \omega_0\), LSP mode resonates with J-aggregate, and an obvious Rabi splitting (\(\Omega \sim 120\) meV) appears in scattering spectrum. Strong coupling criterion \(\Omega > (\gamma_0 + \gamma_p)/2\) is satisfied where \(\gamma_p\) is \(\sim 121\) meV. In this regime, LSP and TLS exchange energy very fast and form a new hybrid state. The anti-cross curve and scattering spectrums are shown in figures 3(c) and (d). Figure 3(c) gives the simulated dispersion of our exciton–hexagon hybrid system showing an obvious anticross curve and figure 3(d) shows that one energy level splits into two energy levels due to the strong interaction in our hybrid system.
In this section, we consider a several quantum dots (QDs) interact with LSP mode shown in figure 4(a).

3.2. Hexagon strong coupled with a several excitons

Localized LSP modes. This inspires a new way to detect the number of molecules. The single-exciton coupling coefficient $g$ can be calculated by $g = \omega / (\sqrt{2}\hbar \omega_0 \varepsilon_0 \hat{V}_{\text{eff}}) \mathbf{d} \cdot \mathbf{f}_x(\mathbf{r})$, where $\mathbf{d}$ is the dipole moment, $\mathbf{f}_x(\mathbf{r}) = \mathbf{E}_0(\mathbf{r})/\max(\mathbf{E}_0(\mathbf{r}))$ represents the normalized electric field of LSP modes. $\hat{V}_{\text{eff}}$ is the effective mode volume and can be calculated by $\hat{V}_{\text{eff}} = n^2(r) \times V_m$. We can simulate and obtain mode volume $V_m$ by using $V_m = \int \varepsilon(\mathbf{r}) |\mathbf{E}(\mathbf{r})|^2 \, dV / \max(\varepsilon(\mathbf{r}) |\mathbf{E}(\mathbf{r})|^2)$. The mode volume of $x$-polarization LSP ($x$LSP) mode and $y$-polarization LSP ($y$LSP) mode are $\sim$915 nm$^3$ and $\sim$735 nm$^3$, respectively. The total coupling coefficient $g_{\text{tot}} = 64.6$ meV calculated from equation (6), which satisfies our strong coupling criterion and we calculated that there are $\sim$14.3 and $\sim$11.4 excitons strongly interact with $x$LSP mode and $y$LSP mode, respectively. Although a layer of J-aggregate is coated to structure, the strong interaction between excitons and plasmon only happens in the sharp corner of regular hexagon due to the strongly localized LSP modes. This inspires a new way to detect the number of molecules.

The diameter of QD is 8 nm which is also simulated by Lorentzian model. $\varepsilon_\infty$ is set to 6.7 and the resonance has a red shift to 645 nm. $f$ and $\gamma_0$ are set to 0.4 and 53 meV, respectively [42]. We use 4QDs and 2QDs interact with $x$LSP mode and $y$LSP mode, respectively. The calculation and simulation results are shown in figure 4. Figure 4(a) gives the Rabi oscillation curves of four QDs (blue line) and two QDs (orange dotted line) exciton–hexagon hybrid system in time domain when the polarization of incident light are along $x$ axis and $y$ axis respectively. Although figure 4(b) demonstrate that Rabi splitting in frequency domain is both $\sim$103 meV for 4QDs and 2QDs, there are obvious difference between two Rabi oscillations in time domain. As is shown, the Rabi oscillation frequency of 2QDs hybrid system (orange dotted line) is larger than 4QDs hybrid system (blue line). This confirms our view that Rabi oscillation frequency is associated with coupling coefficient and decay rather than exciton number $N$. Meanwhile, two more excitons result in the same Rabi splitting of two hybrid systems.

Additionally, when two more QDs participate in the coupling, a larger Rabi splitting is expected according to $\Omega \propto \sqrt{N}$. However, figure 4(b) show that two Rabi splittings are both $\sim$103 meV. Considering $g \propto d \cdot E$, we suppose that the $y$LSP mode caused by $y$-polarization is more localized and 2QDs get a larger $E$ than 4QDs. To confirm it, we calculate mode volumes of both situations, and obtain $\sim$71 nm$^3$ for 4QDs and $\sim$40 nm$^3$ for 2QDs. This result demonstrates that the coupling strength is not only determined by the number of excitons, but also the localization of LSP mode. Besides, localization of optical field causes weak coupling in the farther field where the intensity of electric field is weaker. This results in weak coupling performing as the Lorentzian-like shape peak, which is the third peaks appearing at the middle of two energy levels shown in figure 4(b). Thus although strong localization can produce stronger intensity of electric field, it also leads to smaller effective overlap of QD and electric field causing weak coupling in the farther area. This contradiction between the number of excitons and the localization of LSP mode in strong
Figure 4. (a) Rabi oscillation curves of two QDs (orange dotted line) and four QDs (blue line) exciton–hexagon hybrid system in time domain. The insets are simulated electric field distributions of \(x\) LSP mode and \(y\) LSP mode. White arrows indicate the polarization of incident light. (b) Simulated normalized scattering spectrum of several QDs strongly interact with hexagon. The blue curve is the scattering of four QDs strongly coupled with silver hexagon, when the incident light is along \(x\) axis. The dotted orange curve is the scattering of two QDs strongly coupled with silver hexagon, when the incident light is along \(y\) axis. The black curve is obtained from our theoretical method.

Figure 5. Simulated relation between Rabi splittings \(\Omega\) and mode volume \(V\) for 2QD hybrid system (red line), 4QD hybrid system (blue line), and Ag bowties structure (black line). Two QDs (red line) and four QDs (blue line) exciton–hexagon hybrid system is the same as figure 4, where the polarization of incident light are along \(x\) axis and \(y\) axis. The Ag bowties structure is constructed by two equilateral triangles with one angle of each opposite \([18]\). The mode volume \(V\) is varied by changing the curvature of structure’s corner and the distance from QD and nanoparticle. The blue line start with 71nm\(^3\) which is the minimal mode volume of 4QD hybrid system.

coupling regime provides a new path in molecular detection application. We can use it to detect the number, the distance from metallic nanoparticle and even the shape of molecular.

3.3. Relation between mode volume and coupling strength

Finally, we find a novel phenomenon about the relation between mode volume \(V\) and Rabi splitting. In this section, we obtain mode volume and scattering spectrum in three different situations by simulating the hybrid system of quantum dots and hexagon plasmonic nanocavity.

Here we take the Ag bowties structure into account, which is constructed by two equilateral triangles with 60 nm side length, 30 nm thickness. One QD of 8 nm diameter is placed at the center of the gap created by two triangles. This structure is suggested to compare with two polarization modes of hexagon shown in figure 5. We alter the mode volume of three different systems by changing the curvature of structure’s corner and the distance from QD to nanoparticle. We find that for 2QD hybrid system and Ag bowties with one QD, Rabi splitting, which is proportional to coupling strength, is dramatically decreasing with the increase of mode volume when the mode volume \(V\) is small. Continuously increasing mode
volume, the decrease of Rabi splitting slow down until the systems degenerate to weak coupling regime. However, for 4QD system the Rabi splitting perform an abnormal phenomenon, where the maximal Rabi splitting appears at a medium mode volume rather than the smallest one. We suppose that the contradiction between the number of excitons and the localization of LSP mode mentioned above is responsible for this unusual result. Ultrasmall mode volume is necessary to realize strong coupling for one or two excitons. However, for multiple excitons, strong enhancement of electric field is not the only pursuit to achieve stronger coupling, but also the overlap of excitons and optical mode plays an important role. Comparing to designing new structure for ultrasmall mode volume, it is undoubtedly more efficient to control the balance of field enhancement and the overlap of excitons and electric field. This provides a new way in realizing larger Rabi splitting for plasmonic nanocavity.

4. Conclusions

To conclude, we obtain the damping Rabi oscillation in an emitter–plasmon hybrid system by using a full-quantum method, which is suitable for an arbitrary high-dissipation plasmonic nanocavity. We obtain a new strong coupling criterion about mode volume from our quantum model, which is more intuitive and provides a significant guidance plasmonic nanocavities nanofabrication. Furthermore, we reveal the relation between Rabi oscillation and Rabi splitting in three situations—no coupling, weak coupling and strong coupling, which is beneficial for exploring the time-domain property through frequency-domain response in plasmonic hybrid system. To confirm our theoretical method, we design a hybrid system of silver regular hexagon nanocavity strongly coupled with emitters. The simulation is in a good agreement with our theoretical results. Additionally, we confirm that the strong interaction between excitons and plasmon only happens in the sharp corner of regular hexagon due to the strongly localized LSP modes. Moreover, we find an abnormal relation between Rabi splitting and mode volume. This novel phenomenon of contradiction between the number of excitons and the mode volume of optical field opens perspective for chemical and biology regime such as single-molecular detection and modulation.

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