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Quantum theory of the nonlinear Fano effect in hybrid metal-semiconductor nanostructures: The case of strong nonlinearity

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We develop a quantum theory of the field-tunable nonlinear Fano effect in the hybrid metal-semiconductor nanostructures, in which the plasmon (semicontinuous collective intraband excitation) and the exciton (discrete single-particle interband excitation) are treated on the same footing. Our quantum theory shows that the quantum interference due to the plasmon-exciton interaction leads to the nonlinear Fano effect described by a generalized complex field-tunable Fano factor for the systems with strong external field and dephasing. We establish the relation between quantum and semiclassical theories and show that the results of the quantum and semiclassical theories differ both qualitatively and quantitatively in the strongly nonlinear regime—in particular, the quantum theory predicts the absence of nonlinear instability in the hybrid systems with plasmon relaxation.

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I. INTRODUCTION

Recently, there have been many theoretical and experimental studies on the hybrid nanostructures based on metal nanoparticles (MNPs) and molecules and/or semiconductor quantum dots (SQDs).[1–8] Building blocks of hybrid supernanostructures, metal and semiconductor nanoparticles, have very different electronic, optical, and thermal properties. The superstructures composed of nanoparticles take the advantages of the properties of their composite units and show many interesting phenomena, for instance, plasmon-induced fluorescence enhancement and quenching,[6] plasmon-assisted Forster energy transfer,[7] spin-plasmon interactions,[9] etc. Moreover, the hybrid systems have much more tunability for optical and nonlinear properties due to the exciton-plasmon interaction.

Nanoscale superstructures made from different materials are also interesting from the point of view of fundamental physics. The elementary excitation of MNPs is a plasmon, the intraband excitation of collective charge oscillation. While the elementary excitation of SQD is an exciton–the electron-hole pair, an interband excitation. Both plasmons and excitons play very important roles in modern nano-optics. The interaction among the elementary excitations of different natures in hybrid semiconductor-metal systems leads to interesting properties. For example, recent studies show that the exciton-plasmon interaction leads to the formation of a unique quasiparticle, a hybrid exciton with a renormalized frequency and a lifetime.[1] The long-range Coulomb interaction between excitons and plasmons leads to optimal hybrid nanostructures due to the competition between the field enhancement (dominant for large interparticle distances) and the quench effect (dominant for small interparticle distances).[2] Moreover, it was found that the plasmon-exciton interaction induces the nonlinear Fano effect,[1] which has already been observed in the experiments for the quantum-dot system with tunnel coupling[10] and in the plasmonic nanocrystal coupled with a polymer shell.[11] The nonlinear Fano effect is a generalization of the usual Fano effect[12] originally found in atomic and molecular systems. Much recent attention has been paid to the Fano effects in nanostructures, such as quantum dots[13,14] and plasmonic nanoparticles.[15] For a recent review, see Ref. 16.

Our previous studies on the nonlinear Fano effect in exciton-plasmon systems were based on the semiclassical theory,[1,2] where the exciton was described in the quantum theory framework, while the description of the plasmon was within the classical electromagnetic dynamics. The Fano effect in quantum systems is a consequence of quantum interference. Therefore, a quantum theory is needed for a reliable description of the nonlinear Fano effect. We note that, previously, a fully quantum theory for the nonlinear Fano effect in the quantum dots coupled with a continuum via a single-particle tunneling process was given in Ref. 10. In contrast to Ref. 10, we develop here a nonlinear theory for the exciton-plasmon systems with long-range Coulomb coupling between excitations of quite different natures, which have peculiar properties and are under active investigations in the current literature. In our theory, the exciton and plasmon are treated on the same footing. Importantly, we compare the semiclassical and quantum theories and, in particular, we show that the results coming from these theories are quantitatively and qualitatively different in the strong nonlinear regime. In contrast to the semiclassical theory, the quantum theory involving the fast relaxation of plasmons predicts no nonlinear instability and transparency effects in the nonlinear regime. Our quantum theory can be applied to other systems, where the quantum nature is important, for example, to the systems with a discrete spectrum showing the confined Fano effect.

II. THEORETICAL FORMULISM

We consider a hybrid nanoparticle molecule consisting of a SQD and a MNP in the presence of an external field $E = E_0\cos(\omega_0 t)$ [Fig. 1(a)]. The interaction between the exciton and plasmon is described by the Hamiltonian $\hat{H} = \hat{H}_0 + \hat{H}_{\rm int}$,

$$\hat{H}_0 = \varepsilon_0 c_0^+ c_0 + \varepsilon_e c_e^+ c_e - E\mu/\varepsilon_{\rm eff1}(c_0^+ c_e + c_e^+ c_0) + \sum_j \varepsilon_j c_j^+ c_j - E\sum_j \mu_j (c_0^+ c_j + c_j^+ c_0), \quad (1)$$

$$\hat{H}_{\rm int} = \sum_j (H_{je} c_j^+ c_e + H_{ej} c_e^+ c_j), \quad (2)$$

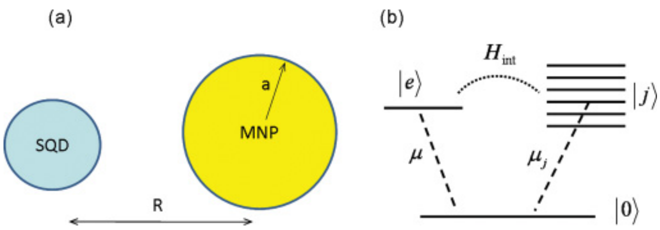

FIG. 1. (Color online) (a) Model and geometry of the hybrid system. (b) Schematic diagram of the system.

where c_0 is the annihilation operator for the common ground state, $c_e(c_j)$ is the annihilation operator for the excited state of the exciton (plasmon), $\mu(\mu_j)$ is the dipole between the ground state and exciton-excited state (plasmon-excited states), $\varepsilon_{\rm eff1} = (2\varepsilon_{ba} + \varepsilon_s)/(3\varepsilon_{ba})$ is the screening factor with ε_{ba}, ε_s being the dielectric constants of the background medium and SQD, respectively (the possible screening factor for MNP could be moved to the definition of μ_j), and H_{ej} is the interaction amplitude between the exciton and plasmon. The schematic diagram of the system is shown in Fig. 1.

The dynamics of the system is described by the equation of motion for the density matrix of the form

$$\frac{\partial\hat{\rho}}{\partial t} = -\frac{i}{\hbar}[\hat{H},\hat{\rho}] - \Gamma\cdot\hat{\rho},$$

where the last term describes possible dissipation effects (for instance, that from the spontaneous phonon emission).

Using the rotation wave approximation, we have the following equations:

$$\begin{aligned}
\frac{d\rho_{ee}}{dt} &= i\Omega_1(\bar{\rho}_{0e} - \bar{\rho}_{e0}) + (i/\hbar)\sum_j (H_{je}\rho_{ej} - H_{ej}\rho_{je}) \\
&\quad - \Gamma_e\rho_{ee},\\
\frac{d\bar{\rho}_{0e}}{dt} &= i(\omega_0 - \omega)\bar{\rho}_{0e} + i\Omega_1(\rho_{ee} - \rho_{00}) + (i/\hbar) \\
&\quad \times \sum_j H_{je}\bar{\rho}_{0j} + i\sum_j \Omega_j\rho_{je} - \Gamma_{0e}\bar{\rho}_{0e},\\
\frac{d\rho_{ej}}{dt} &= i(\omega_j - \omega_0)\rho_{ej} - i\Omega_j\bar{\rho}_{e0} + (i/\hbar)H_{ej}\rho_{ee} \\
&\quad - (i/\hbar)H_{ek}\rho_{kj} + i\Omega_1\bar{\rho}_{0j} - \Gamma_{ej}\rho_{ej},\\
\frac{d\bar{\rho}_{0j}}{dt} &= i(\omega_j - \omega)\bar{\rho}_{0j} + i\sum_k \Omega_k\rho_{kj} - i\Omega_j\rho_{00} \\
&\quad + (i/\hbar)H_{ej}\bar{\rho}_{0e} + i\Omega_1\rho_{ej} - \Gamma_{0j}\bar{\rho}_{0j},\\
\frac{d\rho_{jj}}{dt} &= i\Omega_j(\bar{\rho}_{0j} - \bar{\rho}_{j0}) \\
&\quad + (i/\hbar)(H_{ej}\rho_{je} - H_{je}\rho_{ej}) - \Gamma_j\rho_{jj},
\end{aligned} \tag{3}$$

where the slow variables are defined by $\rho_{ee} = \bar{\rho}_{ee}$, $\rho_{0e} = \bar{\rho}_{0e}e^{i\omega t}$, $\rho_{0j} = \bar{\rho}_{0j}e^{i\omega t}$, and $\rho_{ej} = \bar{\rho}_{ej}$. Also, $\Omega_1 = \mu E_0/2\hbar\varepsilon_{\rm eff1}$, $\Omega_j = \mu_j E_0/2\hbar$, and $\omega_j = (\varepsilon_j - \varepsilon_0)/\hbar$. We consider the steady-state solution for the case with a large dissipation in MNP [i.e., $\Gamma_{ej} \approx (\Gamma_e + \Gamma_j)/2 \approx \Gamma_j/2$, $\Gamma_{0j} \approx \Gamma_j/2$], and for the near resonant regime, i.e., $\omega \approx \omega_0$, we have $\gamma\rho_{ee} = -i\Omega_{\rm eff}\bar{\rho}_{e0} + \text{H.c.}$, $(\omega_0 - \omega + \gamma_{0e})\bar{\rho}_{0e} = \Omega_{\rm eff}\Delta$, where $\Delta = \rho_{00} - \rho_{ee}$ and $\Omega_{\rm eff} = \Omega_1 - \sum_j \frac{\Omega_j H_{je}}{\hbar\omega'_j}$ is the renormalized field felt by SQD, with $\Omega_R = \text{Re}(\Omega_{\rm eff})$, $\Omega_I = \text{Im}(\Omega_{\rm eff})$, $\omega'_j = \omega_j - \omega + i\Gamma_{0j}$, $\gamma_{0e} = \sum_j \frac{\Omega_j^2}{\omega_j'^*} - \sum_j \frac{|H_{ej}|^2}{\hbar^2\omega'_j} + \sum_j 4\text{Im}[\frac{1}{\hbar\omega'_j}]\frac{H_{je}\Omega_j\Omega_1}{\gamma} + i\Gamma_{0e}$, and $\gamma = \Gamma_e + \sum_j \frac{2|H_{ej}|^2\Gamma_{ej}}{\hbar^2[(\omega_j-\omega_0)^2+\Gamma_{ej}^2]}$. We can obtain the solution for ρ_{ee} and $\bar{\rho}_{0e}$ as

$$\begin{aligned}
\rho_{ee} &= \frac{Y|\Omega_{\rm eff}|^2}{K^2 + \tilde{\Gamma}_{0e}^2 + 2Y|\Omega_{\rm eff}|^2},\\
\bar{\rho}_{0e} &= \frac{(K\Omega_R + \tilde{\Gamma}_{0e}\Omega_I) + i(K\Omega_I - \tilde{\Gamma}_{0e}\Omega_R)}{K^2 + \tilde{\Gamma}_{0e}^2 + 2Y|\Omega_{\rm eff}|^2},
\end{aligned} \tag{4}$$

where $K \equiv \omega_0 - \omega + \delta, \delta = \text{Re}(\gamma_{0e}), \tilde{\Gamma}_{0e} = \text{Im}(\gamma_{0e})$, and $Y = 2\tilde{\Gamma}_{0e}/\gamma$. Because of a fast dissipation in MNP, we assumed above that $\rho_{00} + \rho_{ee} \approx 1$. It is easy to see the renormalized frequency and the lifetime for exciton, i.e., the behavior of the hybrid exciton.[1]

III. THE NONLINEAR FANO EFFECT

Now we consider the absorption of our hybrid system. Some algebraic calculation shows that the total energy absorption rate takes the form $Q_{\rm tot} = Q_{\rm MNP} + Q_{\rm SQD}$, with $Q_{\rm SQD} = \Gamma_e\rho_{ee}\hbar\omega_0$ and

$$Q_{\rm MNP} = \sum_j \Gamma_j\rho_{jj}\hbar\omega_j = \sum_j \frac{2\Gamma_{0j}\mu_j^2\hbar\omega_j}{(\omega_j - \omega_0)^2 + \Gamma_{0j}^2}\frac{E_0^2}{(2\hbar)^2}F(K), \tag{5}$$

$$F(K) = \frac{(K - \beta_R)^2 + (\tilde{\Gamma}_{0e} - \beta_I)^2 + [\beta^2(Y-1) + Y]|\Omega_{\rm eff}|^2}{K^2 + \tilde{\Gamma}_{0e}^2 + 2Y|\Omega_{\rm eff}|^2}, \tag{6}$$

where $\beta_R = \beta\Omega_R$, $\beta_I = \beta\Omega_I$, and $\beta = H_{ej}/\hbar\Omega_j$. $F(K)$ can be written as $F = \frac{|\varepsilon+q|^2}{1+\varepsilon^2} = \frac{(\varepsilon+q_R)^2}{1+\varepsilon^2} + \frac{q_I^2}{1+\varepsilon^2}$, $\varepsilon = \frac{\omega-\omega_0-\delta}{\sqrt{\tilde{\Gamma}_{0e}^2+2Y|\Omega_{\rm eff}|^2}}$, $q = q_R + iq_I$, $q_R = \frac{H_{ej}\Omega_R}{\hbar\Omega_j}\frac{1}{\sqrt{\tilde{\Gamma}_{0e}^2+2Y|\Omega_{\rm eff}|^2}}$, and $q_I = \sqrt{\frac{(\tilde{\Gamma}_{0e}-\beta_I)^2+[\beta^2(Y-1)+Y]|\Omega_{\rm eff}|^2}{\tilde{\Gamma}_{0e}^2+2Y|\Omega_{\rm eff}|^2}}$.

Usually $H_{ej} \propto \mu\cdot\mu_j$, then q_R, q_I is independent on the index *j*. In the case of a strong laser field, or a weak laser field with a short interparticle distance, the absorption rates of our hybrid systems are dominated by the absorption rate of MNP. It is because of the saturation effect of SQD (for strong laser field), and/or the fast dissipation in MNP (for the

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case of strong interparticle interaction, i.e., short interparticle distance). Since Γ_{0j} is large, the absorption line shape is mainly determined by $F(K)$, as also seen in Refs. 1 and 2.

In the weak field limit, the q_R term in $F(K)$ is electric-field independent and the usual linear Fano effect is recovered when $\Gamma_{0e}$is zero. The appearance of zero in the Fano function $F(K)$ at $\varepsilon = -q_R$ is a consequence of quantum interference. In the presence of dephasing (which is unavoidable in a real system), this zero is lifted due to the q_I term. Thus, the q_I term describes the effect of dephasing on the quantum interference. It is apparent, when Γ_{0e} is very large, that the q_I term dominates and the linear Fano effect is washed out. It is just because the discrete state is much broadened and becomes a wide or fat state, and the condition for the Fano effect (a discrete state or narrow state) is not satisfied.

A nonlinear Fano effect (with a field-dependent Fano factor q_R) appears for a strong external field, where the dipole of the SQD (proportional to $\mathrm{Re}[\bar{\rho}_{0e}]$) has a nonlinear dependence on the external electric field [see Eq. (4)]. Figure 2 and the above formula of q_R show that the external field makes the Fano factor q_R smaller, and thus makes the Fano effect more pronounced, i.e., the line shape is more asymmetric, which has been observed in experiment.[10] In general, q_R and q_I determine the field-dependent line-shape and dephasing effect. Here we also mention that the nonlinear Fano factor has a more complicated and interesting relation with the interaction strength compared to that for the usual linear Fano factor.

IV. THE QUANTUM-SEMICLASSICAL CORRESPONDENCE (QSC)

Now we establish the quantum-semiclassical relation. Consider the response of MNP to the external field $E = E_0 e^{-i\omega t}$. The induced polarization can be calculated as[17]

$$P(t) = e^{-i\omega t} \sum_j \frac{-|\mu_j|^2}{\hbar(\omega - \omega_j + i\Gamma_{0j})} E_0.$$

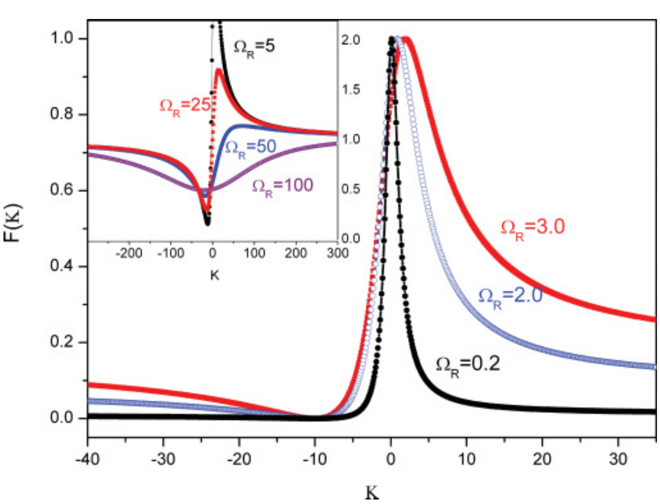

FIG. 2. (Color online) The generalized Fano function $F(K)$. $\beta_R = -10.0$, $Y = 1$. Ω_I has been set to zero for simplicity and $\tilde{\Gamma}_{0e}$ is set as the unit. We have neglected the weak field dependence of $\tilde{\Gamma}_{0e}$. The peak values have been normalized to 1 in the main panel. Insert: The generalized Fano function $F(K)$ in the regime of the very strong external field.

Therefore, we have

$$P(\omega)/E(\omega) = \sum_j \frac{-|\mu_j|^2}{\hbar(\omega - \omega_j + i\Gamma_{0j})} \equiv \varepsilon_{ba}\chi(\omega)a^3$$

for a spherical MNP with radius a, which corresponds to $\varepsilon_{ba}\frac{\varepsilon_m(\omega)-\varepsilon_{ba}}{\varepsilon_m(\omega)+2\varepsilon_{ba}}a^3$ in the classical theory, with ε_m the bulk dielectric constant of the metal. Consider the dipole interaction between a MNP and a SQD with $H_{ej} = -s_\alpha \mu \cdot \mu_j/\varepsilon_{\mathrm{eff2}}R^3$, where $\varepsilon_{\mathrm{eff2}} = (2\varepsilon_{ba} + \varepsilon_s)/3$, R is the interparticle distance, and $s_\alpha = 2$ (-1) for the exciton-dipole orientation parallel (perpendicular to) the axis of the hybrid molecule of SQD and MNP. Then we have

$$\sum_j \frac{|H_{ej}|^2}{\hbar(\omega_j - \omega - i\Gamma_{0j})} = \frac{s_\alpha^2 \mu^2 \chi(\omega) a^3}{\hbar \varepsilon_{\mathrm{eff1}}\varepsilon_{\mathrm{eff2}} R^6},$$

which is just the $G(\omega)$ defined in Ref. 1 accounting for the shift and broadening of the exciton peak. With the QSC, the physical meaning of some of the equations in the quantum theory becomes also quite clear. For instance, in the weak field regime, we obtain the following formula from Eq. (3):

$$\sum_j \mu_j \bar{\rho}_{0j} \propto \sum_j \mu_j \frac{\Omega_j - H_{ej}\bar{\rho}_{e0}/\hbar}{\omega\prime_j^*}$$
$$\propto \chi(\omega)a^3(E_0 + s_\alpha \mu \bar{\rho}_{0e}/\varepsilon_{\mathrm{eff2}}R^3). \qquad (7)$$

The left-hand side of the above equation is the dipole of MNP. The right-hand side is the dipole of MNP induced by the external field (the first term) and the dipole field from SQD (the second term, $P_{\mathrm{SQD}} \propto \mathrm{Re}[\mu\bar{\rho}_{0e}]$).

Using Eqs. (5) and (6) and the QSC, we can also obtain the absorption rate for MNP $Q_{\mathrm{MNP}} = \frac{3}{2}E_0^2 a^3 \omega |\frac{\varepsilon_{ba}}{2\varepsilon_{ba}+\varepsilon_m}|^2 \mathrm{Im}[\varepsilon_m]F(K)$ in the quantum theory with the Fano factor $q_R = A\sqrt{\Delta}$, $A = \frac{s_\alpha \mu^2 \Omega_R}{\hbar\varepsilon_{\mathrm{eff1}}\varepsilon_{\mathrm{eff2}}\Omega_1 R^3 \tilde{\Gamma}_{0e}}$. The semiclassical result is $q_C = A\Delta$, which agrees with the quantum result in the weak field regime $\Delta \approx 1$. It is easy to see that the semiclassical theory gives a smaller Fano factor than that in quantum theory ($\sqrt{\Delta} \geqslant \Delta$).

To further establish the relation between semiclassical theory and quantum theory and apply our theory to concrete material systems, we first calculated the energy absorption spectra for colloidal systems. In the calculation, we have used the parameters for colloidal systems with a background dielectric constant $\varepsilon_{ba} = 2.0$ (polymer) and $\varepsilon_{\mathrm{SQD}} = 7.2$ (CdTe); the constant ε_m (Au) is taken from Ref. 18. $R = 15$ nm, and $a = 7.5$ nm. In Fig. 3, we show the dependence of total energy absorption rate on the laser intensity and dipole moments of excitons ($\mu = er_0$). It is clearly seen that in the case with a weak field or strong field off-resonance ($\Delta \approx 1$), there is good agreement for the total absorption rate between the semiclassical theory and the quantum theory. For the case with a strong field near resonance (Δ is small), the quantum results are quite different from the semiclassical results. In general, the absorption line shape in quantum theory looks more symmetric than that in the semiclassical theory, which is consistent with the fact that $q_R > q_C$. Importantly, in the case of a strong external field and large exciton dipole moment [as shown in Fig. 3(f)], the absorption spectrum from the semiclassical theory shows instabilities in the small energy

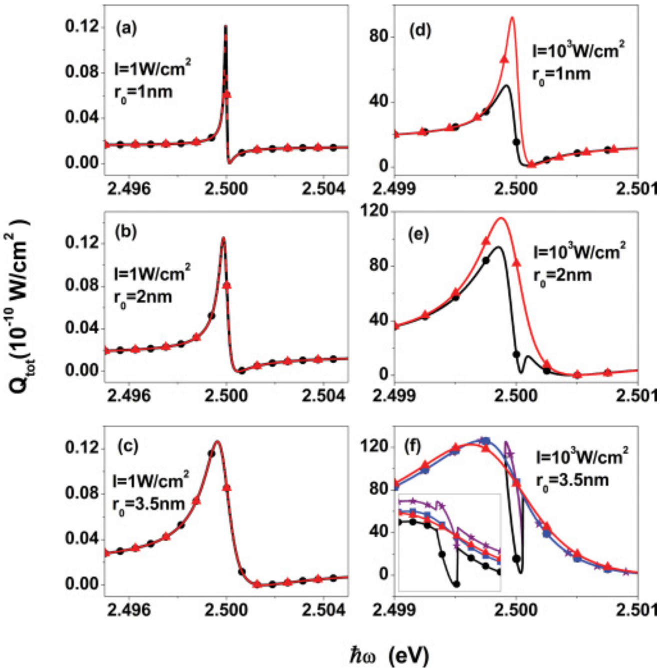

FIG. 3. (Color online) Total energy absorption rates obtained from the semiclassical theory (black curves with dots) and the quantum theory (red curves with triangles) for different values of dipole moment of SQD. (a)–(c) are for the weak field regime with different exciton dipole moments. (d)–(f) are for the strong field regime with different exciton dipole moments. In (f), the black curve with dots, the blue curve with squares, and the purple curve with stars show the three nonlinear steady states from the semiclassical theory. The inset in (f) is the magnification of the near resonance regime; in this graph, we have shifted the curves to make them clearer.

window near the resonance, as predicted before in Ref. 4. In this near resonance regime, there are three steady-state solutions (represented by the black curve with dots, the blue curve with squares, and the purple curve with stars) with different values of the exciton excited state population and total absorption rate for each frequency. Beyond this energy window, these three curves collapse to one curve, seen as the blue curve with squares. This instability is described by three nonlinear steady states [black curve with dots, blue curve with squares, and purple curve with stars in Fig. 3(f)] and one of the states [black curve with dots in Figs. 3(e) and 3(f)] shows zero total absorption that is an induced transparency effect. The absorption spectrum from the quantum theory (red lines with triangles) does not show instabilities and a transparency effect. The physical reason for the absence of the instability and transparency effects in the quantum theory is that the continuum states cannot be heavily populated since the population nonlinearity for the metal component cannot be achieved at the considered moderate light powers.

Figure 4 shows the comparison between quantum theory and semiclassical theory for the self-assembled systems. There is little difference between the total absorption rate from semiclassical theory and that from quantum theory in the weak field regime. In the strong field regime, a large difference between the two theories appears. The absorption has an

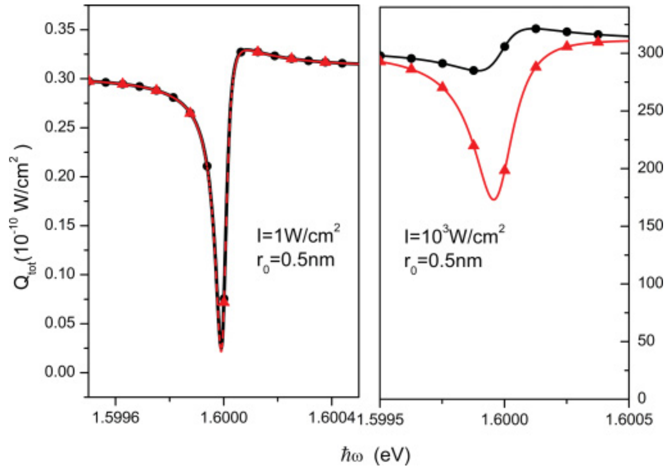

FIG. 4. (Color online) The total energy absorption rates obtained from semiclassical theory (the black curves with dots) and quantum theory (the red curves with triangles) for self-assembled systems with parameters$\varepsilon_{ba} = \varepsilon_{\rm SQD} = 12$.

asymmetric line shape in the semiclassical theory, while it develops antiresonance in the quantum theory.

V. THE NONLINEAR CONFINED FANO EFFECT

Our theory of the nonlinear Fano effect is based on the Hamiltonian [Eqs. (1) and (2)] with a general interaction term, and thus can be applied to more general systems. In particular, it can be applied to systems where the quantum nature is essential and the semiclassical theory is invalid. Bar-Ad *et al.* have studied the confined Fano effect,[19] where the spacing between the energy levels in the "semicontinuous" spectrum may become large due to the confinement effect. In this case, a full quantum theory is needed. If we consider the "picket fence" model[19,20] with $\omega_j = j\Delta\varepsilon$, $-\infty < j < \infty$, assume $\mu_j = \bar{\mu}$, $H_{ej} = V$ to be constants, and neglect the small Γ_{0e}, a simple calculation shows that $\tilde{\Gamma}_{0e} = \pi(|V|^2/\hbar^2 + \Omega_1^2\bar{\mu}^2/\mu^2)/\Delta\varepsilon$ and $q_R = V\mu/\hbar\bar{\mu}\sqrt{\tilde{\Gamma}_{0e}^2 + 2Y|\Omega_{\rm eff}|^2}$, which reduces to $q_0 = \frac{\hbar\mu\Delta\varepsilon}{\pi\bar{\mu}V}$ in the case $E_0 \to 0$. The field-independent confined Fano factor q_0 found in Ref. 19 is recovered. q_0 is proportional to $\Delta\varepsilon$. With increasing $\Delta\varepsilon$, q_0 increases and the asymmetric line shape changes to symmetric line shape gradually, and thus the Fano effect tends to disappear. In general, the linear dependence of q_R on $\Delta\varepsilon$ becomes nonlinear in the presence of an external field and q_R is always smaller than q_0. Here one sees that the competition between the confinement effect and nonlinear effect leads to the field-tunable nonlinear confined Fano effect. The confinement induces enhancement of the Fano factor on one side, and the external field reduces the Fano factor by decreasing $\Delta\varepsilon$ effectively on the other side, increasing the possibility of observing an asymmetric Fano line shape—the confined Fano effect.[19]

Finally, we would like to discuss the parameters. The largest parameter in the case we considered is the damping rate of plasmon. It is ∼10–100 meV, which is much larger than the dissipation rates of the exciton $\Gamma_e \approx \mu$eV, $\Gamma_{0e} \approx \mu$eV. For the case of weak field, for instance, $I \approx 1$–10 W/cm^2, $\hbar|\Omega_{\rm eff}|(\approx\mu{\rm eV}) \ll \tilde{\Gamma}_{0e}$ ($\approx$10 μeV), the system basically shows the linear behavior (with $\Delta \approx 1$). For a stronger field with $I \approx 1000\ \mathrm{W/cm^2}$, i.e., $\hbar|\Omega_{\mathrm{eff}}| \approx 10\ \mu\mathrm{eV}$ ($\ll$meV). In this case, $\tilde{\Gamma}_{0e}^2 \approx 2|\Omega_{\mathrm{eff}}|^2$, the nonlinear effect shows up and Δ has an appreciable difference from 1, indicating that the quantum effect becomes important. We also want to point out that our quantum theory can be applied to cases where the semiclassical treatment of the Fano problem is invalid. In our quantum approach, we can still use in our theory explicit information about an electronic structure of the materials. Our theoretical framework for the nonlinear Fano effect can be applied to other experimental systems, such as a metallic tip–quantum dot and/or molecule system.

To conclude, we present a quantum theory for the hybrid MNP-SQD molecules, where the elementary excitations of very different natures, the exciton and plasmon, are treated on the same footing. The nonlinear Fano effect is described by the Fano function $F = \frac{|\varepsilon+q|^2}{1+\varepsilon^2}$ with the generalized field-dependent complex Fano factor $q = q_R + iq_I$, which includes both the nonlinear and dephasing effects. We show that the results from quantum theory differ from those of semiclassical theory, especially in the strong field regime. For example, in contrast to the semiclassical theory, the quantum treatment of the exciton-plasmon Fano problem points out that there are no bistable states in the strongly nonlinear regime. Our general theoretical framework can be applied to other hybrid systems with strong confinement, for instance, those with an interesting nonlinear confined Fano effect.

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