Slow Light in Artificial Hybrid Molecules

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Abstract

The optical properties of hybrid molecules composed of semiconductor and metal nanoparticles with a weak probe in a strong pump field are investigated theoretically. Excitons in such a hybrid molecule demonstrate novel optical properties due to the coupling between exciton and plasmon. It is shown that a non-absorption hole induced by coherent population oscillation appears at the absorption spectrum of the probe field and there exists slow light effect resulting in the great change of the refractive index. The numerical results indicate that with the different center-to-center distance between the two nanoparticles the slow light effects are greatly modified in terms of exciton-plasmon couplings.

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I. INTRODUCTION

Due to the rapid advances of nanotechnology, it is possible to combine nanocrystal of various material with very different characteristics in one superstructures\[1, 2, 3\], then the investigation on the properties of artificial hybrid molecules has attracted much interest in recent years\[4, 5, 6, 7, 8\]. One of the central problems related to these hybrid complex is the interaction between the nanoscale building blocks. If individual nanoparticles do not exchange carriers, then the interparticle Coulomb interaction becomes the main mechanism of coupling and can strongly change the physical properties of these nanoscale building blocks. On the other hand, there is a great deal of researches devoted to investigate the slow light based on electromagnetically induced transparency (EIT)\[9, 10, 11\] and coherent population oscillation (CPO)\[12\] in various of potential materials. During the last decade, coherent population oscillation(CPO) approach have proven to be a power technique that can eliminate the absorption at the resonant frequency of transition and dramatically change the refractive index which leads to the slowdown of light speed in various of mediums\[13, 14, 15\]. One of the more effective thing is that the slow light based on the CPO approach can be achieved at room temperature, comparing with that based on electromagnetically induced transparency method. But most materials mentioned above are only focused on the conventional materials, such as optical fiber, Pb vapours, room-temperature solid and low dimensional semiconductors. The investigation of slow light in hybrid complex is obviously different from that of the conventional low dimensional semiconductors, then it is hoped that some novel optical properties will be found in the hybrid complex.

In the present paper, we will study the slow light in hybrid complex composed of a metal nanoparticle(MNP) and a semiconductor quantum dot(SQD). Elementary excitations in a SQD and a MNP, an exciton and a plasmon, have very different properties. The basic excitations in the MNP are surface plasmons with a continuous spectrum. In SQDs, the excitations are discrete interband excitons. There is no direct tunneling between the MNP and the SQD. However, long-range coulomb interaction couples the excitons and plasmons\[16\]. The effect of coupling between excitons and plasmons can strongly alter the optical properties of superstructure compared to a single SQD.

The structure of the article is as follows: In Sec.II, the theoretical model is given. The numerical results are presented in Sec.III. In Sec.IV, a summary of our results is given.
II. THEORY

We now consider a hybrid molecule composed of a spherical MNP with radius $a$ and a spherical SQD with radius $r$ with a weak probe in a strong pump field. The center-to-center distance between the two nanoparticles is $R$. For the SQD, we assume a two-level model for such a SQD which consists of the ground state $|0\rangle$ and the first excited state (single exciton) $|1\rangle$. The two-level scheme with coherent drive in atomic systems has been studied by Yu et al.\cite{17} which measured the emission spectrum of two-level-like Ba atoms driven by a continuous wave bichromatic field. The hybrid molecule via exciton interacts with a probe field of the frequency $\omega_s$ and is coherently driven by a strong control field of frequency $\omega_c$. As usual, this two-level system can be characterized by the pseudospin $\frac{1}{2}$ operators $S^\pm$ and $S^z$. Then the Hamiltonian of the system in a rotating frame at the control field frequency $\omega_c$ reads as follows:

$$
H = \hbar(\omega_{ex} - \omega_c)S^z - \hbar\Omega(S^+ + S^-) - \mu(S^zE_s e^{-i\delta t} + S^- E^*_s e^{i\delta t}),
$$

(1)

where $\hbar\omega_{ex}$ is the energy of exciton binding, $\Omega = \mu E_{SQD}/\hbar$, $\mu$ is the interband dipole matrix element, $E_{SQD}$ is the total field felt by the SQD and is given by

$$
E_{SQD} = E_c + \frac{S_\alpha P_{MNP}}{\varepsilon_{eff1} R^3},
$$

(2)

with $\varepsilon_{eff1} = \frac{2\varepsilon_m + \varepsilon_s}{3\varepsilon_0}$, $\varepsilon_0$ and $\varepsilon_s$ are the dielectric constants of the background medium and SQD, respectively, $E_c$ is the slowly varying envelope of the control field and $S_\alpha = 2$ for electric field polarizations $\alpha = z$ which direction corresponds to the axis of the hybrid molecule. The dipole $P_{MNP}$ comes from the charge induced on the surface of the MNP. It depends on the total electric field with superposition of the control and the dipole field due to the SQD, and is given by

$$
P_{MNP} = \gamma a^3(E_c + \frac{S_\alpha P_{SQD}}{\varepsilon_{eff2} R^3}),
$$

(3)

where $\gamma = \frac{\varepsilon_m(\omega) - \varepsilon_0}{3\varepsilon_0 + \varepsilon_m(\omega)}$, $\varepsilon_{eff2} = \frac{2\varepsilon_m + \varepsilon_s}{3}$, $\varepsilon_m(\omega) = 1 - \frac{\omega_p^2}{\omega^2}$ is the dielectric constant of the metal, and $\omega_p$ is the surface plasmon frequency of the MNP. The dipole moment of the SQD is expressed via the off-diagonal elements of the density matrix: $P_{SQD} = \mu S^-$. $E_s$ is the slowly varying envelope of the probe field. $\delta = \omega_s - \omega_c$ is the detuning of the probe and the control field. $\omega_s$ is the frequency of the probe field. The temporal evolutions of the exciton on the SQD are determined by the Heisenberg equation of motion, and are given by
\[ \frac{dS^-}{dt} = -i\Delta S^- - 2i\Omega S^z - \frac{2i\mu}{\hbar} S^z E_s e^{-i\delta t}, \quad (4) \]
\[ \frac{dS^z}{dt} = i\Omega(S^+ - S^-) + i\mu(S^+ E_s e^{-i\delta t} - S^- E^*_s e^{i\delta t}), \quad (5) \]

where \( \Delta = \omega_{ex} - \omega_c \). In what follows we ignore the quantum properties of \( S^- \), \( S^z \), then the semiclassical equations read as follows

\[ \frac{dS^-}{dt} = \left( -\frac{1}{T_2} - i\Delta \right) S^- - 2i\Omega S^z - \frac{2i\mu}{\hbar} S^z E_s e^{-i\delta t}, \quad (6) \]
\[ \frac{dS^z}{dt} = \frac{1}{T_1}(S^z + \frac{1}{2}) + i\Omega(S^+ - S^-) + i\mu(S^+ E_s e^{-i\delta t} - S^- E^*_s e^{i\delta t}), \quad (7) \]

where \( T_1 \) is the exciton lifetime, \( T_2 \) is the exciton dephasing time. For simplicity we define \( p = \mu S^- \), \( w = 2S^z \), and then we have

\[ \frac{dp}{dt} = -(\frac{1}{T_2} + i\Delta)p - \frac{i\mu^2}{\hbar} wE, \quad (8) \]
\[ \frac{dw}{dt} = \frac{1}{T_1}(w + 1) + 4Im(pE^*), \quad (9) \]

where \( E = E_{SQD} + E_s e^{-i\delta t} \). In order to solve Eqs.(8) and (9), we make the ansatz \[22\]

\[ p = p_0 + p_1 e^{-i\delta t} + p_{-1} e^{i\delta t}, \quad (10) \]
\[ w = w_0 + w_1 e^{-i\delta t} + w_{-1} e^{i\delta t}, \quad (11) \]

on substituting Eqs.(10) and (11) into Eqs.(8) and (9) and on working to the lowest order in \( E_s \) but to all orders in \( E_c \), we obtain in the steady state:

\[ 0 = -(\frac{1}{T_2} + i\Delta)p_0 - \frac{i\mu^2 B}{\hbar} w_0 p_0 - \frac{i\mu^2 A}{\hbar} E_c, \quad (12) \]
\[ -i\delta p_1 = -(\frac{1}{T_2} + i\Delta)p_1 - \frac{i\mu^2 B}{\hbar} (w_0 p_1 + w_1 p_0) - \frac{i\mu^2}{\hbar} (A_w E_c + w_0 E_s), \quad (13) \]
\[ i\delta p_{-1} = -(\frac{1}{T_2} + i\Delta)p_{-1} - \frac{i\mu^2 B}{\hbar} (p_0 w_{-1} + p_{-1} w_0) - \frac{i\mu^2 A}{\hbar} w_{-1} E_c, \quad (14) \]
\[ 0 = -\frac{1}{T_1}(w_0 + 1) + \frac{2i}{\hbar} [A(p_0 E_c - p_0 E^*_c)], \quad (15) \]
\[ -i\delta w_1 = -\frac{1}{T_1} w_1 + \frac{2i}{\hbar} [A(p_{-1} E_c - p_{-1} E^*_c) + B(p_{-1} p_0 - p_1 p_0^*) + p_0^* E_s], \quad (16) \]
\[ i\delta w_{-1} = -\frac{1}{T_1} w_{-1} + \frac{2i}{\hbar} [A(p_1 E_c - p_{-1} E^*_c) + B(p_1^* p_0 - p_{-1} p_0^*) - p_0 E^*_s], \quad (17) \]
where \( A = 1 + \frac{\gamma a^3 S^2}{\varepsilon_{\text{eff}} R^2} \), \( B = \frac{\gamma a^3 S^2}{\varepsilon_{\text{eff}} R^2} \).

From the solution of Eqs. (12)-(17), we can yield

\[
W_1 = -\frac{2\mu^2 A w_0 E_s E_c^*}{D(\delta_c) T_2^2} [2 - i(\delta_c + B_c w_0)] [1 + i(\Delta_c + B_c w_0)] [(1 - i(\delta_c + \Delta_c + B_c w_0)],
\]

\[
p_1 = \frac{\mu^2 E_s T_2}{\hbar [1 + i(\Delta_c - \delta_c + B_c w_0)]} \times \left\{ -i w_0 + \frac{2A^2 \Omega^2 w_0}{D(\delta_c)} (1 + i\Delta_c) [1 - i(\Delta_c + \delta_c + B_c w_0)] \right\},
\]

where

\[
D(\delta_c) = (2 - i \delta_c) [1 + (\Delta_c + B_c w_0)^2][(1 - i \delta_c)^2 + (\Delta_c + B_c w_0)^2] + 4A^2 \Omega^2 (1 - i \delta_c)(1 + \Delta_c)^2;
\]

where \( T_1 = \frac{T_2}{2}, \delta_c = \delta T_2, \Omega^2_c = \mu^2 |E_c|^2 T_2^2, \Delta_c = \Delta T_2, B_c = \frac{\omega^2 B}{\hbar} T_2. \) The linear optical susceptibility can be expressed by

\[
\chi^{(1)}_{\text{eff}} = N \frac{p_1}{E_s} = \frac{N \mu^2 T_2}{\hbar} \chi^{(1)}(\omega_s);
\]

where \( N \) is the number density of hybrid molecule and the dimensionless susceptibility is given by

\[
\chi^{(1)}(\omega_s) = \frac{1}{\hbar [1 + i(\Delta_c - \delta_c + B_c w_0)]} \times \left\{ -i w_0 + \frac{2A^2 \Omega^2 w_0}{D(\delta_c)} (1 + i\Delta_c) [1 - i(\Delta_c + \delta_c + B_c w_0)] \right\},
\]

The population inversion of exciton is determined by equation:

\[
w_0 = \frac{-2\Omega^2 w_0}{1 + (\Delta_c + B_c w_0)^2} - 1.
\]

The cubic equation has either a single root or three real roots. The latter case just corresponds to the intrinsic optical bistability which arises from the Coulomb interaction.

In terms of this model, we can also determine the light group velocity\[23, 24\]

\[
v = \frac{c}{n + \omega_s (dn/d\omega_s)},
\]
where $n \approx 1 + 2\pi \chi$, and then

$$\frac{c}{v_g} = 1 + 2\pi \text{Re} \chi(\omega_s)_{\omega_s=\omega_{ex}} + 2\pi \text{Re} \left( \frac{d\chi}{d\omega_s} \right)_{\omega_s=\omega_{ex}}.$$  \hfill (25)

It clear from this expression for $v_g$ that when $\text{Re} \chi(\omega_s)_{\omega_s=\omega_{ex}}$ is zero and the dispersion is very steep and positive, the group velocity is significantly reduced, and then

$$\frac{c}{v_g} - 1 = \frac{2\pi \omega_{ex} N \mu^2 T_2}{\hbar} \text{Re} \left( \frac{d\chi}{d\omega_s} \right)_{\omega_s=\omega_{ex}} = \frac{\sum}{T_2} \text{Re} \left( \frac{d\chi}{d\omega_s} \right)_{\omega_s=\omega_{ex}},$$  \hfill (26)

where

$$\sum = \frac{2\pi \omega_{ex} N \mu^2 T_2^2}{\hbar}.$$

A weak pulse propagating through a distance $L$ with a reduced group velocity is delayed as compared with propagation in free space by the time delay $T_g$, which is often the quantity measured in experiments,

$$T_g = \frac{L}{c} \left( \frac{c}{v_g} - 1 \right) = \frac{2\pi \omega_{ex} N L \mu^2 T_2}{c \hbar} \text{Re} \left( \frac{d\chi}{d\omega_s} \right)_{\omega_s=\omega_{ex}},$$  \hfill (27)

where $c$ is the free-space speed of light.

### III. NUMERICAL RESULTS AND DISCUSSIONS

In the following, as an example, we consider a Au MNP with radius $a = 7.5nm$ and the dielectric constants of the background medium and SQD: $\varepsilon_0 = 1$, and $\varepsilon_s = 6$. For the relaxation time and dipole moment in SQD, we take $T_2 = 0.8ns$ and $\mu = e r_0$, with $r_0 = 0.65nm$.

Figure 1 shows the effect of $B_c$ due to the distance between SQD and NMP and the dielectric constants of the metal and the semiconductor quantum dot on the linear absorption spectrum for three cases of the control field ($\Omega_c^2$). It is clear from the figure that the absorption peak moves along to the larger $\Delta_s (\Delta_s = (\omega_s - \omega_{ex})T_2)$ with the larger value of $B_c$. This implies that the SQD and the NMP close more and the Coulomb interaction between them becomes stronger. As a result, the Stark shift is significant.

The linear optical absorption $\text{Im} \chi^{(1)}$ as a function of the detuning $\Delta_s$ for the case $\Delta_c = 1.5$, $B_c = -0.1$ is presented in Figure 2. In the figure the dotted line is a normal excitonic absorption in the hybrid complex system as the control field is absent ($\Omega_c^2 = 0$). When we turn on the control field ($\Omega_c^2 = 5$), a non-absorption hole appears at $\Delta_s = 0$, thus the
system becomes transparent for the probe field. The hole is induced by coherent population oscillation which is mediated by the plasmon-exciton interaction. The physical reason is that the control field pump the electrons from the ground state to the first excited state. Turning on the probe field, the probe field will adjust the amplitude of the control field. The adjust function of the probe field and the long dephasing time $T_1$ will make the electronic population oscillating with the beat frequency $\delta$ between two energy levels. This coherent population oscillation can lead to the decrease of the absorption of the probe field. But here the plasmon effects will modify such a CPO and is beneficial to the slow light (also see Fig.6 below). Figure 3 demonstrates the behavior of the real part of the linear optical susceptibility $\chi^{(1)}$ as a function of the detuning $\Delta_s$ for $\Delta_c = 1.5, B_c = -0.1, \Omega_c^2 = 5$. At $\Delta_s = 0$, there is a steep slope corresponding to a large dispersion, which results in slow group velocity without absorption which shows in the solid line in Figure 2.

Figure 4 illustrates the imaginary part of $\chi^{(1)}$ as a function of $\Delta_s$ with $\Omega_c^2 = 5, B_c = -0.1$ for the three values of $\Delta_c$. Only $\Delta_c = 1.5$, the transparency will occur at $\Delta_s = 0$. This condition is important in the formation of the transparency at the center of the exciton absorption, otherwise the transparency will disappear at the center of exciton absorption. The imaginary part of the linear optical susceptibility $\chi^{(1)}$ as a function of the detuning $\Delta_s$ for the three values of $\Omega_c^2$ with $\Delta_c = 1.5, B_c = -0.1$ is shown in Figure 5(a). At the resonant condition $\Delta_s = 0$, the linear absorption decreases and the transparency window of the absorption spectrum between two absorption peaks becomes broader with larger value $\Omega_c^2$. Figure 5(b) shows the real part of the linear optical susceptibility $\chi^{(1)}$ as a function of the detuning $\Delta_s$ for the three value of $\Omega_c^2$ with $\Delta_c = 1.5, B_c = -0.1$. At $\Delta_s = 0$, the slope of refractive index can be flatter and the width between two peaks can be broader by increasing $\Omega_c^2$.

The group velocity index $n_g$ as a function of the Rabi frequency $\Omega_c^2$ with $\Delta_c = 1.5$ is plotted (in units of $\Omega$) for three value of $B_c$ in the resonance case ($\Delta_s = 0$) in Figure 6. It is shown that the group velocity is very sensitive to the Rabi frequency of the control field and the exciton-plasmon interaction. The slope of the dispersion become steeper as $\Omega_c^2$ decreases, leading to an increasingly group velocity. The value of $B_c$ is smaller, which corresponds to the interparticle interaction $R$ is larger, so the local field resulting in the Coulomb interaction is weaker. Because of the negative value of $B_c$, this local field will make the total field weaken. So the total field felt by the SQD is weaker with the larger
absolute value of $B_c$. That is the reason of smaller group velocity index with smaller absolute value of $B_c$.

IVA. CONCLUSIONS

In conclusion, the slow light based on the coherent population oscillation mediated by exciton-plasmon interaction in hybrid complex is investigated theoretically. The numerical results show that the exciton-plasmon interaction have a significant effect on the absorption spectrum and the dispersion spectrum and slow light can be achieved in such a hybrid complex. Finally, we hope that the proposed effect of this work will be testified in the near future experiments.

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**Figure Captions**

Fig 1 The imaginary part of the linear optical susceptibility as a function of the detuning $\Delta_s$ with parameters $\Delta_c = 1.5, \Omega_c^2 = 0$, for $B_c = 0, -1, -7$.

Fig 2 The imaginary part of the linear optical susceptibility as a function of the detuning $\Delta_s$ with parameters $\Delta_c = 1.5, B_c = -0.1$, for $\Omega_c^2 = 0, 5$.

Fig 3 The real part of the linear optical susceptibility as a function of the detuning $\Delta_s$ with parameters $\Delta_c = 1, B_c = -0.1, \Omega_c^2 = 5$.

Fig 4 The imaginary part of the linear optical susceptibility as a function of the detuning $\Delta_s$ with parameters $\Omega_c^2 = 5, B_c = -0.1$, for $\Delta_c = 1, 1.5, 2$.

Fig 5.a The imaginary part of the linear optical susceptibility as a function of the detuning $\Delta_s$ with parameters $\Delta_c = 1, B_c = -0.1$, for $\Omega_c^2 = 6, 7, 8$.

Fig 5.b The real part of the linear optical susceptibility as a function of the detuning $\Delta_s$ with parameters $\Delta_c = 1, B_c = -0.1$, for $\Omega_c^2 = 6, 7, 8$.

Fig 6 The group velocity index $n_g (= c/v_g)$ (in units of $\Sigma$) as a function of the detuning $\Delta_s$ with parameters $\Delta_c = 1.5, B_c = -0.1, -0.3, -0.5$. 


\[ \Omega_c^2 = 5 \]
\[ \Omega_c^2 = 0 \]
\[ \Omega_c^2 = 5 \]
\[ \Delta \text{c} = 1 \quad \Delta \text{c} = 1.5 \quad \Delta \text{c} = 2 \]
Detuning $\Delta_s$, $\Omega_c^2 = 6$, $\Omega_c^2 = 7$, $\Omega_c^2 = 8$
\[ \Omega_c^2 = 6 \]
\[ \Omega_c^2 = 7 \]
\[ \Omega_c^2 = 8 \]
Detuning $\Delta_s$