Quantum-dot exciton dynamics with a surface plasmon: Band-edge quantum optics

Y. N. Chen,[1] G. Y. Chen,[2,*] D. S. Chuu,[3] and T. Brandes[4]
[1]*Department of Physics and National Center for Theoretical Sciences, National Cheng-Kung University, Tainan 701, Taiwan*
[2]*Institute of Physics, National Chiao Tung University, Hsinchu 300, Taiwan*
[3]*Department of Electrophysics, National Chiao Tung University, Hsinchu 300, Taiwan*
[4]*Institut für Theoretische Physik, Technische Universität Berlin, Hardenbergstrasse, 36 D-10623 Berlin, Germany*
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We examine the spontaneous emission of a two-level emitter, quantum-dot exciton, into surface plasmons propagating on the surface of a cylindrical nanowire. The numerically obtained dispersion relations are found to influence the spontaneous emission rate strongly. At certain values of the exciton band gap, the emission rates can go to infinity due to the band-edge feature of the dispersion relations. Borrowing the idea from the photonic crystals, we model the quantum-dot exciton dynamics with a non-Markovian way and demonstrate that the decay can undergo an oscillatory behavior. In addition, we also show that the remote entangled states can be generated via super-radiance by using the one-dimensional propagating feature of the nanowire surface plasmons.

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I. INTRODUCTION

Great attention has been focused on the so-called plasmonics recently since surface plasmons reveal strong analogies to light propagation in conventional dielectric components [1]. For example, it is now possible to confine them to subwavelength dimensions [2] and develop novel approaches for waveguiding below the diffraction limit [3]. The useful subwavelength confinement, single mode operation [4], and relatively low power propagation loss [5] of surface-plasmon polaritons could be applied to miniaturize photonic circuits [6].

Plasmon-induced modification of the spontaneous emission (SE) is naturally an extended issue [7]. Sun *et al.* [8] recently calculated the Lamb shift of a hydrogen atom due to the surface-plasmon polariton. Strong enhancement of fluorescence due to surface plasmons was also observed [9]. Coherent coupling between individual optical emitters and guided plasmon excitations in conducting nanowires at optical frequencies was also pointed out [10] and may be used as a novel single-photon transistor [11].

In this work, we investigate the SE of a quantum-dot (QD) exciton coupled to surface plasmons in a metal nanowire. SE of a QD exciton into different modes of surface plasmons is considered separately. The emission rate is found to approach infinity at certain values of QD exciton band gap, which is similar to the band-edge effect in photonic crystals. In addition, application of such a system in generating remote entangled states is also pointed out and may be useful in future quantum information processing.

II. DISPERSION RELATIONS OF SURFACE PLASMONS

Consider now a colloidal CdSe/ZnS QD near a cylindrical silver nanowire with radius a. The QD and nanowire are assumed to be separated by a GaN layer [12] as shown in Fig. 1. One of the main reasons to choose a CdSe/ZnS QD exciton as the two-level emitter is that it is now possible to isolate single colloidal QD and measure its exciton lifetime [13]. The other reason is that its exciton band gap is around 2–2.5 eV, depending on the size and environment of the dot [14]. The plasmon energy $\hbar\omega_p$ of bulk silver is 3.76 eV with the corresponding saturation energy $\hbar\omega_p/\sqrt{2}\approx 2.66$ eV in the dispersion relation [15]. As we shall see below, variations in the dispersion relations in energy just match the exciton band gap of colloidal CdSe/ZnS QDs.

Surface-plasmon modes are created due to the nonzero local charge density on the surface of a nanowire. The nth surface-plasmon mode components of the electromagnetic field at the surface can be obtained by solving Maxwell's equations in a cylindrical geometry (ρ and φ denote the radial and azimuthal coordinates, respectively) with the appropriate boundary conditions [16],

$$E_\rho = \left[\frac{ik_z}{K_\xi}\frac{d\psi_n^\xi(K_\xi\rho)}{d(K_\xi\rho)}A_n^\xi - \frac{\mu_\xi\omega n}{K_\xi^2\rho}\psi_n^\xi(K_\xi\rho)B_n^\xi\right]\phi_n,$$

$$E_\varphi = -\left[\frac{nk_z}{K_\xi^2\rho}\psi_n^\xi(K_\xi\rho)A_n^\xi - \frac{i\mu_\xi\omega}{K_\xi}\frac{d\psi_n^\xi(K_\xi\rho)}{d(K_\xi\rho)}B_n^\xi\right]\phi_n,$$

$$E_z = [\psi_n^\xi(K_\xi\rho)A_n^\xi]\phi_n,$$

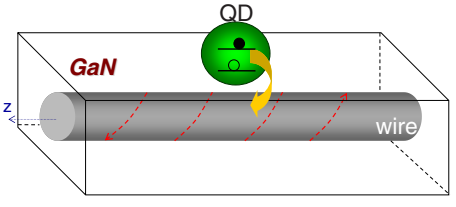

FIG. 1. (Color online) Schematic view of the model. Spontaneous emission of a two-level emitter (QD exciton) into nanowire surface plasmons, which act like photons in a cavity.

*guangyinchen.py95g@nctu.edu.tw

$$H_\rho = \left[\frac{n(K_\xi^2+k_z^2)}{\mu_\xi \omega K_\xi^2 \rho}\psi_n^\xi(K_j\rho)A_n^\xi + \frac{ik_z}{K_\xi}\frac{d\psi_n^\xi(K_\xi\rho)}{d(K_\xi\rho)}B_n^\xi\right]\phi_n,$$

$$H_\varphi = \left[\frac{i(K_\xi^2+k_z^2)}{\mu_\xi \omega K_\xi}\frac{d\psi_n^\xi(K_\xi\rho)}{d(K_\xi\rho)}A_n^\xi - \frac{nk_z}{K_\xi^2\rho}\psi_n^\xi(K_\xi\rho)B_n^\xi\right]\phi_n,$$

$$H_z = [\psi_n^\xi(K_\xi\rho)B_n^\xi]\phi_n, \tag{1}$$

with

$$K_\xi^2 = \omega^2\epsilon_\xi(\omega)/c^2 - k_z^2 \quad (\xi = I \text{ or } O),$$

$$\psi_n^J(K_I\rho) = J_n(K_I\rho), \quad \psi_n^O(K_O r) = H_n^{(1)}(K_O\rho),$$

$$\phi_n = \exp(in\varphi + ik_z z - i\omega t),$$

where $J_n(K_I\rho)$ and $H_n^{(1)}(K_O\rho)$ are Bessel and Hankel functions, respectively. I (O) stands for the component inside (outside) the wire. The dielectric function is assumed as $\epsilon(\omega)=\varepsilon_\infty[1-\frac{\omega_p^2}{\omega(\omega+i/\tau)}]$, where $\epsilon_\infty=9.6$ (for Ag) and $\epsilon_\infty=5.3$ (for GaN). The plasma energy ($\hbar\omega_p$) of bulk silver is 3.76 eV, and $\tau=3.1\times10^{-14}$ s is the relaxation time due to ohmic metal loss [17], which has been taken into account in the following calculations. The magnetic permeabilities $\mu_{I,O}$ are unity everywhere since we consider nonmagnetic materials here. A_n^ξ and B_n^ξ are constants to be determined by normalizing the electromagnetic field to the vacuum fluctuation energy, $\int\epsilon(|E_\rho|^2+|E_\varphi|^2+|E_z|^2)d\mathbf{r}=\hbar\omega(\mathbf{k})$, and matching the boundary conditions. According to the experimental report [18], in general, the length of the nanowire is very long compared to the size of the QD. Therefore, it is legitimate to treat the length of the nanowire as effectively infinite. In this case, the dispersion relations of the surface plasmons with a continuum spectrum can be obtained by solving the following transcendental equation numerically [16]:

$$S(k_z,\omega) = \left[\frac{\mu_I}{K_I a}\frac{J_n'(K_I a)}{J_n(K_I a)} - \frac{\mu_O}{K_O a}\frac{H_n^{(1)'}(K_O a)}{H_n^{(1)}(K_O a)}\right] \times \left[\frac{(\omega/c)^2\varepsilon_I(\omega)}{\mu_I K_I a}\frac{J_n'(K_I a)}{J_n(K_I a)} - \frac{(\omega/c)^2\varepsilon_O(\omega)}{\mu_O K_O a}\frac{H_n^{(1)'}(K_O a)}{H_n^{(1)}(K_O a)}\right] - n^2k_z^2\left[\frac{1}{(K_O a)^2} - \frac{1}{(K_I a)^2}\right]^2 = 0. \tag{2}$$

Figure 2(a) shows the dispersion relations of the $n=0$ mode for different radii. Here, one unit of the effective radii $R(\equiv\omega_p a/c)$ is roughly equal to 53.8 nm. As can be seen, the behavior of these curves is very similar to the two-dimensional case [17], i.e., $\Omega(\equiv\omega/\omega_p)$ gradually saturates with increasing wave vector $K(\equiv k_z c/\omega_p)$. This is because the fields for the $n=0$ mode are independent of the azimuthal angle φ. However, the behaviors for the $n\neq0$ modes are quite different as shown in Figs. 2(b) and 2(c). The first interesting point is the discontinuities around $\omega/c\approx k_z$. Further analysis shows that the solutions of ω are "almost real"

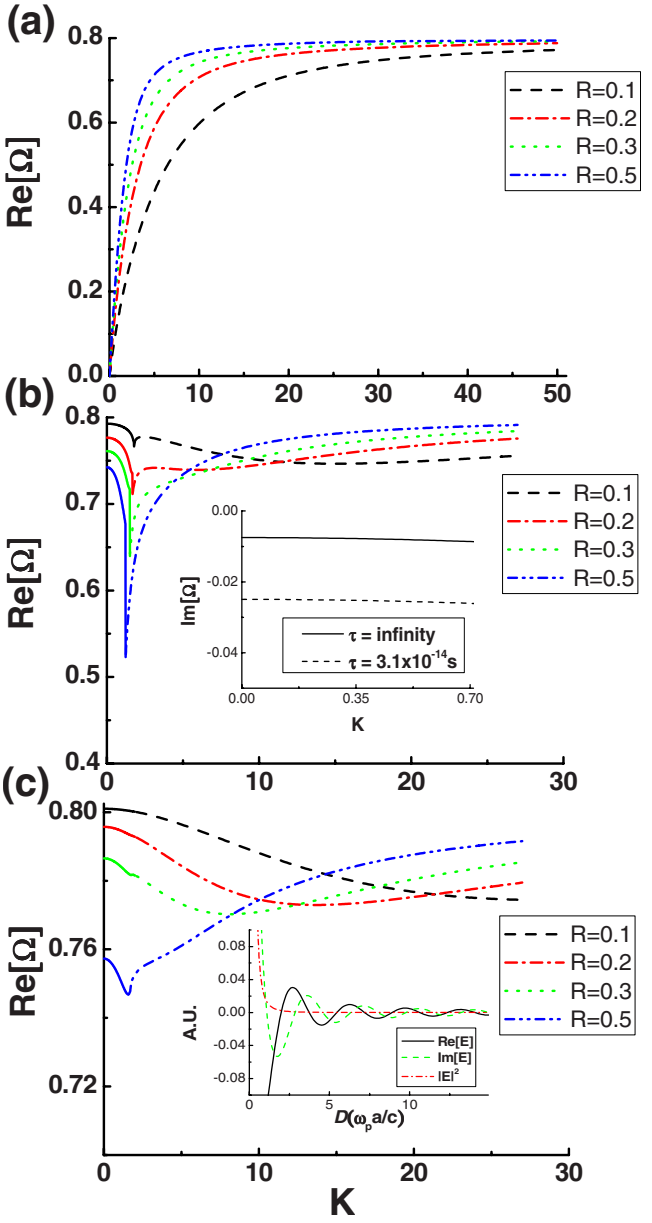

FIG. 2. (Color online) (a), (b), and (c) represent the dispersion relations of surface plasmons for the modes $n=0$, 1, and 2, respectively. The nonsolid (solid) lines represent the bound (nonbound) modes. The units for vertical and horizontal lines are $\Omega=\omega/\omega_p$ and $K=k_z c/\omega_p$, and $R\equiv\omega_p a/c$. The inset in (c) represents the real part, imaginary part, and intensity of the electric field for $n=1$ nonbounded mode as a function of distance away from the wire surface.

[19] as $k_z>\mathrm{Re}[\omega]/c$. In this case, the first kind of Hankel function of order n, $H_n^{(1)}(K_\xi\rho)$, decays exponentially. This means that the surface plasmons in this regime are confined on the surface (*bound modes*). For $k_z<\mathrm{Re}[\omega]/c$, however, the solutions of ω are *complex*. The form of $H_n^{(1)}(K_\xi\rho)$ in this case is similar to a traveling wave (*nonbound modes*), for which its lifetime is finite. One might think that the reason for the finite lifetime is totally from the ohmic metal loss. However, as shown in the inset of Fig. 2(b), the frequency is still complex (the solid line) even without the metal loss τ. We thus conclude that the finite lifetime in the regime of k_z

$<\mathrm{Re}[\omega]/c$ is actually influenced by both metal and radiation loss.

To calculate, the SE rate of a QD or atom within a structured reservoir, one, in general, considers the contributions from the scattered fields for different surface geometry of the surrounding scatters. There are some well-developed means to deal with such calculations. For instance, making use of the Green's tensors, one can calculate the scattered fields and obtain the local density of states for an atomic dipole [20]. Moreover, once the surfaces of scatters are metallic, the presence of surface plasmons is expected to dominate the SE rate due to the strong coupling between surface plasmons and QD [10]. In our case, we would like to focus on the effect of surface plasmons on the SE rate since other contributions of the scattering fields are much smaller than that of the surface plasmons.

The general decay rate of a QD or atom coupled to multimode electromagnetic fields can be directly obtained from Fermi's golden rule [21] within the dipole approximation,

$$\Gamma_{sp} = \frac{2\pi}{\hbar} \int d\vec{k} |\vec{d}_0 \cdot \vec{E}(\vec{k})|^2 \delta(\omega_0 - \omega_{\vec{k}}), \tag{3}$$

where $\omega_{\vec{k}}$ and $\vec{k}$ are the frequency and wave vector of the field $\vec{E}(\vec{k})$, respectively. $\vec{d}_0$ is the dipole moment of the QD exciton and ω_0 is the exciton band gap of the QD. Once the electromagnetic fields are determined, the SE rate, Γ_{sp}, of the QD excitons into bound surface plasmons can be obtained via Eq. (3). Since the surface plasmons are confined on the surface [22] of the cylindrical nanowire, the integral of $\vec{k}$ in Eq. (3) stands for the summation of the contributions from all the possible final states, i.e., a two-dimensional integral of k_φ and k_z. Because n is the quantum number governing the φ component of the wave function, summing over all n modes is equivalent to the integral over all k_φ. For convenience, we assume that the dipole moment $\vec{d}_0$ is along the ρ direction. By transforming the argument of the delta function from $\omega_{\vec{k}}(=\omega_{n,k_z})$ to k_z, the SE rate can then be written as

$$\Gamma_{sp} = \sum_{n=0}^{\infty} \Gamma_n = \frac{2\pi}{\hbar} \sum_{n=0}^{\infty} \frac{\sum_{k_{z_i}} |\vec{d}_0 \cdot \vec{E}_\rho(k_{z_i})|^2}{\left| \frac{d(\omega_0 - \omega_{n,k_z})}{dk_z} \right|_{k_{z_i}}}, \tag{4}$$

where Γ_n is the SE rate into the nth mode and k_{z_i} stands for the values of k_z that make the argument in the δ function vanish.

For the purpose of discussion, we display the SE rate into the first few modes $(\Gamma_n, n=0,1,2,3)$ as shown in Fig. 3 for $R=0.1$ and 0.5, respectively. In plotting Fig. 3, the distance between the dot and the wire surface is fixed as $d=10.76$ nm. We find that the latter modes $(n>3)$ contribute much less to the decay rate. For certain ranges of ω_0, the contributions to the decay rate Γ_{sp} mainly come from the first few modes. For example, if we set $\omega_0=0.746\,47$, which is the minimum point of the $n=1$ mode dispersion curve, the decay rate (for $R=0.1$ case) is mainly from $n=0$ and $n=1$ modes as seen from Fig. 3(a). In addition, the different feature here is that the SE rate approaches infinity at certain values of the exciton band gap ω_0. Mathematically, one might think that at these values the corresponding slopes of the dispersion relation are zero [23]. Physically, however, this infinite rate is not reasonable since it is based on perturbation theory. Therefore, one has to treat the dynamics of the exciton around these values more carefully, i.e., the *Markovian* SE rate is not enough. One has to consider the *non-Markovian* behavior around the band edge, which means that the band abruptly appears or disappears across certain values of ω_{n,k_z}.

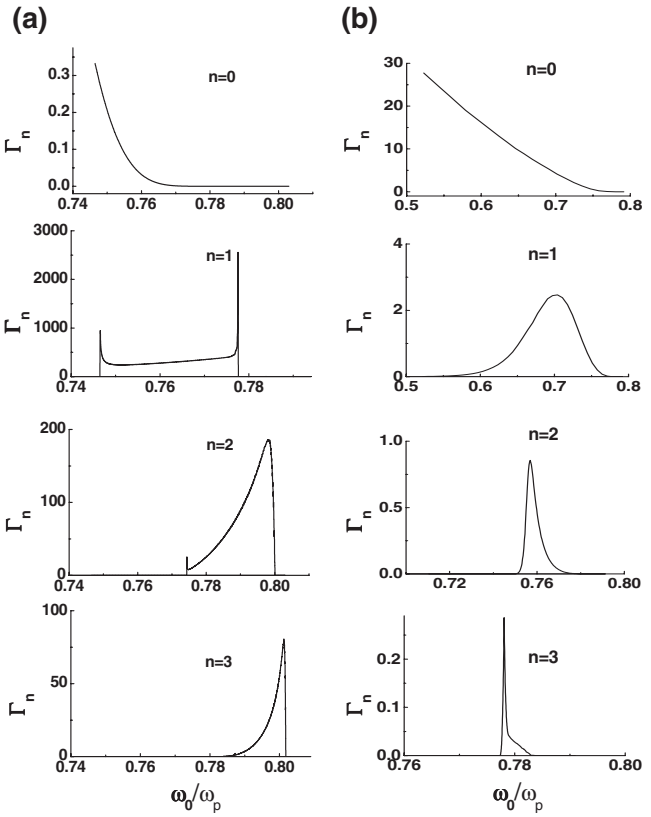

FIG. 3. Spontaneous emission rate (Γ_n) into $n=0\sim3$ modes for (a) $R=0.1$ and (b) $R=0.5$. The unit of Γ_n is normalized to the free-space decay rate γ_0.

III. NON-MARKOVIAN DYNAMICS OF QD EXCITONS

When an open quantum system interacts with a structured reservoir, there exists non-Markovian memory effect in the form of oscillatory behavior of decay dynamics which reflects the exchanges of information back and forth between system and reservoir. Recently, Piilo *et al.* [24] developed a non-Markovian quantum jump method which generalized the proven Monte Carlo wave-function method for the Markovian system in order to deal with the non-Markovian problems. In our work, we will numerically solve the time-dependent Schrödinger equation to obtain the time-dependent population on the excited state.

To obtain the non-Markovian dynamics of the exciton, we first write down the Hamiltonian of the system in the inter-

action picture (with the rotating wave approximation),

$$H_{ex\text{-}sp} = \sum_{n,k_z} \hbar\Delta_{n,k_z}\hat{a}^{\dagger}_{n,k_z}\hat{a}_{n,k_z} + i\hbar\sum_{n,k_z}(g_{n,k_z}\hat{a}^{\dagger}_{n,k_z}\sigma_{\downarrow\uparrow} - g^{*}_{n,k_z}\hat{a}_{n,k_z}\sigma_{\uparrow\downarrow}), \quad (5)$$

where $\sigma_{ij}=|i\rangle\langle j|(i,j=\uparrow,\downarrow)$ are the atomic operators; $\hat{a}_{n,k_z}$ and $\hat{a}^{\dagger}_{n,k_z}$ are the radiation field (surface plasmon) annihilation and creation operators; $\Delta_{n,k_z}=\omega_{n,k_z}-\omega_0$ is the detuning of the radiation mode frequency ω_{n,k_z} from the excitonic resonant frequency ω_0, and $g_{n,k_z}=\vec{d}_0\cdot\vec{E}_{n,k_z}$ is the atomic field coupling.

Assuming that there is an exciton in the dot with no plasmon excitation in the wire initially, the wave function of the system then has the form

$$|\psi(t)\rangle = b_e(t)|\uparrow,0\rangle + \sum_{n,k_z} b_{n,k_z}(t)|\downarrow,1_{n,k_z}\rangle e^{-i\Delta_{n,k_z}t}. \quad (6)$$

The state vector $|\uparrow,0\rangle$ describes an exciton in the dot and no plasmons present, whereas $|\downarrow,1_{n,k_z}\rangle$ describes the exciton recombination and a surface plasmon emitted into mode k_z. With the time-dependent Schrödinger equation, the solution of the coefficient $b_e(t)$ in z space is straightforwardly given by

$$\tilde{b}_e(z) = \left[z + \sum_{n=0}^{\infty}\int g_{n,k_z}g^{*}_{n,k_z}\frac{dk_z}{z+i(\omega_{n,k_z}-\omega_0)}\right]^{-1}. \quad (7)$$

We use the dispersion relations obtained from Eq. (2) to numerically calculate the integral over the whole spectra of n and k_z in Eq. (7). Consequently, $b_e(t)$ can be obtained by performing a numerical inverse Laplace transformation to Eq. (7).

The dashed, dotted, and dashed-dotted lines in Fig. 4(a) represent the decay dynamics of the QD excitons for different detunings, $\delta=-0.4\gamma_0$, $0.4\gamma_0$, and $0.8\gamma_0$, respectively. Here, $\delta=\omega_0-\omega_{n=1,k_z}$ is the detuning from the local minimum of the $n=1$ mode and γ_0 is the decay rate of the QD exciton into free space. The radius of the wire and the wire-dot separation are $R=0.1$ and $d=0.34$, respectively. Apparently, there exists oscillatory behavior in the decay profile, demonstrating that decay dynamics is non-Markovian. If one considers only the contribution from the $n=1$ mode and set the detuning $\delta=0$, the probability amplitude would saturate to a steady limit as shown by the solid line. This *quasidressed* state is an analogy of Rabi oscillation in cavity quantum electrodynamics and also appears in the systems of photonic crystals [25]. In the investigations for SE of a two-level atom near the edge of a photonic band gap, the density of states becomes singular, and the dispersion relation near the band edge can be approximated as a parabolic curve [25]. The oscillatory behavior during the decay can be then obtained by treating the transition from the excited state to the intermediate state as the other decay channel. The oscillatory behavior in the photonic crystal case is a direct consequence of strong interaction between the atom and its own localized radiation. In our case, the coupling between the QD exciton and surface plasmons can be very strong as well, resulting from a similar feature of local extremum in the dispersion curve. So, the oscillations in decay dynamics shown in Fig. 4(a) can be understood as the SE near a band edge.

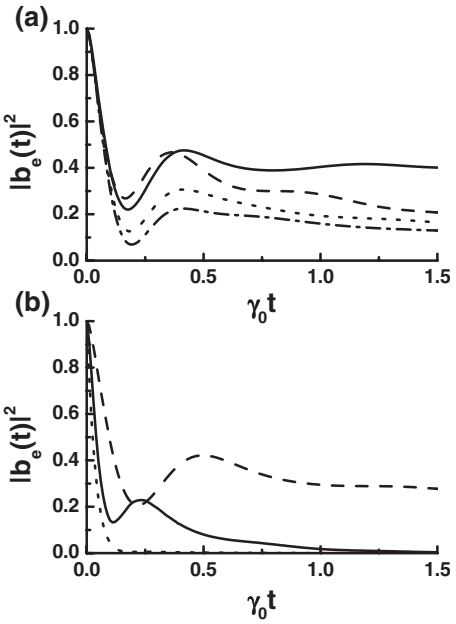

FIG. 4. (a) Non-Markovian decay dynamics of QD excitons for $\delta=-0.4\gamma_0$ (dashed line), $0.4\gamma_0$ (dotted line), and $0.8\gamma_0$ (dashed-dotted line). As $\delta=0$, the solid line represents the result for the contribution from the $n=1$ mode. (b) By setting $\delta=0$, the dotted, solid, and dashed lines represent the results for dot-wire separation $d=0.2$, 0.3, and 0.35, respectively. Here, one unit of d is $\omega_p a/c=53.8$ nm.

Another interesting discovery is shown in Fig. 4(b) if one sets the detuning $\delta=0$ and plots the dynamics of the exciton for different dot-wire separations: $d=0.2$ (dotted line), $d=0.3$ (solid line), and $d=0.35$ (dashed line). As can be seen, the oscillatory behavior is diminished with the decrease in the dot-wire separation. This is because as ω_0 is chosen to be close to the local minimum of the dispersion relation of the $n=1$ mode, the decay dynamics is mainly dominated by the contributions from the $n=0$ and $n=1$ modes. Since the non-Markovian oscillatory behavior is mainly from the local minimum of the $n=1$ mode, the contribution from the $n=1$ mode can be overwhelmed by that from the $n=0$ mode if the dot is put close enough to the wire surface. This leads to a degradation of the oscillatory behavior.

IV. APPLICATION IN ENTANGLEMENT GENERATION

Let us now put another QD close to the wire, the interaction between the wire and QDs can now be written as

$$H' = \sum_{n,k_z} g_{n,k_z}(\sigma_{1+} + \sigma_{2+}e^{ik_z z_0})\hat{a}_{n,k_z} + \text{H.c.}, \quad (8)$$

where a_{n,k_z} is the surface-plasmon operator and σ_{j+} is the creation operator of the jth QD. Note that in Eq. (8) we have assumed no detuning and the two dots have the same separation from the metal wire. Since the propagating modes are along the z direction only, the phase difference acquired by the second dot is $ik_z z_0$, where z_0 is the separation between the

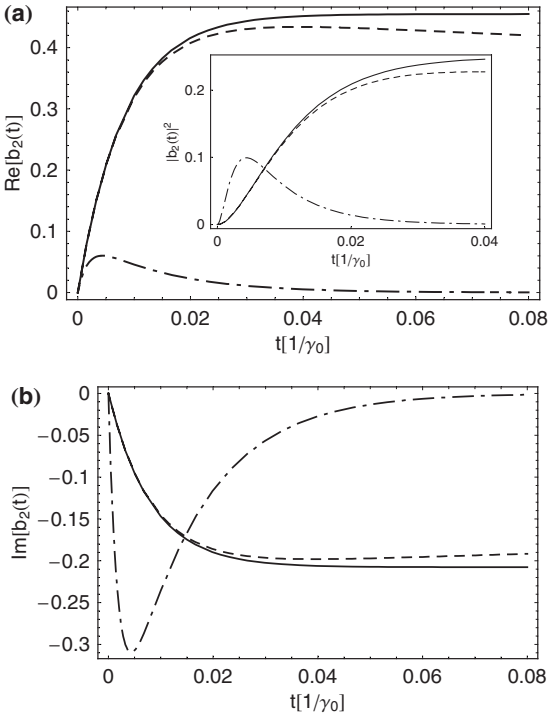

FIG. 5. Variations in $\mathrm{Re}[b_2(t)]$, $\mathrm{Im}[b_2(t)]$, and $|b_2(t)|^2$ [inset of (a)] as functions of time for $\omega_0=0.602\omega_p$ (solid curves) and $0.748\omega_p$ (dashed-dotted curves). In plotting the figure, the interdot distance z_0 is set equal to $0.35(\omega_p a/c)$ with radius $R=0.1$. The dashed curves in (a) and (b) are the results for $\omega_0=0.602\omega_p$ with the inclusion of the contributions from other channels: the free-space decay rate $\Gamma_f(=\gamma_0)$ and nonradiative decay rate $\Gamma_{\mathrm{non}}(\sim\gamma_0)$.

two dots. If one further assumes that only QD-1 is initially excited, the state vector of the system can be written as

$$|\Psi(t)\rangle = b_1(t)|\uparrow\downarrow;0\rangle + b_2(t)|\downarrow\uparrow;0\rangle + \sum_{n,k_z} b_{n,k_z}(t)|\downarrow\downarrow;1_{n,k_z}\rangle, \tag{9}$$

with $b_1(0)=1$ and $b_2(0)=b_{n,k_z}(0)=0$. Here, $|\uparrow\downarrow;0\rangle$ ($|\downarrow\uparrow;0\rangle$) means that QD-1 (QD-2) is excited, while $|\downarrow\downarrow;1_{n,k_z}\rangle$ represents that both the QDs are de-excited with the presence of single surface plasmon. If we let the exciton band gap ω_0 far away from the band edge, $b_1(t)$ and $b_2(t)$ can be obtained easily by solving the time-dependent Schrödinger equation.

Figure 5 shows the time variations in $\mathrm{Re}[b_2(t)]$, $\mathrm{Im}[b_2(t)]$, and $|b_2(t)|^2$ for different values of the exciton band gap. For $\omega_0=0.748\omega_p$, the population of the second dot vanishes quickly as seen from the dashed-dotted curves. For $\omega_0=0.602\omega_p$, however, it approaches (quasi-) stationary limit [26] as shown by the solid curves. This is because, for the latter case, only $n=0$ mode contributes to the decay rate Γ_{sp}. In this case, the system is similar to a one-dimensional superradiant one with the populations written as

$$b_1(t) = e^{-2\Gamma_{sp}t}(1+e^{2\Gamma_{sp}t})/2,$$

$$b_2(t) = e^{-ik_0z_0-2\Gamma_{sp}t}(-1+e^{2\Gamma_{sp}t})/2. \tag{10}$$

From Eq. (10), one realizes that there is always a 50% chance for the two dots to evolve into the state, $|\uparrow\downarrow\rangle + e^{-ik_0z_0}|\downarrow\uparrow\rangle$. It means that, for example, the singlet [triplet] entangled state can be created if $k_0z=(2m+1)\pi$ [$2m\pi$] with m being an integer [27]. One might argue that the QD excitons can also have other decay channels (free-space radiation and nonradiative decay due to metal loss), such that the generation of the entanglement might be invalid. We thus plot in Fig. 5 the dashed curves for $\omega_0=0.602\omega_p$ with the contributions from the free-space decay rate $\Gamma_f(=\gamma_0)$ and nonradiative decay rate Γ_{non}, which is assumed to be roughly equal to γ_0. As seen, the (quasi-) stationary state is not greatly degraded in the short-time regime, $t \ll 1/(\Gamma_f+\Gamma_{\mathrm{non}})$. In other words, as long as the SE rate into surface plasmons is large enough, the generated entanglement is still observable.

V. SUMMARY

We have numerically calculated the dispersion relations of the nanowire surface plasmons and have shown that the SE of QD excitons into surface plasmons can be greatly enhanced at certain values of the exciton band gap. The enhancement is due to the band-edge effect [28] in dispersion relation, and one has to treat the decay dynamics with a non-Markovian way. In addition, an idea of creating remote entangled states between two QDs is also proposed and can be tested with current technology [29].

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