Spontaneous emission of quantum dot excitons into surface plasmons in a nanowire

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The spontaneous emission (SE) of quantum dot (QD) excitons into surface plasmons in a cylindrical nanowire is investigated theoretically. Maxwell's equations with appropriate boundary conditions are solved numerically to obtain the dispersion relations of surface plasmons. The SE rate of QD excitons is found to be greatly enhanced at certain values of the exciton bandgap. Application in generation of remote entangled states via superradiance is also pointed out and may be observable with current technology. © 2008 Optical Society of America

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When a light wave strikes a metal surface, a surface plasmon polariton can be excited [1,2]. Investigations of the dispersion relations of surface plasmons for different geometries have been reported [3] since the 1970s. Recently, great attention has been focused on the so-called plasmonics since surface plasmons reveal strong analogies to light propagation in conventional dielectric components [4–6]. Plasmon-induced modification of the spontaneous emission (SE) rate is naturally an extended issue [7–9]. Recently, strong and 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 Letter, we investigate the SE rate of a II–VI colloidal quantum dot (QD) (nanocrystals) exciton coupled to surface plasmons in a silver nanowire. SE of a QD exciton into different modes of surface plasmons is considered separately. The emission rate is found to be greatly enhanced with a discontinuous feature if one varies the energy bandgap of the QD exciton. Application of such a system in generating remote entangled states via collective decay (superradiance) is also pointed out and may be useful in future quantum information processing.

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 as shown in Fig. 1(a). The nth surface plasmon mode's 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 [3]. The dispersion relations of the surface plasmons are thus obtained by solving the following transcendental equation numerically:

$$S(k_z,\omega)=\left[\frac{\mu_I}{K_Ia}\frac{J_n'(K_Ia)}{J_n(K_Ia)}-\frac{\mu_O}{K_Oa}\frac{H_n^{(1)\prime}(K_Oa)}{H_n^{(1)}(K_Oa)}\right]\times\left[\frac{(\omega/c)^2\varepsilon_I(\omega)}{\mu_IK_Ia}\frac{J_n'(K_Ia)}{J_n(K_Ia)}-\frac{(\omega/c)^2\varepsilon_O(\omega)}{\mu_OK_Oa}\frac{H_n^{(1)\prime}(K_Oa)}{H_n^{(1)}(K_Oa)}\right]-n^2k_z^2\left[\frac{1}{(K_Oa)^2}-\frac{1}{(K_Ia)^2}\right]^2=0, \quad (1)$$

where $K_\xi^2=\omega^2\epsilon_\xi(\omega)/c^2-k_z^2$ ($\xi=I$ or O) and $\phi_n=\exp(in\varphi+ik_zz-i\omega t)$. Here, $J_n(K_I\rho)$ 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 $\epsilon_I(\omega)=\varepsilon_\infty[1-\omega_p^2/\omega(\omega+i/\tau)]$, where $\epsilon_\infty=9.6$ (for Ag), $\epsilon_\infty=5.3$ (for GaN), and τ is the relaxation time due to ohmic metal loss [9]. The magnetic permeabilities $\mu_{I,O}$ are unity everywhere since we consider nonmagnetic materials here.

Figure 1(b) shows the dispersion relations of the $n=0$ mode for different radii. Here, one unit of the effective radii R ($\equiv\omega_pa/c$) is roughly equal to 53.8 nm. As can be seen, the behavior of these curves is very similar to the two-dimensional case [12]. 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. 1(c) and 1(d). The first interesting point are the discontinuities around $\omega/c\approx k_z$. Further analysis shows that the solutions of ω are "almost real" [13] as $k_z>\mathrm{Re}[\omega]/c$. In this case, the first kind Hankel function of order n, $H_n^{(1)}(K_\xi\rho)$, decays exponentially. This

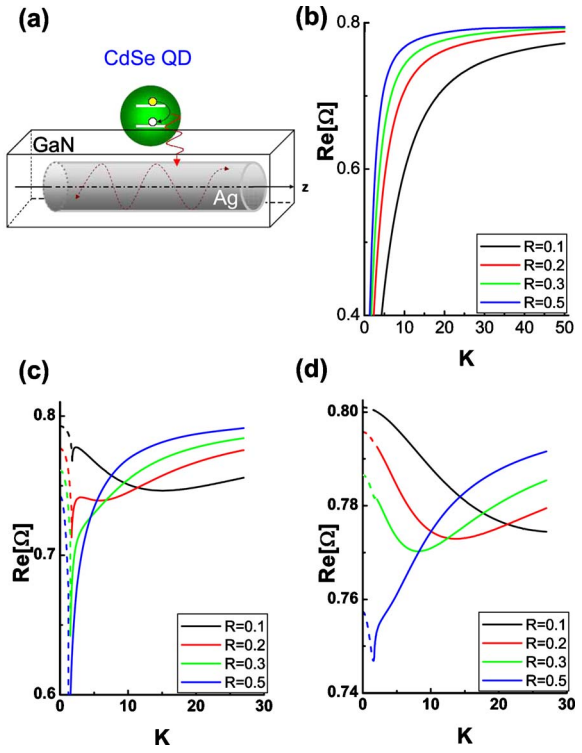

Fig. 1. (Color online) (a) Schematic view of the model; a silver nanowire is embedded inside GaN material and a colloidal QD is put on top of it. (b)–(d) Dispersion relations of surface plasmons for the modes $n=0$, 1, and 2, respectively. The solid (dashed) curves represent the bound (nonbound) modes. The units for vertical and horizontal lines are $\Omega=\omega/\omega_p$ and $K=k_z c/\omega_p$.

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 like a traveling wave with finite lifetime (nonbound modes).

Once the electromagnetic fields are determined, the SE rate, Γ_{sp}, of the QD excitons into bound surface plasmons can be obtained via Fermi's golden rule and is given by

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

where $\mathbf{d}$ is the dipole moment of the QD exciton, $\mathbf{E}_\rho$ is the electric field in ρ direction, k_{z_i} stands for the values of k_z that make the argument in the δ function vanish, and ω_0 is the exciton bandgap of the colloidal CdSe–ZnS QD. Note that in deriving Eq. (2), we have assumed that the dipole moment $\mathbf{d}$ is along the ρ direction for convenience.

The SE rates of the first few modes ($n=0,1,2,3$) are shown in Fig. 2 for $R=0.1$ and 0.5, respectively. In plotting the figures, the distance between the dot and the wire surface is fixed at 10.76 nm. The first novel feature is that the SE rate approaches infinity at certain values of the exciton bandgap ω_0. The reason is that at these values the corresponding slope of the dispersion relation is zero, such that the rate Γ_{sp} in Eq. (2) is greatly enhanced. One might think that, in Fig. 1(b), the slope seems to approach zero in the limit of large K. Therefore, there should also be the enhanced feature there. However, one should note that the SE rate also depends on the strength of the electric field, i.e., the contribution from the numerator in Eq. (2). For the case of $n=0$, the overall effects do not give the enhanced phenomenon. The main difference between this Letter and [10] should be reminded here. In [10], only the fundamental guided mode ($n=0$) is considered. All other modes have cutoff in the limit of vanishing nanowire radius ($R\to 0$). It was found that large decay rate enhancement can be obtained over broad wavelength regions by enhanced mode confinement. However, this Letter considered the enhancement owing to large density of states at resonant conditions.

Let us now put another QD close to the wire; the interaction between the wire and QDs can now be written as $H' = \Sigma_{n,k_z} D_{n,k_z}(\sigma_{1+}+\sigma_{2+}e^{ik_z z_0})a_{n,k_z} + h.c.$, where a_{n,k_z} is the surface plasmon operator, σ_{j+} is the creation operator of the jth QD, and $D_{n,k_z}(\propto \mathbf{d}\cdot\mathbf{E})$ is the coupling strength. Note that we have assumed 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 two dots. If one further assumes that only QD-1 is initially excited, the state vector of the system can be written as

$$|\Psi(t)\rangle = b_1(t)|\uparrow\downarrow;0\rangle + b_2(t)|\downarrow\uparrow;0\rangle + \sum_{n,k_z} b_{n,k_z}(t)|\downarrow\downarrow;1_{n,k_z}\rangle, \tag{3}$$

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(-2) is excited, while $|\downarrow\downarrow;1_{n,k_z}\rangle$ represents that both the QDs are deexcited with the presence of a single surface plasmon; $b_1(t)$ and $b_2(t)$ can be obtained easily by solving the time-dependent Schrödinger equation.

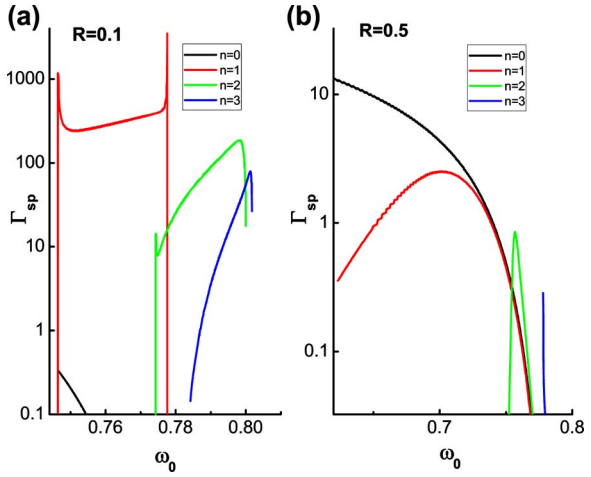

Fig. 2. (Color online) SE rates (Γ_{sp}) into $n=0-3$ modes for (a) $R=0.1$ and (b) $R=0.5$. The unit of Γ_{sp} is normalized to free-space decay rate (γ_0).

Figure 3 shows the time variations of $\mathrm{Re}[b_2(t)]$, $\mathrm{Im}[b_2(t)]$, and $|b_2(t)|^2$ for different values of the exciton bandgap. 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 the (quasi-)stationary limit [14] 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 just like a one-dimensional superradiant one with the populations written as

$$\begin{cases} b_1(t) = e^{-2\Gamma_{\mathrm{sp}}t}(1+e^{2\Gamma_{\mathrm{sp}}t})/2, \\ b_2(t) = e^{-ik_0z_0-2\Gamma_{\mathrm{sp}}t}(-1+e^{2\Gamma_{\mathrm{sp}}t})/2. \end{cases} \tag{4}$$

From Eq. (4), 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 [15]. 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. 3 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.

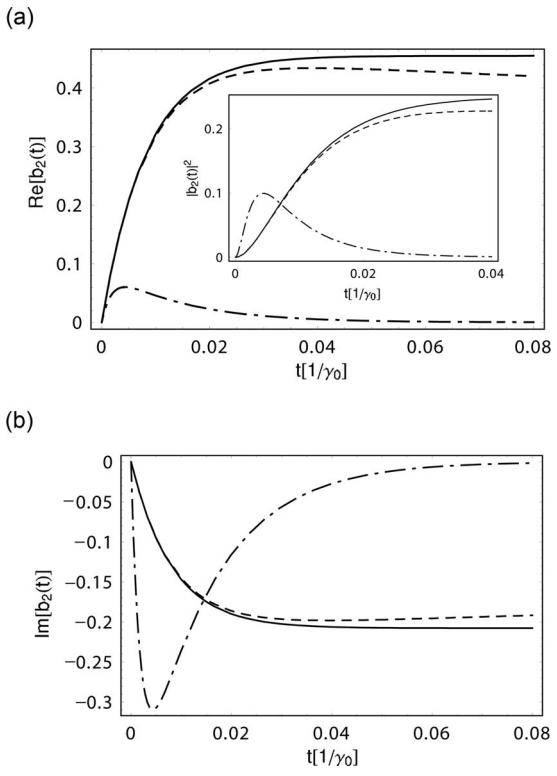

Fig. 3. Variations of $\mathrm{Re}[b_2(t)]$, $\mathrm{Im}[b_2(t)]$, and $|b_2(t)|^2$ [inset of (a)] as functions of time for $\omega_0=0.602\omega_p$ (solid curves) and $0.748\omega_p$ (dashed–dotted curves). In plotting the figures, the interdot distance z_0 is set equal to $0.35(\omega_p a/c)$ with 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 $\Gamma_{\mathrm{non}}(\sim\gamma_0)$.

In summary, we have shown that SE of colloidal QD excitons into surface plasmons can be enhanced even more strongly at certain values of the exciton bandgap. The enhancement is due to zero slope in dispersion relation. Applications of such a phenomenon in cavity quantum electrodynamics and entanglement generation are also pointed out and deserve to be tested with current technology [16].

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References and Notes

1. R. H. Ritchie, Phys. Rev. **106**, 874 (1957).
2. H. A. Atwater, Sci. Am. **296**, 56 (2007).
3. C. A. Pfeiffer, E. N. Economou, and K. L. Ngai, Phys. Rev. B **10**, 3038 (1974).
4. R. Zia and M. L. Brongersma, Nat. Nanotechnol. **2**, 426 (2007).
5. W. L. Barnes, A. Dereux, and T. W. Ebbesen, Nature **424**, 824 (2003).
6. J. Takahara, S. Yamagishi, H. Taki, A. Morimoto, and T. Kobayashi, Opt. Lett. **22**, 475 (1997).
7. H. F. Arnoldus and T. F. George, Phys. Rev. A **37**, 761 (1988).
8. A. Neogi, H. Morkoç, T. Kuroda, and A. Tackeuchi, Opt. Lett. **30**, 93 (2005).
9. R. Paiella, Appl. Phys. Lett. **87**, 111104 (2005).
10. D. E. Chang, A. S. Sørensen, P. R. Hemmer, and M. D. Lukin, Phys. Rev. Lett. **97**, 053002 (2006).
11. D. E. Chang, A. S. Sørensen, E. A. Demler, and M. D. Lukin, Nat. Phys. **3**, 807 (2007).
12. J. A. Stratton, *Electromagnetic Theory* (McGraw-Hill, 1941).
13. From the numerical results, the imaginary parts of ω for the bound modes are actually very small ($10^{-4}\sim10^{-5}$ of the real parts).
14. In fact, the exciton eventually decays into free space. Since the coupling to the surface plasmons is so strong, one can roughly neglect the effect from vacuum fluctuations in the regime of $t<1/\gamma_0$.
15. Y. N. Chen, D. S. Chuu, and T. Brandes, Phys. Rev. Lett. **90**, 166802 (2003).
16. A. V. Akimov, A. Mukherjee, C. L. Yu, D. E. Chang, A. S. Zibrov, P. R. Hemmer, H. Park, and M. D. Lukin, Nature **450**, 402 (2007).