Quantum Plasmonic of Finite-Size Particles with Coherent Anti-Stokes Raman Scattering

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We have developed a semi-analytical theory to study a quantum plasmonics system composed of a quantum particle and a metallic particle with finite size. The quantum particle is composed of molecules driven by three laser fields and produces coherent anti-Stokes Raman scattering (CARS) signal which is enhanced by plasmonic resonance of the metallic particle. The enhanced CARS field and the control laser form a Λ scheme that gives highly nonlinear and spatially nonlocal effects. We obtain absorption spectral peaks that show quasi-periodic oscillations with radius of the spherical quantum particle and the transformation between drifting Fano dip and double peaks. Plasmonic resonance gives very large absorption power as well as vanishing absorption at specific values of frequency and particle radius. The space between the two particles provides a resonant cavity effect which explains the oscillations of absorption power with interparticle distance.

PACS numbers:

I. INTRODUCTION

The study of interactions between metallic nanostructures and quantum emitters such as molecules [1] and quantum dots [2]-[7] has been a subject of active investigations due to its significance in both fundamental and applied sciences. Such topic is fundamentally interesting as it involves the interactions between elementary excitations (plasmons in metals and excitons in dielectric materials) coupled with photons to form polaritons in the hybrid nanostructure. The coupling between photons, plasmon and exciton in low dimensional structures such as metallic nanostructure and quantum emitter like quantum dot or wire often leads to novel and exotic quantum optical effects [8]. Several interesting results from the study of strong coupling [9] between a semiconductor quantum dot and a spherical metallic nanoparticle have been reported. These include nonlinear Fano effect [2], [3], exciton-induced transparency [4], nonclassical photon statistics [5], effects of spin coupling in the presence of magnetic fields [6], inhibition of optical excitation and enhancement of Rabi oscillations [7] and coherent control of nano-amplifier [10]. Beside spherical metallic nanoparticle, systems that involve coupling between a quantum dot and multiple metallic nanoparticles [11], [12], metallic nanowire [13] and metallic-dielectric nanoshell [14] have also been studied. Recent studies in quantum nonlinear plasmonic include nonlinear plasmonic effects of metallic nanoparticle on a three-level quantum system with electromagnetically induced transparency [15], and quantum properties of light in a four-level double Raman configuration in a quantum plasmonic system [16].

This field of research has also found its way through a wide range of applications such as the development of biological markers [17], nanosensors [18], [19] and control of emission from semiconductor nanocrystals [20]-[22]. One of the most remarkable applications of plasmon-exciton coupling is in molecular spectroscopy such as surface-enhanced Raman scattering (SERS) [23] technique that provides species-specific molecular fingerprints. The proximity of the system to metallic nanostructure enables local field enhancement by surface plasmon resonance (SPR) resulting in the amplification of the SERS signal in the order of $10^3 - 10^{10}$ times [24]. Spectroscopic signal can be further enhanced through coherent nonlinear optical processes such as coherent anti-Stokes Raman scattering (CARS) [25],[26]. Surface enhanced CARS signal [27] near hot spot regions of metallic nanostructures can be much higher than that of SERS, enabling single molecule detection [28] by maximizing the scattered CARS signal at the Fano resonance frequency (dip) [29] where absorption vanishes. On the other extreme, large absorption peaks due to excitation of plasmon resonances produce very intense electric fields in a localized region that can cause photo-disintegration of biomolecules and this would be useful for destruction of cancer cells.

In this paper, we investigate the plasmonic effects of a nearby spherical metallic particle (MP) on a quantum system (QS) of spherical particle composed of molecules driven by lasers in CARS scheme, as illustrated in Fig. 1. In particular, we develop a theory to study the effects of size of the particles on the absorption spectra of the hybrid structure. The molecular quantum system is modelled as discrete four energy levels coupled to three laser fields that produce the CARS field through four-wave mixing process. We know from previous studies that strong field coupling can cause splitting of spectroscopic peaks and create Fano dip that would complicate the spectra. It is interesting to explore how these strong coupling effects are affected by the size of the quantum and metallic particles. Spatial inhomogeneity of the laser fields inside the spherical quantum particle is accounted by the Mie theory. We compute the absorption power spectra of these particles and study the effects of the radius of the particles ranging from nanometer to micrometer regimes as well as the effects of interparticle
distance. For mesoscopic particle with radius larger than 1 μm, the CARS signal in the QS can be enhanced to a sufficiently high level that it couples strongly with the control laser field to form a Raman transition beyond the electromagnetic induced transparency (EIT) regime, and further contributes to the CARS signal. Together with the plasmonic resonance from the MP and the spatial dependent local field as well as the laser fields, the mesoscopic effects contribute to an exotic plasmonic nonlinear quantum coherence.

II. SCATTERED FIELDS WITH LOCAL FIELD ENHANCEMENT

In the far field approximation, the coherent anti-Stokes Raman scattering electric field in frequency domain $\omega$ generated by four-wave mixing process in a particle of volume $V$ is [30],[31]

$$E(R, \omega) = \frac{\omega^2}{c^2} \int_V \frac{\mathbf{P}(r, \omega) e^{i k(\omega) |\mathbf{R} - \mathbf{r}|}}{4 \pi \varepsilon_0 |\mathbf{R} - \mathbf{r}|} d^3r$$

(1)

where $k(\omega) = \omega/c$ and $\mathbf{P}(r, \omega) = \hat{\mathbf{\Theta}} P_\Theta (r, \omega) + \hat{\mathbf{\Phi}} P_\Phi (r, \omega)$ is the polarization vector transverse to the radial direction $\hat{R}$.

In the QS-MP system, the field scattered by one particle becomes the local field at the position inside another particle. Specifically, the electric field at $\mathbf{R}$ scattered by the QS of radius $\rho$ is:

$$E_{QS}^S(\mathbf{R}, \omega) = - \int_{\Omega(\mathbf{R} - \mathbf{r}) < \rho} C_{QS}(\mathbf{R} - \mathbf{r}, \omega) N_{QS} \hat{\psi}_{ca}^\dagger \hat{\rho}_{ac} (\mathbf{r}, \omega) d^3r$$

(2)

where $N_{QS}$ is the number density of the molecules, $\hat{\psi}_{ca}$ is the transverse transition matrix element of electric dipole moment operator. The coherence of the $a - c$ transition due to both the CARS and the $\Lambda$ (Raman) transition coupled by the CARS field and the control laser field is given by

$$\hat{\rho}_{ac} (\mathbf{r}, \omega) = \hat{\rho}_{ac}^{NL} (\mathbf{r}, \omega) + \frac{\varepsilon_{e f f}^{NL}}{\varepsilon_{QS}^{NL}} (\mathbf{r}, \omega).$$

(3)

which is the Fourier transform of $\rho_{ac}(\mathbf{r}, t)$ (in Schrodinger picture) while the expressions for $\hat{\rho}_{ac}^{NL}$ and $\varepsilon_{e f f}^{NL}$ are given in the Appendix A and B, respectively. The effective dielectric function is

$$\varepsilon_{e f f}^{NL} (\mathbf{r}, \omega) = \frac{2 \varepsilon_b + \varepsilon_1^{(2)} (\mathbf{r}, \omega)}{3 \varepsilon_b}$$

(4)

appears because of the local field effect which is exclusively applied only to the linear response part of the polarization, where $\varepsilon_b$ is the background dielectric function around the QS and the dielectric function is given by Eq. 75 in Appendix B. Thus, Eq. 3 provides semi-analytical approach that includes both CARS and Raman, like the case of fluorescence and Raman [32].

The transverse dipole moment in the standard spherical polar coordinates is given by [30]

$$\hat{\psi}_{ca}^\dagger = \hat{\psi}_{ca, \Theta} \hat{\Theta} + \hat{\psi}_{ca, \Phi} \hat{\Phi} =$$

$$\left( \begin{array}{ccc} 0 & 0 & 0 \\
\cos \Theta \cos \Phi & \cos \Theta \sin \Phi & - \sin \Theta \\
- \sin \Theta & \cos \Phi & 0 \end{array} \right) \left( \begin{array}{c} R P_{\Theta}^{NL} \\
\hat{\Theta} P_{\Phi}^{NL} \\
\hat{\Phi} P_{\Theta}^{NL} \end{array} \right).$$

(5)

and the nonlocal response function is (assuming $R >> |\mathbf{r}|$)

$$C_{QS}(\mathbf{R} - \mathbf{r}, \omega) = \frac{\omega^2 e^{i k(\omega) \mathbf{R} - \mathbf{r}}}{c^2 4 \pi \varepsilon_0 R}$$

(6)

Similarly, the electric field at $\mathbf{R}'$ scattered by the MP of radius $a$ is...
\[
E_{MP}^{\perp}(\mathbf{R'}, \omega) = -\int_{|\mathbf{r}'|<a} C_{MP}(\mathbf{R}'-\mathbf{r}', \omega) \alpha(\mathbf{r}', \omega) E_{MP}^{\perp}(\mathbf{r}', \omega) \frac{d^3r'}{V_{MP}} \tag{7}
\]

\[
= -\int_{|\mathbf{r}'|<a} C_{MP}(\mathbf{R}'-\mathbf{r}', \omega) \frac{1}{\varepsilon_{MP}^{loc}(\mathbf{r}', \omega)} \left[ \varepsilon_0 \chi_{MP}(\mathbf{r}', \omega) E_{MP}^{\perp}(\mathbf{r}', \omega) \right] d^3r'
\]

where \(V_{MP}\) is the volume of the MP with the nonlocal response function (assuming \(R' >> |\mathbf{r}'|\)).

Note that the term in the square bracket of Eq. 7 is the linear polarization of the MP. Also, the scattered far-field \(E_{MP}^{\perp}(\mathbf{r}', \omega)\) depends on the transversal components of the local fields \(E_{MP,\Theta'}^{loc}(\mathbf{r}', \omega)\) and \(E_{MP,\Phi'}^{loc}(\mathbf{r}', \omega)\), through

\[
E_{MP}^{\perp}(\mathbf{r}', \omega) = \hat{\Theta} E_{MP,\Theta'}^{loc}(\mathbf{r}', \omega) + \hat{\Phi} E_{MP,\Phi'}^{loc}(\mathbf{r}', \omega). \tag{9}
\]

We define the linear susceptibility \(\chi_{MP}\), Drude-type dielectric function for metal \(\varepsilon_{MP}\) [33] and effective dielectric function \(\varepsilon_{MP}^{eff}\), respectively, as

\[
\chi_{MP}(\mathbf{r}', \omega) = \frac{\varepsilon_{MP}(\mathbf{r}', \omega) - \varepsilon_b}{\varepsilon_b}, \tag{10}
\]

\[
\varepsilon_{MP}(\mathbf{r}', \omega) = \varepsilon_{\infty} - \frac{\omega_p^2}{\omega(\omega + i\Gamma)}, \tag{11}
\]

\[
\varepsilon_{MP}^{eff}(\mathbf{r}', \omega) = \frac{2\varepsilon_b + \varepsilon_{MP}(\mathbf{r}', \omega)}{3\varepsilon_b}, \tag{12}
\]

which are related to the polarizability \(\alpha\) and the enhancement factor \(\gamma\) by

\[
\alpha(\mathbf{r}', \omega) = V_{MP} \varepsilon_0 \gamma(\mathbf{r}', \omega), V_{MP} = \frac{4\pi a^3}{3} \tag{13}
\]

\[
\gamma(\mathbf{r}', \omega) = 3 \frac{\varepsilon_{MP}(\mathbf{r}', \omega) - \varepsilon_b}{\varepsilon_{MP}(\mathbf{r}', \omega) + 2\varepsilon_b} = \frac{\chi_{MP}(\mathbf{r}', \omega)}{\varepsilon_{MP}^{eff}(\mathbf{r}', \omega)} \tag{14}
\]

where \(\varepsilon_{\infty}\) is the background dielectric function at frequencies far beyond \(\omega_p\) due to positive ion cores in noble metals, \(\omega_p\) is the plasma frequency and \(\Gamma\) is the electron damping coefficient.

Using the plasmonic resonance condition \(2\varepsilon_b + \varepsilon_{MP}(\omega) = 2\varepsilon_b + \varepsilon_{\infty} - \frac{\omega_p^2}{(\omega + i\Gamma)} = 0\) we define the plasmonic resonant frequency \(\omega_r = \sqrt{\frac{\omega_p^2}{2\varepsilon_b + \varepsilon_{\infty}} - \Gamma^2}\). We assume the CARS peak (transition frequency) to be at \(\omega_{ac}\), i.e. resonant to the separation between level \(a\) and level \(c\). Then, we would have plasmonic resonance by setting \(\omega_{ac} = \omega_r\), which gives

\[
\omega_{p0} = \sqrt{(\omega_{ac}^2 + \Gamma^2)(2\varepsilon_b + \varepsilon_{\infty})}, \tag{15}
\]

the plasma frequency that gives plasmonic enhancement at the transition frequency \(\omega_{ac}\).

### III. LOCAL FIELDS FOR METALLIC PARTICLE-QUANTUM SYSTEM

According to Fig. 1 the MP is displaced by \(\mathbf{D} = \hat{\varepsilon} \mathbf{D}\) from QS, so \(\mathbf{r}'\) becomes \(\mathbf{D} + \mathbf{r}'\).

#### A. Local Field for MP at \(\mathbf{r}'\)

The local field \(E_{MP}^{loc}(\mathbf{D} + \mathbf{r}', \omega)\) at \(\mathbf{D} + \mathbf{r}'\) of the MP consists of the scattered fields from all points \(\{\mathbf{s}\}\) inside the QS which depends on the quantum coherence \(\hat{\rho}_{ac}\), i.e.
\[
E_{MP}^{\text{loc}}(D + r', \omega) = -N_{QS} \omega^2 \int_{Q_S,|s|<\rho} C_{QS}(D + r' - s, \omega) \hat{p}_{ac}(s, \omega) \, d^3s
\]  
(16)

\[
C_{QS}(D + r' - s, \omega) = \frac{\omega^2 e^{i\epsilon(\omega)(|D + r'| - \hat{n}_+ \cdot s)}}{4\pi\varepsilon_0 |D + r'|}
\]  
(17)

Assuming the z-axis is along the interparticle axis (as depicted in Fig. 1)

\[
\hat{n}_+ = \frac{D + r'}{|D + r'|} \approx \hat{z}
\]  
(18)

\[
D + r' = (\hat{z} D + r' \hat{z}')
\]  
(19)

\[
|D + r'| = \sqrt{r'^2 \sin^2 \theta' + (D + r' \cos \theta')^2} \approx D\left(1 + \frac{1}{2} \frac{r'^2 + 2D r' \cos \theta'}{D^2}\right)
\]  
(20)

\[
\hat{n}_+ \cdot s = \frac{s(D \cos \theta + r'u)}{|D + r'|} \approx r \cos \theta
\]  
(21)

\[
u(\theta', \phi', \theta, \phi) = \hat{r}' \cdot \hat{s}
\]  
(22)

where \( \hat{z} \cdot s = s \cos \theta, \hat{r}' = (\sin \theta' \cos \phi', \sin \theta' \sin \phi', \cos \theta'), \hat{s} = (\sin \theta \cos \varphi, \sin \theta \sin \varphi, \cos \theta) \) and \( s = ss' \). We may not neglect \( r'u \) since it may be comparable to \( D \cos \theta \) when \( \theta \) is close to 90°.

### B. Local Field for QS at r

The local field \( E_{QS}^{\text{loc}}(r, \omega) \) at \( r \) in the QS comprises of the integrated polarization corresponding to scattering by all points \( \{D + r'\} \) inside the MP, i.e.

\[
E_{QS}^{\text{loc}}(r, \omega) = -\varepsilon_0 \int_{MP,|r'|<a} C_{MP}(r - (D + r'), \omega) \frac{\chi_{MP}(D + r', \omega) \varepsilon_{\text{eff}}(D + r', \omega)}{\varepsilon_{\text{eff}}(D + r', \omega)} E_{MP}^{\text{loc}}(D + r', \omega) \, d^3r'
\]  
(23)

where \( \varepsilon_{\text{eff}} \) (given by Eq. 12) is due to the local field enhancement to the linear polarization like the second term in Eq. 3.

We assume that there is no external CARS field or incident laser field which may be quasi-resonant with the \( a - c \) transition or CARS transition. Inserting Eq. 16 into Eq. 23 we have

\[
E_{QS}^{\text{loc}}(r, \omega) = \varepsilon_0 \int_{MP,|r'|<a} C_{MP}(r - D - r', \omega) \frac{\chi_{MP}(D + r', \omega)}{\varepsilon_{\text{eff}}(D + r', \omega)} \left[N_{QS} \omega^2 \int_{Q_S,|s|<\rho} C_{QS}(D + r' - s, \omega) \hat{p}_{ac}(s, \omega) \, d^3s\right] d^3r'
\]  
(24)

where

\[
C_{MP}(r - D - r', \omega) = \frac{\omega^2 e^{i\epsilon(\omega)(|r - D| - \hat{n}_- \cdot r')} \varepsilon_{\text{eff}}(r - D)}{4\pi\varepsilon_0 |r - D|}
\]  
(25)

\[
\hat{n}_- = \frac{r - D}{|r - D|} \approx -\hat{z}
\]  
(26)

\[
r - D = (r \hat{r}' - \hat{z} D)
\]  
(27)

\[
|r - D| = \sqrt{r'^2 \sin^2 \theta + (D - r \cos \theta)^2}
\]  
(28)

\[
\hat{n}_- \cdot r' = \frac{r' (r u - D \cos \theta')}{|r - D|} \approx -r' \cos \theta' (\theta' \text{ not } 90^\circ)
\]  
(29)

\[
\hat{r} = [r \sin \theta \cos \varphi, r \sin \theta \sin \varphi, r \cos \theta].
\]  
(30)
For small separation $kD \lesssim 1$ or near-field regime, the longitudinal field component is present when the contribution of the operator $\nabla \nabla$ is evaluated exactly [34], but this involves more elaborate calculations. To avoid further complication to the theory, we confine our study to the far field regime $kD > 1$.

Assuming $r' \ll D$ the phase factor in $C_{QS}$ can be expanded as

$$
e^{i k(\omega)|\mathbf{D} + r'| - \hat{n}_+ \cdot \mathbf{s}} \approx e^{i k(\omega)|\mathbf{D} + r'|} e^{-i k(\omega)\hat{n}_+ \cdot \mathbf{s}}$$

where we use $|\mathbf{D} + r'| \approx D + \hat{z} \cdot r'$ and

$$
e^{i k(\omega)|\mathbf{D} + r'|} \approx \frac{e^{i k(\omega)D}}{D} e^{i k(\omega)\hat{z} \cdot r'}$$

The product of the two phase factors becomes

$$
e^{i k(\omega)|\mathbf{r} - \mathbf{D} - \hat{n}_- \cdot \mathbf{r}'|} e^{i k(\omega)|\mathbf{D} + \mathbf{r}'| - \hat{n}_+ \cdot \mathbf{s}} \approx e^{i k(\omega)|\mathbf{r} - \mathbf{D}| - \hat{n}_- \cdot \mathbf{r}'|} e^{i k(\omega)|\mathbf{D} + \mathbf{r}'|} e^{-i k(\omega)\hat{n}_+ \cdot \mathbf{s}}$$

where we use

$$
(\hat{z} - \hat{n}_-) \cdot \mathbf{r}' = r' \cos \theta' - \frac{r'(r u - D \cos \theta)}{|\mathbf{r} - \mathbf{D}|} \approx 2 r' \cos \theta' - \frac{r r' u}{D},
$$

$$
\hat{n}_+ \cdot \mathbf{s} = s \frac{|D \cos \theta + r' u|}{|\mathbf{D} + \mathbf{r}'|} \approx s \cos \theta' + \frac{r r' u}{D}
$$

Inserting the $C_{QS}$ into Eq. 16, the local field at MP becomes

$$
\mathbf{E}_{MP}^{\text{loc}} (\mathbf{D} + \mathbf{r}', \omega) = -\frac{\omega^2}{c^2} \frac{e^{i k(\omega)|\mathbf{D} + \mathbf{r}'|}}{4 \pi \varepsilon_0 |\mathbf{D} + \mathbf{r}'|} N_{QS} \overline{\varepsilon}_{ca} \Psi (\mathbf{r}', \omega)
$$

where we define the integral in $\mathbf{E}_{MP}^{\text{loc}}$ as

$$
\Psi (\mathbf{r}', \omega) = \int_{|\mathbf{s}| < \rho} e^{-i k(\omega)\hat{n}_+ \cdot \mathbf{s}} \mathcal{P}_{ac} (\mathbf{s}, \omega) d^3 s
$$

Inserting the $C_{MP}$ and $C_{QS}$ into Eq. 24, the local field at Q can be expressed as

$$
\mathbf{E}_{QS}^{\text{loc}} (\mathbf{r}, \omega) = A (\mathbf{r}, \omega) \int_{|\mathbf{r}'| < a} \int_{Q_1, |\mathbf{s}| < \rho} \frac{\chi_{MP} (\mathbf{D} + \mathbf{r}', \omega)}{\varepsilon_{MP} (\mathbf{D} + \mathbf{r}', \omega)} \frac{1}{1 + \frac{r^2 + 2D r' \cos \theta'}{2D^2}}
$$

$$
e^{i k(\omega)(\hat{z} - \hat{n}_-) \cdot \mathbf{r}' - i k(\omega)\hat{n}_+ \cdot \mathbf{s}} \mathcal{P}_{ac} (\mathbf{s}, \omega) d^3 s d^3 r'
$$

$$
A (\mathbf{r}, \omega) = \frac{\overline{\varepsilon}_{ca} \varepsilon_0 N_{QS} (\omega^2 / 4 \pi \varepsilon_0 c^2)^2 e^{i k(\omega)D}}{|\mathbf{r} - \mathbf{D}|}.
$$

For $\frac{r'}{D} \ll 1$ the $\Psi (\mathbf{r}', \omega)$ becomes $\Psi (\omega)$, almost independent of space as $e^{-i k(\omega)s \cos \theta}$ is approximately $e^{-i k(\omega)s \cos \theta}$. If the metallic particle is assumed to have a homogeneous charge density or plasma frequency, $\varepsilon_{MP} (\omega)$ would be independent of space too. Hence, the local field simplifies to

$$
\mathbf{E}_{QS}^{\text{loc}} (\mathbf{r}, \omega) = A (\mathbf{r}, \omega) \frac{\chi_{MP} (\omega)}{\varepsilon_{MP} (\omega)} \Psi (\omega) V_{MP}
$$

$$
V_{MP} = \int_{|\mathbf{r}'| < a} e^{i k(\omega)(1 + \frac{\rho}{|\mathbf{r} - \mathbf{D}|}) \mathbf{r}' \cos \theta' d^3 r'}
$$
IV. NONLINEAR INTEGRAL EQUATION

Equations 39 and 38 form a highly nonlinear and spatially nonlocal problem for \( \tilde{E}_{\text{QS}}^{\text{loc}} \) because the coherence \( \tilde{\rho}_{ac}^L \) in the integral also depends on the local field \( \tilde{E}_{\text{QS}}^{\text{loc}} \) as given by Eq. 69. The \( \tilde{\rho}_{ac}^{NL} \) in \( \Psi \) is due to three lasers interacting to produce nonlinear four-wave mixing CARS field, which contributes partly to the local field \( \tilde{E}_{\text{QS}}^{\text{loc}} \) and at the same time it can also be sufficiently strong to drive the \( a - c \) transition in the \( \Lambda \) system to produce the coherence \( \tilde{\rho}_{ac}^L \) given by Eq. 69. Phase matching of the CARS is implicitly included in \( \tilde{\rho}_{ac}^{NL} \) (given by Eq. 64) through the laser wavevectors contained in the spatial distribution functions of the respective focused laser fields as computed exactly by the Mie theory (Appendix E in ref. [31]).

Please note that, while \( \tilde{\rho}_{ac}^L = \tilde{\rho}_{ac}^L(r, \omega, J_1^\parallel, \epsilon) / \hbar \), \( I \sim 4 \), \( J \sim 0 \), \( |\alpha \rangle \sim \tilde{\rho}_{ac}^{NL}(\tilde{r}, \omega) r^2 \sin \theta \theta d\theta dr d\phi \) the coherence \( \tilde{\rho}^{NL}(\tilde{r}, \omega) r^2 \sin \theta \theta d\theta dr d\phi \)

and the second term (spatially nonlocal) has the response given by

\[
H(s, \omega) = \int_R \int_\pi \int_\phi^2 e^{-ik(\omega)n_s} \tilde{\rho}^{NL}(r, \omega) r^2 \sin \theta d\theta dr d\phi
\]

and the second term (spatially nonlocal) has the response given by

\[
H(s, \omega) = \int_R \int_\pi \int_\phi^2 e^{-ik(\omega)n_s} \tilde{\rho}^{NL}(r, \omega) r^2 \sin \theta d\theta dr d\phi
\]

where \( \eta = \frac{N_{\text{tot}}|\phi|}{k_{\text{e}}} \). We have used the pieces in the Appendix B to express the ratio as

\[
\frac{\tilde{\rho}_{ac}^L(s, \omega)}{\tilde{\rho}_{ac}^{NL}(s, \omega)} = \frac{F(s, \omega) \chi_{\text{QS}}(s, \omega)}{\eta \tilde{\rho}_{ac}^{NL}(s, \omega)} = iF(s, \omega) \frac{\chi_{\text{QS}}(s, \omega)}{1 + \frac{1}{3}\chi^{(1)}(s, \omega)} \left( \frac{n_{ac}I_c - n_{ac}J_b(\omega)}{J_c(\omega)J_b(\omega) - I_{41}I_c} \right)
\]

where \( I_c, J_4, J_b, J_c \) and \( \chi^{(1)} \) are defined in Appendix B.

When the CARS field is small the \( |F(s, \omega)|^2 \) in Eq. 48 may be neglected, giving

\[
F(r, \omega) = F_0(r, \omega) + C(r, \omega) \int F(s, \omega) H_0(s, \omega) d^3 s
\]

with the kernel

\[
H_0(s, \omega) = i \frac{e^{-ik(\omega)n_s}}{B(s, \omega) + \frac{1}{3}\eta}
\]
On the other extreme, when the CARS field is large such that \(|F(s, \omega)|^2 >> I_c, |\Upsilon_{\alpha\beta}|\), we have

\[
F(r, \omega) = F_0(r, \omega) + C(r, \omega) \left( \frac{n_{ac}}{\Gamma_{ac}} \right) \int \frac{F(s, \omega) e^{-ik(s, \omega)}r_s}{1 + \frac{1}{3}i\eta \gamma_{ac}} d^3 s
\]  

(54)

In either case both Eqs. 52 and 54 are the Fredholm equation of the second kind. Note that we may not avoid getting the Fredholm equation simply by reformulating the scattered field Eq. 1 such that the linear polarization is transformed away because the linear dielectric function has spatial dependence.

We may employ the method of ref. [36] to solve the Fredholm or the so-called Lippmann-Schwinger equation by multiplying Eq. 52 with the kernel \(H_0(r, \omega)\) (Eq. 53) and integrating, we have

\[
\int F(r, \omega) H_0(r, \omega) d^3 r = \int F_0(r, \omega) H_0(r, \omega) d^3 r + \int C(r, \omega) H_0(r, \omega) d^3 r \int F(s, \omega) H_0(s, \omega) d^3 s
\]  

(55)

and the integral to be evaluated

\[
\xi_0 = \int F(r, \omega) H_0(r, \omega) d^3 r = \frac{\int F_0(r, \omega) H_0(r, \omega) d^3 r}{1 - \int C(r, \omega) H_0(r, \omega) d^3 r}.
\]  

(56)

Hence, the (approximate) initial solution obtained by neglecting the \(|F|^2\) in the kernel of Eq. 48 is

\[
F_i(r, \omega) = F_0(r, \omega) + C(r, \omega) \xi_0
\]  

(57)

which shows the physics of the field enhancement through the second term. To obtain a better solution, we replace \(F(r, \omega)\) in the kernel of Eq. 48 by \(F_i(r, \omega)\), the approximate solution Eq. 57, giving

\[
H(r, \omega) \simeq H_i(r, \omega) = i \frac{e^{-ik(s, \omega)}r_s}{1 + \frac{1}{3}i\eta \gamma_{ac}} \left( \frac{A(r, \omega) - n_{ac}|F_i(r, \omega)|^2}{B(r, \omega) + \Upsilon_{ac}(\omega)|F_i(r, \omega)|^2} \right).
\]  

(58)

Integrating as before

\[
\int F(r, \omega) H_i(r, \omega) d^3 r = \int F_0(r, \omega) H_i(r, \omega) d^3 r + \int C(r, \omega) H_i(r, \omega) d^3 r \int F(s, \omega) H_i(s, \omega) d^3 s
\]  

(59)

we have

\[
\xi_i = \int F(r, \omega) H_i(r, \omega) d^3 r = \frac{\int F_0(r, \omega) H_i(r, \omega) d^3 r}{1 - \int C(r, \omega) H_i(r, \omega) d^3 r}
\]  

(60)

which is used to compute a more precise solution

\[
F(r, \omega) = F_0(r, \omega) + C(r, \omega) \xi_i
\]  

(61)

We may iterate the cycle by setting \(F(r, \omega)\) as \(F_i(r, \omega)\) in the kernel and repeat the computation to further obtain a more precise \(F(r, \omega)\). In our computation we repeat the cycle up to 2 times as further iteration would not yield significantly more different or precise results.

Using the solutions of the local fields we compute the absorption spectra of the quantum system and the metallic particle as

\[
Q_{QS} = \frac{\varepsilon}{2\pi} \int \text{Im} \left[ \varepsilon_{QS}(r, \omega) \right] \left| \vec{E}_{QS}^0(r, \omega) \right|^2 dV
\]  

(62)

\[
Q_{MP} = \frac{\varepsilon}{2\pi} \int \text{Im} \left[ \varepsilon_{MP}(r', \omega) \right] \left| \vec{E}_{MP}^0(r', \omega) \right|^2 dV
\]  

(63)

which depend on the imaginary parts of \(\varepsilon_{QS}\) and \(\varepsilon_{MP}\). Thus, a negative sign would mean emission rate.

V. RESULTS AND DISCUSSIONS

In our simulations we have used the following parameters (unless stated otherwise): number density of molecules \(N_{qs} = 2 \times 10^{26} \text{ m}^{-3}\); characteristic radiative decay in soft/condensed matter \(\gamma_0 = 5 \times 10^{12} \text{ s}^{-1}\); coherence rates: \(\gamma_{ab} = \gamma_{de} = \gamma_{ac} = \gamma_0, \gamma_{bc} = 0.6\gamma_0\); detunings of the fields: \(\Delta_3 = 0 \text{ s}^{-1}, \Delta_4 = \Delta_5 = \Delta_6 = 0 \text{ s}^{-1}\); transition frequencies: \(\omega_{ab} = 3.586 \times 10^{15} \text{ s}^{-1}, \omega_{bc} = 3.54 \times 10^{15} \text{ s}^{-1}, \omega_{db} = 2.96 \times 10^{15} \text{ s}^{-1}, \omega_{dc} = 3.006 \times 10^{15} \text{ s}^{-1}, \omega_{bc} = 4.6 \times 10^{14} \text{ s}^{-1}, \omega_{ac} = \omega_{db} + \omega_{bc}\); Rabi frequencies for pump beam: \(\Omega_{p0} = 1.5\gamma_{ac}\); for Stokes beam,
due to approximations in Eqs. 8 and 6. Power $10$ is enhanced more than other side peaks, with the peak

$$\omega_{\text{peak}} = 10$$

is as high as $10^3$ times but the spectrum is not affected by plasmonic resonance. For medium particle size, the CARS process should give EIT peaks at $\omega_{\text{peak}}$ due to CARS and the slightly lower $\omega_{\text{side}}$. We also notice that the CARS peaks do not show up. We also notice that the CARS peaks are complicated by several factors: broadening, spatially inhomogeneous fields in the quantum particle and mesoscopic resonances as the result of Mie scattering as the size of the QS particle increases. Thus, any variation to the peaks is due to finite size effect. In the figures we have plotted, $Q_{\text{QS}}$ and $Q_{\text{MP}}$ are in unit of watts and we set the regime of validity as $D = 3(a + \mu_{\text{max}}) > 3\mu_{\text{max}}$ due to approximations in Eqs. 6 and 8.

**Spectra versus interparticle separation** Figures 2 and 3 show the variation of the power $Q_{\text{QS}}$ and $Q_{\text{MP}}$ with frequency, i.e. absorption spectra, for a range of interparticle distance $D$ and several values of particle radii $\rho = a = 10$ nm, 100 nm and 1 $\mu$m. For small particle size $\rho = a = 10$ nm and interparticle distance in the range $D = 60$ nm-140 nm, the spectral peaks in $Q_{\text{QS}}$ are similar to the Mollow’s triplets, with the principal peak at around $\omega \approx \omega_{\text{ac}}$ due to CARS and the slightly lower side peaks at $\omega_{\text{ac}} \pm (\Omega_{\rho0} + \Omega_{s0} + \Omega_{d0}) \approx \omega_{\text{ac}} \pm 5\gamma_{\text{ac}}$ due to ac Stark shifts by the laser fields in the Raman process. Secondary peaks spaced by $\gamma_{\text{ac}}$ are superimposed to the three main peaks. For $Q_{\text{MP}}$, the same peaks as in $Q_{\text{QS}}$ appear, but the central peak is lower than the side peaks. Here, $Q_{\text{MP}}$ is larger than $Q_{\text{QS}}$ but the spectrum is not affected by plasmonic resonance. For $Q_{\text{QS}}$ with the plasmonic resonance ($\omega_p = \omega_{\rho0}$) the central peak is enhanced more than other side peaks, with the peak power $10^4$ times higher than the peak without plasmonic resonance ($\omega_p = 0.9\omega_{\rho0}$).

For medium particle size $\rho = a = 100$ nm and interparticle distance in the range $D = 600$ nm-1400 nm, the peak power absorbed by the QS is typically $3 \times 10^8$ W, slightly larger than $Q_{\text{MP}}$, around $8 \times 10^8$ W, and the power drops with $D$. The multiple peaks in $Q_{\text{MP}}$ and $Q_{\text{QS}}$ almost disappear and are no longer resolved. However, a small dip starts to appear at $\omega = \omega_{\text{ac}}$ which is connected to nonlinear coupling with Fano effect and double resonance [35]. Plasmonic enhancement factor of $Q_{\text{QS}}$ is as high as $10^2$ times but $Q_{\text{MP}}$ is almost unaffected by the plasmonic effect.

For large particle size $\rho = a = 1$ $\mu$m and interparticle range $D = 6$ $\mu$m-14 $\mu$m, $Q_{\text{MP}}$ and $Q_{\text{QS}}$ show only a single dominant peak around $\omega_{\text{ac}}$. The plasmonic enhancement here is $3 \times 10^4$ times, slightly lower than the case of medium particle size. At $\omega_{\text{ac}} \pm (\Omega_{\rho0} + \Omega_{s0} + \Omega_{d0})$ the peaks have vanished and become the absorption minima, a clear sign of the Fano interference effect of the nonlinear coupling.

It is interesting that for medium and large particle radii, the logarithmic plots (on the right panels) show periodic oscillations at intervals of about $\lambda_d/2 = 263$ nm in the absorption power as $D$ varies. This interference effect happens only in the case of plasmonic resonance where standing waves of the CARS fields fit in between the two particles separated by $n\lambda_d/2$, multiples of half-wavelength of the $a-c$ transition, where $\lambda_d = 525.28$ nm.

**Stronger laser fields** We now look at right hand side panels in Figs. 2 and 3 with stronger laser fields: $\Omega_{\rho0} = 2\gamma_{\text{ac}}, \Omega_{s0} = 3\gamma_{\text{ac}}, \Omega_{d0} = 3\gamma_{\text{ac}}$. Optimization of the absorption power can be achieved by controlling the quantum coherence [37] using the pump, Stokes and control laser fields and this will be a subject of further analysis.

For small particles $\rho = a = 10$ nm the main peaks at $\omega_{\text{ac}} \pm (\Omega_{\rho0} + \Omega_{s0} + \Omega_{d0}) \approx \omega_{\rho0} \pm 5\gamma_{\text{ac}}$ are clearly visible and a smaller central peak at $\omega_{\text{ac}}$. Within the three main peaks there are smaller peaks that appear regularly spaced with period of $\gamma_{\text{ac}}$, as in the case of weaker fields. For $\rho = a = 100$ nm (Fig. 2), the plasmonic enhancement peak of QS is $5 \times 10^4$ times higher. The Fano dip does not appear here as it may have been covered by effects of the stronger laser fields. In the case $\omega_p = 0.9\omega_{\rho0}$ (without enhancement) we have negative absorption at $\omega = \omega_{\text{ac}}$ which indicates gain without population inversion since we verify that it is manifested by a finite but small population in level $b$ ($\rho_{bb} = 0.05, n_{ab} = 0.05$) coupled to the strong control laser. Unlike the gain in lasing without inversion (LWI) [38], the gain here is unique as it appears at certain values of $\rho$ and medium range of $D$ only. The negative absorption persists even at plasmonic resonance as well as the oscillations (in log scale) with period of $\lambda_d/2$.

For larger particles $\rho = a = 1$ $\mu$m, at plasmonic resonance $\omega_p = \omega_{\rho0}$, a few series of narrow and high peaks emerge in $Q_{\text{QS}}$ within the frequency range $\omega_{\text{ac}} \pm 2\gamma_{\text{ac}}$ and a small range of interparticle distance, indicating the effect of configurational resonance of the QS-MP system. Here, the absorption power goes as high as $4 \times 10^{16}$ W at distance around $D = 6$ $\mu$m, i.e. the power is enhanced by $10^4$ times relative to the case $\omega_p = 0.9\omega_{\rho0}$. The rate of the oscillations at plasmonic resonance remains the same as the weaker field case, i.e. with period of $\lambda_d/2$, half-wavelength of the $a-c$ transition.

Notice that the three ranges of $D$ in Figs. 2 and 3 are of different order of magnitudes: 60 nm, 600 nm and 6 $\mu$m while the peaks decay at similar rate. This shows that for larger particle sizes the plasmonic resonance does not only enhance the power but also gives long range interparticle interaction effect similar to the Förster interaction. Spectral peaks associated with the four-level system (Raman and CARS) are buried by the mesoscopic size effects for the cases of medium and large particle sizes.
The effects of the particle radius can be seen from a better perspective in Fig. 4 and Fig. 5 which show the variations of the power spectra $Q_{QS}$ and $Q_{MP}$, respectively, with particle radius $\rho$ in three ranges of $\rho$ for $D = 3(a + \rho_{\text{max}})$ with $a = 100$ nm and $\rho_{\text{max}}$ is the highest value for each range. The ranges are chosen to show the significant rise in the power at around 300 nm, 700 nm and 1600 nm. The plots of $\log Q_{QS}$ (Fig. 4) for the radius in small range $\rho = 10$nm $- 0.5$μm with plasmonic resonance condition indicates that the power increases very rapidly with $\rho$, almost as $10^{\sqrt{800\rho(\mu m)^{-8}}}$. For larger $\rho$ there is a saturation effect connected to the Mie focusing where the power increases slowly with $\rho$. Power enhancement is not significant as $\rho$ increases. The power oscillates in a quasi-periodic manner. At $\rho \approx 1$ μm (see plots for medium range $\rho = 0.5$ μm $- 1.2$ μm) and $D = 3.9$μm, the single peak breaks into two peaks with a dip near $\omega_{ac}$ as $\rho$ increases. This is similar to the Fano dip except that here it is due to the mesoscopic size effect. For larger particle size range, $\rho = 1.2$ μm $- 2$ μm, the peaks also drift to frequencies below $\omega_{ac}$. The drifting of peaks is due to the strong level shift by the intense laser fields formed by the focusing effect of the spherical particle. At $\rho \approx 1.4$ μm the frequencies of the double peaks suddenly jump back to around $\omega_{ac}$ and make a transition to a higher power as the focused intensity is diluted in the large particle while the capacity of absorption reaches a higher level. These features, also found in Fig. 5 for $Q_{MP}$, are associated with the Mesoscopic focusing effect that generates field intensity high enough to cause the spectral broadening, splitting and shifting. At plasmonic resonance $\omega_p = \omega_{ab}$, the spectral peaks appear in a non-regular fashion as $\rho$ increases. The plasmonic resonance does not affect the $Q_{MP}$ (Fig. 5) but significantly affects $Q_{QS}$ (Fig. 4) which shows narrow and high peaks and low peaks at certain frequencies and radii, corresponding to plasmonic-induced absorption and transparency, respectively. In particular, at $\rho \approx 420$ nm,$\rho \approx 800$ nm, $\rho \approx 1.9$ μm corresponding to the plasmonic enhancement factors of about $4 \times 10^5, 5 \times 10^4$ and $4 \times 10^4$ times, respectively.

In conclusion, we have developed a theory to compute the local CARS fields and absorption power of spherical QS and MP of finite sizes. We find from the log scale plots that the power scales very rapidly with $\rho$ and $a$ for small particle sizes. The rapid rise of power with particle size is an unexpected finding. It is as significant as the enhancement of narrow peaks at plasmonic resonance. The mesoscopic size of the particles can bury the spectral peaks and give rise to the new kind of Fano dip and shifted two peaks features that are very sensitive to the sizes of the QS and MP. Together with plasmonic resonance, very large and very small absorption powers at certain frequencies can be achieved for certain particle sizes. Also, interparticle spacing can accommodate a new kind of cavity resonance effect at plasmonic resonance. This work provides a useful theoretical and computational basis for further studies of realistic finite-size quantum plasmonic structures, particularly challenging endeavours to obtain exact numerical solutions and extend the theory to excitations by pulsed lasers with high intensity as well as few photons.

VI. APPENDIX A: POLARIZATION DUE TO CARS IN QS

The first term of the coherence in Eq. 3 is given by Eq. D4 of [31]

$$\tilde{\rho}_{ac}^{NL}(r, \omega) = \frac{i\Omega_c(r)\tilde{\rho}_{bc}(r, \omega - \nu_c)}{\Upsilon_{ac}^*(\omega)}$$

(64)

$$\Upsilon_{ac}^*(\omega) = \gamma_{ac} - i(\omega - \omega_{ac})$$

(65)

with the ground state coherence

$$\tilde{\rho}_{bc}(r, \omega - \nu_c) = \frac{\Omega_p(r)\Omega_s^*(r)}{\Upsilon_{bc}^*(\omega) + \delta_S(\omega) \left( \frac{n_{dc}}{\Upsilon_{dc}^*(\omega)} + \frac{n_{db}}{\Upsilon_{db}^*(\omega)} \right)}$$

(66)

$$\delta_S(\omega) = \frac{|\Omega_p|^2}{\Upsilon_{dc}^*(\omega)} + \frac{|\Omega_{s}^p|^2}{\Upsilon_{db}^*(\omega)}$$

(67)

$$\Upsilon_{dc}^*(\omega) = \gamma_{dc} - i(\Delta_p - i(\nu_c - \nu_d))$$

(68a)

$$\Upsilon_{db}^*(\omega) = \gamma_{db} + i(\Delta_p - i(\nu_c - \nu_d))$$

(68b)

$$\Upsilon_{bc}^*(\omega) = \gamma_{bc} - i(\Delta_p - i(\nu_c - \nu_d))$$

(68c)

where $n_{db} = \rho_{dd} - \rho_{bc}, n_{dc} = \rho_{dd} - \rho_{cc}$ are the population differences, $\Delta_p = \nu_p - \omega_{dc}, \Delta_s = \nu_s - \omega_{db}$ are the laser detunings, $\nu_4$ is the carrier frequency of the CARS signal taken to be $\omega_{ac}, \Omega_4$ and $\nu_4$ are the Rabi frequencies and carrier frequencies of the pump ($q = p$), Stokes ($q = s$) and control ($q = c$) lasers, $\omega_{nm}$ and $\gamma_{mn}$ are the atomic transition frequencies and decoherence rates between levels $m$ and $n (m, n = a, b, c, d)$.

VII. APPENDIX B: COHERENCE DUE TO A SCHEME IN QS

The second term in Eq. 3 is responsible for the EIT-type or A transition and has been defined by Eq. 73. Assuming the probe pulse comes after the pump and Stokes pulses are gone, we have the coherence and susceptibility for the EIT case

$$\tilde{\rho}_{ac}^A(r, \omega) = iF(r, \omega) \frac{n_{ab}I_c - n_{ac}J_b(\omega)}{J_b(\omega)\Upsilon_{ac}^* + I_T\Upsilon_{ab}^*(\omega)}$$

(69)

$$= iF(r, \omega) \Upsilon_{bc}^*(\omega) \frac{n_{ab}I_c - n_{ac}J_b(\omega)}{J_c(\omega)J_b(\omega) - I_cI_T}$$

(70)

$$\chi_{QS}^A(r, \omega) = \frac{\eta F(r)}{F(r)\tilde{\rho}_{ac}^A(r, \omega)}$$

(71)
\[ J_c(\omega) = \chi^*_c(\omega) \chi_b^c(\omega) + I_c \]  
(71)

\[ J_b(\omega) = \chi^*_b(\omega) \chi_c^b(\omega) + I_b \]  
(72)

where \( \eta = \frac{\hbar \omega_c}{4\eta n^2_{ac}} \), \( I_c(\mathbf{r}) = |\Omega_c(\mathbf{r})|^2 \), \( I_b(\mathbf{r}) = F(\mathbf{r})^2 \), \( \chi^*_c(\omega) = \gamma_{ac} + i\Delta_c - i(\omega - \nu_c), \chi^*_b(\omega) = \gamma_{bc} - i\Delta_c - i(\omega - \nu_b) \), and \( \nu_c = \nu_b = \nu_c - \nu_d \) and \( \omega_c = \omega_b = \omega_c - \omega_d \). The effective decoherences are \( \gamma_{ac} = \Gamma_a + \gamma_{ac}^d, \gamma_{bc} = \Gamma_b + \gamma_{ac}^d \), with \( \gamma_{ac}^d \) and \( \gamma_{bc}^d \) being the spontaneous emission rates, \( \Delta_c = \nu_c - \omega_c \) and \( \Delta_c = \nu_c - \omega_b \) are the detunings.

The dielectric function \( \varepsilon^{(1)}_Q \) in Eq. 4, susceptibility \( \chi^{(1)}_Q \) and \( \rho^{(1)}_a \) of the QS are obtained from \( \rho^{(1)}_a \) as

\[ \chi^{(1)}_Q(\mathbf{r}, \omega) = \rho^{(1)}_a(\mathbf{r}, \omega) \left|_{I_a=0} \right. \]

\[ = i F(\mathbf{r}) \frac{n_{ab} I_c - n_{ac} \chi^*_b(\omega)}{J_c(\omega) \chi^*_b(\omega)} \]  
(73)

\[ \varepsilon^{(1)}_Q(\mathbf{r}, \omega) = \chi^{(1)}_Q(\mathbf{r}, \omega) \left|_{I_a=0} \right. \]

\[ = i F(\mathbf{r}) \frac{n_{ab} I_c - n_{ac} \chi^*_b(\omega)}{J_c(\omega) \chi^*_b(\omega)} \]  
(74)

\[ \varepsilon^{(1)}_Q(\mathbf{r}, \omega) = \varepsilon_b[1 + \chi^{(1)}_Q(\mathbf{r}, \omega)] \]  
(75)
FIG. 2: Absorption power spectrum of QS versus interparticle distance $D$ for three values of particle radii $\rho = 10\,nm, 100\,nm$ and $1\,\mu m$ for off-resonant and resonant plasma frequencies $\omega_p$. Left two columns are for weaker laser fields: $\Omega_p = 1.5\gamma_{ac}$, $\Omega_s = 1.5\gamma_{ac}$, and $\Omega_c = 2\gamma_{ac}$). Right two columns are for stronger laser fields (right panels): $\Omega_p = 2\gamma_{ac}$, $\Omega_s = 3\gamma_{ac}$, and $\Omega_c = 4\gamma_{ac}$. Other parameters are described in the discussion section. For $\omega_p = \omega_p$ the log10 plot shows the enhanced narrow peaks more clearly.
FIG. 3: Same as in Fig. 2 except it is the absorption power spectrum of the MP plotted versus interparticle distance $D$. 

**Weaker fields**

- **Small particles:** $\rho=10\text{nm}; a=10\text{nm}$
  - $\omega_p=0.9\omega_0$
  - $\omega_p=\omega_0$
  - $\omega_p=0.9\omega_0$

**Stronger fields**

- **Medium particles:** $\rho=100\text{nm}; a=100\text{nm}$
  - $\omega_p=0.9\omega_0$
  - $\omega_p=\omega_0$
  - $\omega_p=0.9\omega_0$

- **Large particles:** $\rho=1000\text{nm}; a=1000\text{nm}$
  - $\omega_p=0.9\omega_0$
  - $\omega_p=\omega_0$
  - $\omega_p=0.9\omega_0$
FIG. 4: Absorption power spectrum for QS versus particle radius $\rho$ in three different ranges of $\rho$ for plasmonic off-resonant and resonant plasma frequencies $\omega_p$. The MP particle radius $a = 100\text{nm}$ and $D = 3(a + \rho_{\text{max}})$, $\rho_{\text{max}}$ is the maximum range of $\rho$ in the figures. Other parameters are described in the discussion section.
FIG. 5: Absorption power spectrum for MP versus three different ranges of particle radius $\rho$ for plasmonic off-resonant and resonant plasma frequencies $\omega_p$. Other parameters are the same as Fig. 4.