Ο **Nanoscale Research Letters**
a SpringerOpen Journal

NANO EXPRESS   **Open Access**

Strong coupling among semiconductor quantum dots induced by a metal nanoparticle

Yong He and Ka-Di Zhu[*]

Abstract

Based on cavity quantum electrodynamics (QED), we investigate the light-matter interaction between surface plasmon polaritons (SPP) in a metal nanoparticle (MNP) and the excitons in semiconductor quantum dots (SQDs) in an SQD-MNP coupled system. We propose a quantum transformation method to strongly reveal the exciton energy shift and the modified decay rate of SQD as well as the coupling among SQDs. To obtain these parameters, a simple system composed of an SQD, an MNP, and a weak signal light is designed. Furthermore, we consider a model to demonstrate the coupling of two SQDs mediated by SPP field under two cases. It is shown that two SQDs can be entangled in the presence of MNP. A high concurrence can be achieved, which is the best evidence that the coupling among SQDs induced by SPP field in MNP. This scheme may have the potential applications in all-optical plasmon-enhanced nanoscale devices.
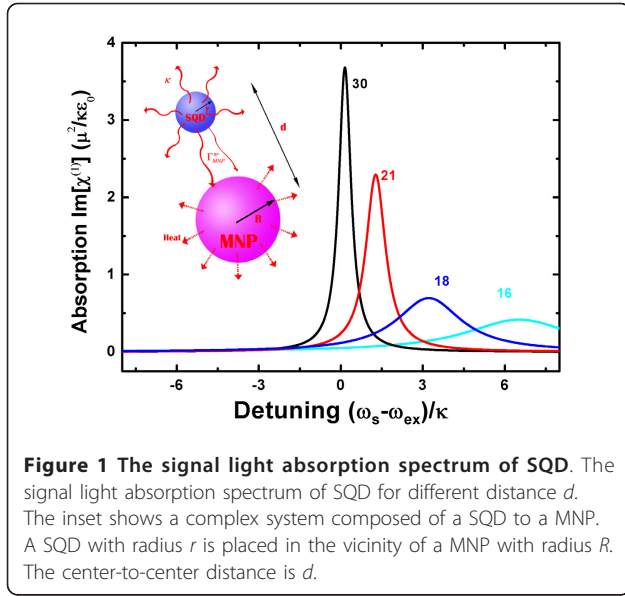
1 Introduction

Due to the advances in modern nanoscience, various nanostructures such as metal nanopartities (MNPs), semiconductor quantum dots (SQDs) and nanowires can be constructed for the applications in photonics and optoelectronics [1,2]. Studies of these nanostructures are essential for further development of nanotechnology. MNPs can be excited to produce surface plasmon polaritons (SPP) [3]. The energy transfer effect in a hybrid nanostruction complex composed of MNPs and SQDs has been observed, which implies the light-matter interaction between SPP field in MNPs and the excitons in SQDs [4,5]. To display the interaction between the exciton and SPP field, the vacuum Rabi splitting has been studied theoretically [6,7] and experimentally [8]. However, in the SQD-MNP coupled system a nonlinear Fano effect can be produced by a strong incident light [9]. Various theoretical [10,11] and experimental [12-14] reports have shown a decrease of the exciton lifetime of SQD placed in the vicinity of MNP. The decrease is related to the distance between SQD and MNP as a result of the coupling of the exciton and SPP field [15]. Moreover, the exciton energy level of SQD can be shifted because of the influence of SPP field [14].

Recently, the coupling among SQDs mediated by SPP field has received increasing attention [16,17]. The complex system like cavity QED system [18] and circuit QED system [19] may be applied in quantum information. Owing to the advantages of the solid-state of SQDs and integrated circuits of these nanostructures, the complex system is a promising candidate to implement the quantum information processing. However, more details about the coupling among SQDs and the role of SPP field need to be further studied. To illustrate clearly these quantum effects, a full quantum mechanics method to describe the coupled SQD-MNP system have to be developed.

In the present article, cavity QED as a quantum optics toolbox provides a full quantum mechanics description of the coupled SQD-MNP system. Under the description we develop a novel quantum transformation method that is suitable for the coupling SQDs to SPP field with large decay rate. The quantum transformation is used to treat master equation of the entire system. Under a certain condition, we obtain an effective Hamiltonian in SQDs' subsystem, and show a modified decay rate for each SQD. The effective Hamiltonian demonstrates an exciton energy shift and the coupling among SQDs. A cross-decay rate is induced by SPP field. It not only changes the decay rate of each SQD but also makes decay between every two SQDs. We analyze the exciton energy shift and the cross-decay rate of every SQD and

* Correspondence: zhukadi@sjtu.edu.cn
Key Laboratory of Artificial Structures and Quantum Control (Ministry of Education), Department of Physics, Shanghai Jiao Tong University, 800 DongChuan Road, Shanghai 200240, China

Figure 1 The signal light absorption spectrum of SQD. The signal light absorption spectrum of SQD for different distance d. The inset shows a complex system composed of a SQD to a MNP. A SQD with radius r is placed in the vicinity of a MNP with radius R. The center-to-center distance is d.

the coupling among SQDs, and find that these parameters are related to the distance between SQD and MNP. An experimental scheme to obtain these parameters is proposed by the observation of the signal light absorption spectrum of SQD in a system consisted of an SQD and an MNP. Based on the achievement of thes parameters, we design a simple model that two identical SQDs interact with an Au MNP for demonstrating the coupling of two SQDs.

2 Theory

We consider multiple SQDs in the vicinity of an MNP. Each SQD consists of the electronic ground state $|0\rangle$ and the first excited state $|ex\rangle$. They interact with SPP field in the MNP. First, we need to quantize SPP field based on the cavity quantum electrodynamics (QED). Recently, a good deal of study had been devoted to quantize SPP field in the metal [20-24]. SPP field in the MNP can be considered as a multiple-modes field. After the second quantization of SPP field, the Hamiltonian can be written as $H_{SPP} = \sum_k \omega_k a_k^+ a_k$ [20,21], where ω_k is the frequency of SPP mode k, $a_k^+(a_k)$ is the creation (annihilation) operation of SPP mode k. Next, we consider the interaction between each SQD and SPP modes. We assume that the coupling strength between each SQD and SPP field is identical for simplicity. The interaction Hamiltonian, under the rotating-wave approximation, can be written as $H_{int} = -\sum_{i,k} (g_k a_k \sigma_+^i + g_k^* a_k^+ \sigma_-^i)$ [22,25], where g_k is the coupling strength between each SQD and SPP mode

k, $\sigma_+^i = |ex\rangle_i \langle 0| (\sigma_-^i = |0\rangle_i \langle ex|)$ is the raising (lowering) operator of the ith SQD. Therefore, the Hamiltonian of the entire system can be written as ($\hbar = 1$)

$$H = \sum_i \omega_{ex} \sigma_z^i + \sum_k \omega_k a_k^+ a_k - \sum_{i,k} (g_k a_k \sigma_+^i + g_k^* a_k^+ \sigma_-^i) \quad (1)$$

where $\sigma_z^i = (1/2) \times (|ex\rangle_i \langle ex| - |0\rangle_i \langle 0|)$. The full quantum dynamics of the coupled nanosystem can be derived from the following master equation for the density operation

$$\partial_t \rho = -i[H, \rho] + \varsigma_{SQD} + \varsigma_{SPP}, \quad (2)$$

with the Liouvillian terms [26,27], $\varsigma_{SQD} = (\kappa/2) \times \sum_i (2\sigma_-^i \rho_+^i - \rho \sigma_+^i \sigma_-^i - \sigma_+^i \sigma_-^i \rho)$ describes the decay of each SQD to Markovian reservoirs, κ is the exciton radiative decay rate in SQDs, $\varsigma_{SPP} = \sum_i (\gamma_k/2) \times (2a_k \rho a_k^+ - \rho a_k^+ a_k - a_k^+ a_k \rho)$ describes the relaxation of SPP mode k with decay rate γ_k. Next, we take a time-independent unity transformation e^{is} on the density operator, where $s = \sum_{i,k} (\pi_k a_k \sigma_+^i + \pi_k^* a_k^+ \sigma_-^i), \pi_k = 2g_k/(\gamma_k + 2i\delta_k), \delta_k = \omega_k - \omega_{ex}$, so that $\partial_t \tilde{\rho} = -i[e^{is} H e^{-is}, \tilde{\rho}] + e^{is} \varsigma_{SQD} e^{-is} + e^{is} \varsigma_{SPP} e^{-is}.$,

$$\partial_t \tilde{\rho} = -i[e^{is} H e^{-is}, \tilde{\rho}] + e^{is} \varsigma_{SQD} e^{-is} + e^{is} \varsigma_{SPP} e^{-is}. \quad (3)$$

If $|\pi_k| \ll 1$, the second-order term remains, and the higher-order terms can be ignored safely. To obtain the reduce density operation of the SQDs' subsystem, we take a trace over the SPP field of the both hands of Eq. (3) by using $Tr_{SPP}[.]$. Here, we assume that the multi-mode plasmon field can be consider as a thermal reservoir and the reservoir variables are distributed in the uncorrelated thermal equilibrium mixture of states, $< a_k > = < a_k^+ > = 0, < a_k^+ a_l > = \bar{n}_k \partial_{kl}$, where the thermal average boson number $(\bar{n}_k)^{-1} = exp[(\omega_k)/(k_B T)] - 1$, k_B is the Boltzmann constant, and T is the temperature. Therefore,

$$Tr_{SPP}[\partial_t \tilde{\rho}] = -i \left[\left\{ \sum_i \omega_{ex} \sigma_z^i + \sum_k \omega_k \bar{n}_k + \sum_i 2\text{Re} \ [\pi_k (\pi_k^* \delta_k + i2g_k^*)](2\bar{n}_k + 1)\sigma_z^i \right. \right.$$
$$\left. + \sum_{i<j} 2\text{Re} \ [\pi_k (\pi_k^* \delta_k + i2g_k^*)](\sigma_+^i \sigma_-^j + \sigma_-^i \sigma_+^j) \right\}, \rho_{SQD} \right]$$
$$+ \varsigma_{SQD} + \sum_{ij} \text{Im}[\pi_k (\pi_k^* \delta_k + i2g_k^*)](2\sigma_-^i \rho_{SQD} \sigma_+^j - \sigma_+^i \sigma_-^j \rho_{SQD} - \rho_{SQD} \sigma_+^i \sigma_-^j), \quad (4)$$

where $\rho_{SQD} = Tr_{SPP}[\tilde{\rho}]$. After some algebraic calculation, the master equation of the reduce density operation of the SQDs' subsystem can be written as

$$\partial_t \rho_{SQD} = -i[H_{eff}, \rho_{SQD}] + \varsigma_{SQD}'. \quad (5)$$

The effective Hamiltonian to reveal the exciton energy shift and the coupling among SQDs is given by

$$H_{eff} = \sum_i (\omega_{ex} - \eta_0)\sigma_z^i - \eta \sum_{i<j}(\sigma_+^i\sigma_-^j + \sigma_-^i\sigma_+^j), \quad (6)$$

where

$$\eta_0 = \eta + \sum_k 8|g_k|^2\delta_k\bar{n}_k/(\gamma_k^2 + 4\delta_k^2),\; \bar{n}_k = <a_k^+a_k>, \quad \text{and}$$

$\eta = \sum_k 4|g_k|^2\delta_k/(4\delta_k^2 + \gamma_k^2)$ are the coupling strength among SQDs induced by quantized SPP modes. We can see that η_0 represents the exciton energy shift as a result of the coupling SQD to all quantized SPP modes. In the bosonic bath composed of all SPP modes, according to the Bose-Einstein distribution function, $\bar{n}_k \ll 1$ at low temperature so that $\eta_0 \cong \eta$. The dissipation term is given by

$$\varsigma'_{SQD} = (\Gamma_{i,j}/2) \times \sum_{i,j}(2\sigma_-^i\rho_{SQD}\sigma_+^j - \sigma_+^i\sigma_-^j\rho_{SQD} - \rho_{SQD}\sigma_+^i\sigma_-^j), \quad (7)$$

$\Gamma_{i,j} = \kappa + 2\tau$ if $i = j$, $\Gamma_{ij} = 2\tau$ if $i \neq j$, where $\tau = \sum_k 2|g_k|^2\gamma_k/(4\delta_k^2 + \gamma_k^2)$. We note that a cross-decay rate 2τ between every two SQDs appears and the exciton lifetime decreases because of the presence of SPP field. The cross-decay rate represents the nonradiative decay rate that can be decomposed into different contributions for each SPP mode, i.e., $2\tau \cong \Gamma_{MNP}^{nr}$ [22].

Our method to treat the Hamiltonian is similar with Schrieffer-Wolff transformation [28]. In cavity (circuit) QED system, when the decay rate of cavity mode is very small as compared to the detuning between the cavity mode frequency and the transition frequency of qubits so that it can be ignored safely, the effective Hamiltonian can be obtained by using Schrieffer-Wolff transformation [18,19]. Under the treatment of Schrieffer-Wolf transformation, one can obtain $\eta = \sum_k |g_k|^2/\delta_k, \tau = 0$. But it is well-known that the decay of SPP field is too large to be ignored in the coupled SQD-MNP system. Taking this fact fully into account, our method is suitable for revealing the exciton energy shift, the modify decay rate and the coupling strength among SQDs.

3 Coupling an SQD to an MNP

Now, we consider a simple complex system composed of an SQD and an MNP. As illustrated in inset of Figure 1, an SQD with radius r is placed in the vicinity of an MNP with radius R. The center-to-center distance is d. The modified decay rate of the SQD includes the radiative decay rate κ and the nonradiative decay rate Γ_{MNP}^{nr} induced by MNP. Owing to the ohmic losses within the metal a significant fraction of absorbed power has be dissipated as heat [3]. We first estimate the parameters η and τ. In the complex system, the SQD can induce polarization of MNP $P_{MNP} = [\gamma s_\alpha R^3 P_{SQD}]/[\varepsilon_{eff1}d^3]$, where $\gamma = [\varepsilon_M(\omega) - \varepsilon_0]/[\varepsilon_M(\omega) + 2\varepsilon_0], \varepsilon_{eff1} = [\varepsilon_s + 2\varepsilon_0]/[3\varepsilon_0], P_{SQD} = \mu(<\sigma_+> + <\sigma_->)$ [9], $\varepsilon_0, \varepsilon_s,$ and ε_M are the dielectric constants of the background medium, the SQD and the MNP, respectively, μ is the electric dipole moment of the exciton, s_α is related to the direction of the coupling. The SPP field induced by the SQD can be expressed as $E_{MNP} = [s_\alpha P_{MNP}]/[4\pi\varepsilon_0\varepsilon_{eff2}d^3]$ that is the mean value of the electric field operator $\widehat{E}_{MNP}$, where $\varepsilon_{eff2} = [\varepsilon_M(\omega) + 2\varepsilon_0]/[3\varepsilon_0]$. The operator can be split into two contributions $\widehat{E}_{MNP}^+ + \widehat{E}_{MNP}^-$ evolving with positive and negative frequencies [29]. Based on the principle of second quantization for SPP field, we have $<\mu\widehat{E}_{MNP}^+> = \sum_k g_k<a_k>$ [26]. The above result is under the dipole approximation when the distance is large comparing to the radius of the MNP. However, if the distance is comparable to the radius of the MNP, we need to consider the multipole polarization in the MNP, including dipole, quadrupole, octopole, and so on. So the multipole polarization can be expressed as $P_{MNP,tot} = \sum_{n=1}(s_n\varepsilon_0\gamma_n R^{2n+1}P_{SQD})/(\varepsilon_{eff1}d^{2n+1})$ [30], where $s_n = (n + 1)^2$ for the polarization parallel to the axis of the complex system, $\gamma_n = [\varepsilon_M(\omega) - \varepsilon_0]/[\varepsilon_M(\omega) + \varepsilon_0(n + 1)/n]$. For simplicity we assume that the distance is larger than the radius of the MNP so that the dipole approximation ($n = 1$) is reasonable. In the dissipative system, the expectation value $<a_k> = Tr[\rho\, a_k]$ of each SPP mode satisfies the equation, $\partial_t<a_k> = (\delta_k - i\gamma_k/2)<a_k> - g_k^*<\sigma_->$. At steady state, we can obtain

$$\sum_k \frac{|g_k|^2}{\delta_k - i\gamma_k/2} = \frac{\gamma(\mu s_\alpha)^2 R^3}{4\pi\varepsilon_0\varepsilon_{eff1}\varepsilon_{eff2}d^6}. \quad (8)$$

Therefore, $\eta = \text{Re}[G]$, $\tau = \text{Im}[G]$, where $G = [\gamma(\mu s_\alpha)^2 R^3]/[4\pi\varepsilon_0\varepsilon_{eff1}\varepsilon_{eff2}d^6]$. We note that, here, $\eta, \tau \sim d^{-6}$. So, it is reasonable that $g_k \sim d^{-3}$. The verdict is in good agreement with the coupling strength between a two-level system and a single mode of SPP field [24,27]. In [9], Zhang et al. found that the interaction between an SQD and an MNP leads to the formation of a hybrid exciton with the shifted exciton frequency and the decreased lifetime in which the SPP field is treated as a classical field rather than a quantized field. Here, we make a same conclusion under the quantized SPP field.

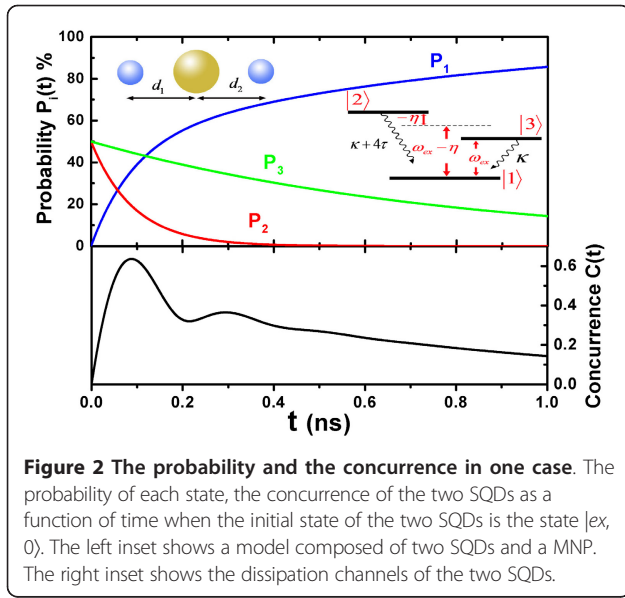An experimental scheme to measure the two parameters is proposed by observation on the absorption

Figure 2 The probability and the concurrence in one case. The probability of each state, the concurrence of the two SQDs as a function of time when the initial state of the two SQDs is the state $|ex, 0\rangle$. The left inset shows a model composed of two SQDs and a MNP. The right inset shows the dissipation channels of the two SQDs.

spectrum of SQD in the system. Now, we consider an SQD in the vicinity of an Au MNP excited a weak signal light E_s with frequency ω_s. According to master equation $\partial_t \rho_{SQD} = -i[H', \rho_{SQD}] + \varsigma'_{SQD}$, where $H' = (\omega_{ex} - \eta)\sigma_z - \mu(E_s\sigma_+ e^{-i\omega_s t} + E_s^*\sigma_- e^{i\omega_s t})$, $\varsigma_{SQD} = (\gamma_{SQD}^{tot}/2) \times (2\sigma_-\rho\sigma_+ - \rho\sigma_+\sigma_- - \sigma_+\sigma_-\rho)$, $\gamma_{SQD}^{tot} = \kappa + 2\tau$, we have

$$\partial_t p = [i(\eta - \omega_{ex}) - \gamma_{SQD}^{tot}]p - i\mu^2 E_s e^{-i\omega_s t} w, \qquad (9)$$

$$\partial_t w = -2\gamma_{SQD}^{tot}(w+1) + 2i\mu(E_s p^* e^{-i\omega_s t} - E_s^* p e^{i\omega_s t}), \quad (10)$$

where $p = \mu\rho_{ex,0}$, $w = \rho_{ex,ex} - \rho_{0,0}$.

The steady state solution can be obtained by setting the left-hand sides of Eqs. (9) and (10) equal to zero. Thus,

$$w = -\frac{1 + (\omega_s + \eta - \omega_{ex})^2 T^2}{1 + (\omega_s + \eta - \omega_{ex})^2 T^2 + 8|\mu E_s|^2 T^2}, \qquad (11)$$

$$p = \frac{\mu E_s w e^{-i\omega_s t}}{(\omega_s + \eta - \omega_{ex})T + i}, \qquad (12)$$

where $T = 1/\gamma_{SQD}^{tot}$. The polarization induced by the signal light can be expressed as $p = \varepsilon_0 \chi E_s e^{-i\omega_s t}$ [31], where χ is the total susceptibility to all order. We can obtain the total susceptibility: $\chi = (|\mu|^2/\varepsilon_0) \times [T - (\omega_s + \eta - \omega_{ex})T^2]/[1 + (\omega_s + \eta - \omega_{ex})^2 T^2 + 8|\mu E_s|^2 T^2]$. It can be expended in powers of the electric field $\chi = \chi^{(1)} + 3\chi^{(3)}|E_s|^2 + \cdots$, where

$$\chi^{(1)} = (|\mu|^2/\varepsilon_0) \times \frac{T - (\omega_s + \eta - \omega_{ex})T^2}{1 + (\omega_s + \eta - \omega_{ex})^2 T^2} \qquad (13)$$

is the first-order (linear) susceptibility.

In what follows, as an example, we consider a CdSe SQD with radius $r = 3.75$ nm [4] and an Au MNP with radius $R = 7.5$ nm. We use $\varepsilon_0 = 1.8$, $\varepsilon_s = 7.2$ [32] and the electric constant of Au $\varepsilon_M(\omega) = \epsilon_b - \omega_p^2/[\omega(\omega + i\eta_p)]$ with $\epsilon_b = 9.5$, $\hbar\omega_\omega = 2.8 eV$, $\hbar\omega_p = 9eV$, $\hbar\eta_p = 0.07 eV$ [22,33]. For the decay rate and dipole moment of the SQD, we take $\kappa = 1.25$ GHz and $\mu = er_0$ with $r_0 = 0.65$ nm. Figure 2 shows the absorption spectrum of the SQD (the imaginary part of linear susceptibility $\text{Im}[\chi^{(1)}]$) as a function of the signal-SQD detuning for $d = 30, 21, 18, 16$ nm. We note that the absorption peak is shifting and broadening with the decreasing distance between the SQD and the MNP. The absorption peak shift represents the exciton energy shift, and the broadened peak implies the increased decay rate of SQD as a result of the presence of SPP field. So, the exciton energy shift η and the cross-decay rate 2τ can be obtained by observation of the absorption spectrum. As shown in Figure 2, the exciton energy shift (full width at half maximum) is about 6.5κ (2.5κ) for a small distance $d = 16$ nm.

4 Coupling of two SQDs

We consider a simple model composed of two identical SQDs and an Au MNP for revealing the coupling between two SQDs induced by SPP field, as shown in left inset of Figure 2. The interaction between the two identical SQDs can be neglected safely in the absence of the MNP if the distance between them is very lager. When the distances between every SQD and the MNP are not equal ($d_1 \neq d_2$), we need to make a modification for the expression of two parameters η, τ. If one of the two distances changed, the expressions of the cross-decay rate and the coupling constant between the two SQDs need to be modified. As mentioned above, $g_k \sim d^{-3}$. The expression of the cross-decay rate and the coupling strength can be rewritten as $\text{Im}[G']$ and $\text{Re}[G']$, respectively, where $G' = [\gamma(\mu s_\alpha)^2 R^3]/[4\pi\varepsilon_0\varepsilon_{eff1}\varepsilon_{eff2}d_1^3 d_2^3]$. However, here, we assume that $d_1 = d_2 = d$ for simplicity. In the SQDs' subsystem, we choose an adequate basic of SQDs' subsystem, i.e., $|1\rangle = |0,0\rangle$, $|2\rangle = (1/\sqrt{2}) \times (|ex,0\rangle + |0,ex\rangle)$, $|3\rangle = (1/\sqrt{2}) \times (|ex,0\rangle - |0,ex\rangle)$, $|4\rangle = |ex,ex\rangle$. The four collective states are the eigenstates of the two coupling SQDs. The master equation of the SQDs' subsystem is given by

$$\partial_t \rho = -i[H'', \rho] + \varsigma_{SQD}, \qquad (14)$$

where $H'' = -(\omega_{ex} - \eta)|1\rangle\langle 1| - \eta|2\rangle\langle 2| + \eta|3\rangle\langle 3| + (\omega_{ex} - \eta)|4\rangle\langle 4|$, $\zeta_{SQD}(\rho) = [(\kappa + 4\tau)/2] \times [2(|2\rangle\langle 4| + |1\rangle\langle 2|)\rho(|4\rangle\langle 2| + |2\rangle\langle 1|) - (|2\rangle\langle 2| + |4\rangle\langle 4|)\rho - \rho(|2\rangle\langle 2| + |4\rangle\langle 4|)] + (\kappa/2) \times [2(|1\rangle\langle 3| - |3\rangle\langle 4|)\rho(|3\rangle\langle 1| - |4\rangle\langle 3|) - (|3\rangle
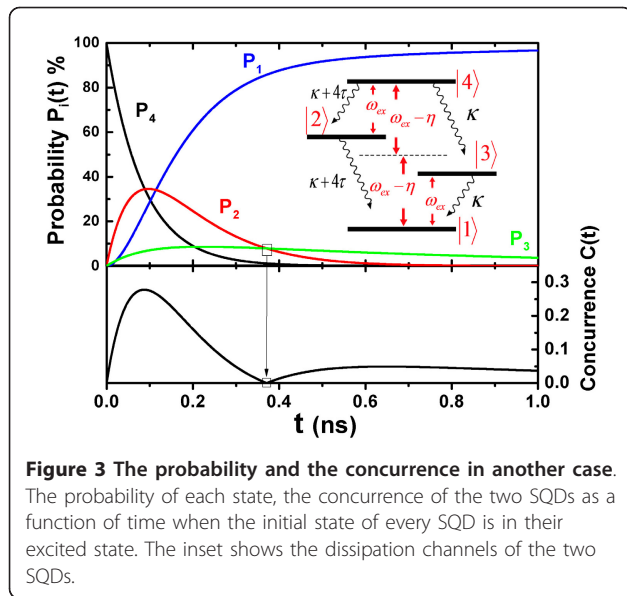
Figure 3 The probability and the concurrence in another case.
The probability of each state, the concurrence of the two SQDs as a
function of time when the initial state of every SQD is in their
excited state. The inset shows the dissipation channels of the two
SQDs.

$\langle 3| + |4\rangle \langle 4|)\rho - \rho(|3\rangle \langle 3| + |4\rangle \langle 4|)]$. It shows two dissipated channels. The first term describes dissipation through one cascade channel $|4\rangle \rightarrow |2\rangle \rightarrow |1\rangle$ with fast decay rate $\kappa + 4\tau$. The second term describes dissipation through another cascade channel $|4\rangle \rightarrow |3\rangle \rightarrow |1\rangle$ with slow decay rate κ (see inset of Figure 3).

In order to illustrate the coupling of the two SQDs, we analyze the following two parameters: (1) The probability of the two SQDs being in the state $|i\rangle$, $P_i(t) = \rho_{i,i}(t)$, for i = 1, 2, 3, 4. (2) The concurrence for quantifying entanglement of the two SQDs,

$$C(t) = \sqrt{[\rho_{2,2}(t) - \rho_{3,3}(t)]^2 + 4\text{Im} \, [\rho_{2,3}(t)]^2} \, [17,34].$$

Here we use the parameters of the above section, and take d = 16 nm.

If the initial state of the two SQDs is prepared in a product state $|ex, 0\rangle$, only two dissipation channels $|2\rangle \rightarrow |1\rangle$ and $|3\rangle \rightarrow |1\rangle$ should been considered (see right inset of Figure 2). To obtain the probability of each state, Eq. (14) can be rewritten as $\partial_t \rho_{i,j}(t) = -i \sum_k \left(H''_{i,k}\rho_{k,j} - H''_{k,j}\rho_{i,k} \right) + < i \left| \varsigma_{SQD} \right| j >$.

According to the initial state density matrix $\rho(0) = (|2\rangle + |3\rangle)(\langle 2| + \langle 3|)/2$, we can obtain the the probability of each state and the concurrence. As shown in Figure 2, with the decrease of $P_2(t)$ and $P_3(t)$, the probability of the two SQDs in the state $|1\rangle$ increases. At about t = 0.08 ns, the concurrence ofthe two SQDs reaches the maximal value. In the figure of the concurrence, a weak oscillation is presented as a result of the coupling of the two SQDs.

Another case is that the initial state is in another product state $|ex, ex\rangle$ ($\rho(0) = |4\rangle \langle 4|$). Figure 3 shows the

probability of each state, the concurrence as a function of time. It shows that the two SQDs can be entangled. Only at about t_0 = 0.275 ns the concurrence is equal to zero (see the figure of the concurrence); and $P_2(t_0)$ = $P_3(t_0)$ (see the figure of probability). This is because two entangled states $|2\rangle$ and $|3\rangle$ make a product state $|ex, 0\rangle$ or $|0, ex\rangle$. The absence of the oscillation in the figure of the concurrence implies that the coupling of the two SQDs cannot play a role in the creation of the concurrence. In the two cases, we can generate the entangled state of the two SQDs because the quantized SPP modes are act as the platform of the energy transfer between the two SQDs. If the MNP is absent ($d \rightarrow \infty$), the coupling strength η and the cross-decay rate τ of the two SQDs are equal to zero so that the SQDs cannot be entangled. We can tune the concurrence of the two SQDs by changing the distance d. In our theoretical calculations presented above, we do not consider size distribution of the SQD. A numerical averaging of the obtained results for different spatial dispersions of the distance will give a perfect prediction of the dispersion effects on the concurrence. Because of size inhomogeneities of CdSe SQD, we assume that the position distribution density satisfies the Gaussian distribution $\rho(r) = exp[-r^2/(2\sigma^2)]/(\sqrt{2\pi}\sigma)$, with the the half-width of Gaussian distribution σ = 16Å. Figure 4 shows a comparison between the original results and the modified results considering the dispersion effects on the concurrence under the two cases. We can see the difference between the two results. The difference becomes slighter with decreasing σ. When the half-
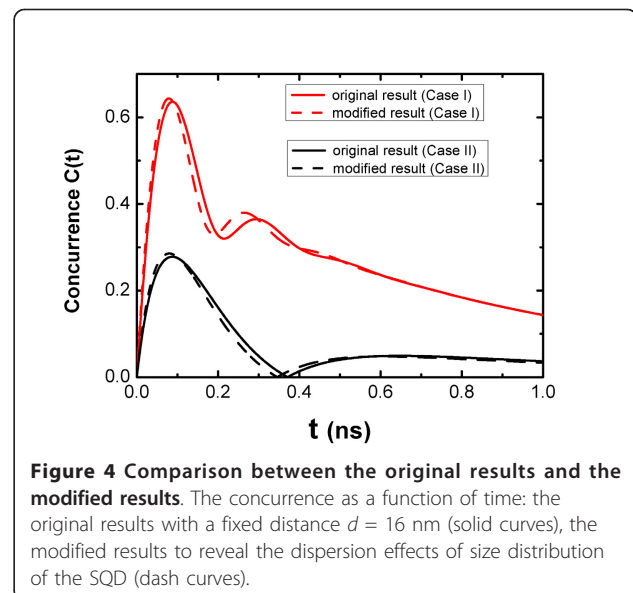

Figure 4 Comparison between the original results and the modified results. The concurrence as a function of time: the original results with a fixed distance d = 16 nm (solid curves), the modified results to reveal the dispersion effects of size distribution of the SQD (dash curves).

width σ is much smaller than the radius of the SQD, there is good agreement between the two results. Moreover, a stationary state with a high concurrence can be achieved by continuous pumping [17].

In conclusion, we have clearly demonstrated the interaction of SQDs and SPP field in MNP via a novel quantum transformation. The SPP field can induce the exciton energy shift and the decay rate modification of each SQD. The expressions of them is given by analysis. They can be measured by the designed scheme. Moreover, the coupling of two SQDs mediated by SPP field has been revealed strongly under two cases. With respect to the coupling among three or more SQDs, it is very significant for multipartite entanglement. The entanglement due to the light-matter interaction in the coupled SQD-MNP system may be applied in all-optical plasmon-enhanced nanoscale devices.

Authors' contributions
YH finished the main work of this paper, including deducing the formulas, plotting the figures, and drafting the manuscript. KDZ participated in the discussion and provided some useful suggestion. All authors are involved in revising the manuscript and approved the final version.

Competing interests
The authors declare that they have no competing interests.

References
1. Noginov M, Zhu G, Belgrave A, Bakker R, Shalaev V, Narimanov E, Stout S, Herz E, Suteewong T, Wiesner U: **Demonstration of a spaser-based nanolaser.** *Nature* 2009, **460**:1110.
2. Schuller JA, Barnard ES, Cai W, Jun YC, White JS, Brongersma ML: **Plasmonics for extreme light concentration and manipulation.** *Nat Mater* 2010, **9**:193.
3. Akimov A, Mukherjee A, Yu C, Chang D, Zibrov A, Hemmer P, Park H, Lukin M: **Generation of single optical plasmons in metallic nanowires coupled to quantum dots.** *Nature* 2007, **450**:402.
4. Govorov AO, Bryant GW, Zhang W, Skeini T, Lee J, Kotov NA, Slocik JM, Naik RR: **Exciton-plasmon interaction and hybrid excitons in semiconductor-metal nanoparticle assemblies.** *Nano lett* 2006, **6**:984.
5. Hosoki K, Tayagaki T, Yamamoto S, Matsuda K, Kanemitsu Y: **Direct and stepwise energy transfer from excitons to plasmons in close-packed metal and semiconductor nanoparticle monolayer films.** *Phys Rev Lett* 2008, **100**:207404.
6. Savasta S, Saija R, Ridolfo A, Di Stefano O, Denti P, Borghese F: **Nanopolaritons: vacuum rabi splitting with a single quantum dot in the center of a dimer nanoantenna.** *ACS nano* 2010, **4**:6369.
7. Manjavacas A, Abajo FJG, Nordlander P: **Quantum plexcitonics: strongly interacting plasmons and excitons.** *Nano lett* 2011, **11**:2318.
8. Passmore BS, Adams DC, Ribaudo T, Wasserman D, Lyon S, Davids P, Chow WW, Shaner EA: **Observation of rabi splitting from surface plasmon coupled conduction state transitions in electrically excited InAs quantum dots.** *Nano lett* 2011, **11**:338.
9. Zhang W, Govorov AO, Bryant GW: **Semiconductor-metal nanoparticle molecules: hybrid ex-citons and the nonlinear Fano effect.** *Phys Rev Lett* 2006, **97**:146804.
10. Merten H, Koenderink A, Polman A: **Plasmon-enhanced luminescence near noble-metal nanospheres: comparison of exact theory and an improved Gersten and Nitzan model.** *Phys Rev B* 2007, **76**:115123.
11. Vandenbem C, Brayer D, Froufe-Pirez L, Carminati R: **Controlling the quantum yield of a dipole emitter with coupled plasmonic modes.** *Phys Rev B* 2010, **81**:085444.
12. Okamoto K, Niki I, Shvartser A, Narukawa Y, Mukai T, Scherer A: **Surface-plasmon-enhanced light emitters based on InGaN quantum wells.** *Nat Mater* 2004, **3**:601.
13. Fedutik Y, Temnov V, Schps O, Woggon U, Artemyev M: **Exciton-plasmon-photon conversion in plasmonic nanostructures.** *Phys Rev Lett* 2007, **99**:136802.
14. Vasa P, Pomraenke R, Schwieger S, Mazur YI, Kunets V, Srinivasan P, Johnson E, Kihm J, Kim D, Runge E: **Coherent exciton-surface-plasmon-polariton interaction in hybrid metal-semiconductor nanostructures.** *Phys Rev Lett* 2008, **101**:116801.
15. Chen C, Wang C, Wei C, Chen Y: **Tunable emission based on the composite of Au nanopar-ticles and CdSe quantum dots deposited on elastomeric film.** *Appl phys lett* 2009, **94**:071906.
16. Lin ZR, Guo GP, Tu T, Li HO, Zou CL, Chen JX, Lu YH, Ren XF, Guo GC: **Quantum bus of metal nanoring with surface plasmon polaritons.** *Phys Rev B* 2010, **82**:241401.
17. Gonzalez-Tudela A, Martin-Cano D, Moreno E, Martin-Moreno L, Tejedor C, Garcia-Vidal FJ: **Entanglement of two qubits mediated by one-dimensional plasmonic waveguides.** *Phys Rev Lett* 2011, **106**:020501.
18. Zheng SB, Guo GC: **Efficient scheme for two-atom entanglement and quantum information processing in cavity QED.** *Phys Rev Lett* 2000, **85**:2392.
19. Majer J, Chow JM, Gambetta JM, Koch J, Johnson BR, Schreier JA, Frunzio L, Schuster DI, Houck AA, Wallraff A, Blais AM, Devoret H, Girvin SM, Schoelkopf RJ: **Coupling superconducting qubits via a cavity bus.** *Nature* 2007, **449**:443.
20. Bergman DJ, Stockman MI: **Surface plasmon amplification by stimulated emission of radiation: quantum generation of coherent surface plasmons in nanosystems.** *Phys Rev Lett* 2003, **90**:27402.
21. Trügler A, Hohenester U: **Strong coupling between a metallic nanoparticle and a single molecule.** *Phys Rev B* 2008, **77**:115403.
22. Hohenester U, Trügler A: **Interaction of single molecules with metallic nanoparticles.** *IEEE J Sel Top Quantum Electron* 2008, **14**:1430.
23. Sugakov V, Vertsimakha G: **Localized exciton states with giant oscillator strength in quantum well in vicinity of metallic nanoparticle.** *Phys Rev B* 2010, **81**:235308.
24. Waks E, Sridharan D: **Cavity QED treatment of interactions between a metal nanoparticle and a dipole emitter.** *Phys Rev A* 2010, **82**:043845.
25. Andersen ML, Stobbe S, Sørensen AS, Lodahl P: **Strongly modified plasmon-matter interaction with mesoscopic quantum emitters.** *Nat Phys* 2011, **7**:215.
26. Loudon R: *The Quantum Theory of Light* Oxford: Oxford University Press; 2000.
27. Ridolfo A, Di Stefano O, Fina N, Saija R, Savasta S: **Quantum Plasmonics with quantum dot-metal nanoparticle molecules: influence of the fano effect on photon statistics.** *Phys Rev Lett* 2010, **105**:263601.
28. Salomaa M: **Schrieffer-Wolff transformation for the Anderson Hamiltonian in a superconductor.** *Phys Rev B* 1988, **37**:9312.
29. Dalton B, Ficek Z, Swain S: **Atoms in squeezed light fields.** *J Mod Opt* 1999, **46**:379.
30. Yan J, Zhang W, Duan S, Zhao XG, Govorov AO: **Optical properties of coupled metal-semiconductor and metal-molecule nanocrystal complexes: role of multipole effects.** *Phys Rev B* 2008, **77**:165301.
31. Boyd RW: *Nonlinear Optics* Amsterdam: Academic Press; 2008.
32. Lu Z, Zhu K: **Slow light in an artificial hybrid nanocrystal complex.** *J Phys B: At Mol Opt Phy* 2009, **42**:015502.
33. De Abajo FJG: **Optical excitations in electron microscopy.** *Rev Mod Phys* 2010, **82**:209.
34. Wootters WK: **Entanglement of formation of an arbitrary state of two qubits.** *Phys Rev Lett* 1998, **80**:2245.