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 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 \( n \)th surface plasmon mode’s components of the electromagnetic field at the surface can be obtained by solving Maxwell’s equations in a cylindrical geometry (\( \rho \) and \( \varphi \) 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:

\[
S(k_z, \omega) = \left[ \frac{\mu_1 J_n^i(K_\omega)}{K_\omega J_n(K_\omega)} - \frac{\mu_0 H_n^{(1)}(K_\omega)}{K_\omega H_n^{(1)}(K_\omega)} \right] \times \left[ \frac{(\omega/c)^2 \varepsilon_\omega J_n^i(K_\omega)}{\mu_1 J_n(K_\omega)} - \frac{(\omega/c)^2 \varepsilon_\omega H_n^{(1)}(K_\omega)}{\mu_0 K_\omega H_n^{(1)}(K_\omega)} \right] - n^2 k_z^2 \left[ \frac{1}{(K_\omega)^2} - \frac{1}{(K_\omega)^2} \right] = 0,
\]

where \( K_\omega^2 = \omega^2 \varepsilon_\omega/\omega^2 - k_z^2 \) (\( \zeta = I \) or \( O \)) and \( \phi_\omega = \exp(i\varphi + ik_z z - i\omega t) \). Here, \( J_n(K_\omega) \) and \( H_n^{(1)}(K_\omega) \) are Bessel and Hankel functions, respectively. \( I \) (O) stands for the component inside (outside) the wire. The dielectric function is assumed as \( \varepsilon_\omega = \varepsilon_\omega [1 - \omega^2/\omega(\omega + i/\tau)] \), where \( \varepsilon_\omega = 9.6 \) (for Ag), \( \varepsilon_\omega = 5.3 \) (for GaN), and \( \tau \) is the relaxation time due to ohmic metal loss [9]. The magnetic permeabilities \( \mu_1, \mu_0 \) 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 \( R (= \omega_0 a/c) \) is roughly equal to 53.8 nm. As can be seen, the behavior of these curves is very similar to the two-dimensional case [12]. This is because the fields for the \( n = 0 \) mode are independent of the azimuthal angle \( \varphi \). However, the behaviors for the \( n \neq 0 \) modes are quite different as shown in Figs. 1(c) and 1(d). The first interesting point are the discontinuities around \( \omega/c = k_z \). Further analysis shows that the solutions of \( \omega \) are “almost real” [13] as \( k_z > \text{Re}[\omega]/c \). In this case, the first kind Hankel function of order \( n \), \( H_n^{(1)}(K_\omega) \), decays exponentially. This
dispersion relation is zero, such that the rate $\Gamma_{sp}$ 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

$$H' = \sum_{n,k_z} D_{n,k_z}(\alpha_{n,k_z} + \alpha^*_{n,k_z})\alpha_{n,k_z} + h.c.,$$

where $\alpha_{n,k_z}$ is the surface plasmon operator, $\alpha^*_{n,k_z}$ is the creation operator of the $j$th QD, and $D_{n,k_z}(\mathbf{z} \cdot \mathbf{E})$ 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 $\pm k\Delta z$, where $\Delta z$ 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

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

(3)

with $b_1(0)=1$ and $b_2(0)=b_{n,k_z}(0)=0$. Here, $|\uparrow\downarrow;0\rangle$ and $|\downarrow\downarrow;1_{n,k_z}\rangle$ represent that both the QDs are decoupled with a single surface plasmon; $b_1(t)$ and $b_2(t)$ can be obtained easily by solving the time-dependent Schrödinger equation.
Figure 3 shows the time variations of $\text{Re}[b_2(t)]$, $\text{Im}[b_2(t)]$, 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 $\Gamma_{sp}$. In this case, the system is just like a one-dimensional superradiant one with the populations written as

$$
\begin{align*}
 b_1(t) &= e^{-2\Gamma_{sp}t}/\left(1 + e^{2\Gamma_{sp}t}\right)/2, \\
 b_2(t) &= e^{-i\omega_0 t - 2\Gamma_{sp}t}(-1 + e^{2\Gamma_{sp}t})/2.
\end{align*}
$$

(4)

From Eq. (4), one realizes that there is always a 50% chance for the two dots to evolve into the state $|\uparrow\downarrow\rangle$. It means that, for example, the singlet [triplet] entangled state can be created if $k_{zp}=(2m + 1)\pi/2m\pi$ 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 $\Gamma_{non}$ which is assumed to be roughly equal to $\gamma_0$. As seen, the (quasi-)stationary state is not greatly degraded in the short time regime $t\ll1/(\Gamma_f + \Gamma_{non})$. In other words, as long as the SE rate into surface plasmons is large enough, the generated entanglement is still observable.

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

13. From the numerical results, the imaginary parts of $\omega$ for the bound modes are actually very small ($10^{-4}~10^{-5}$ 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 $t\ll1/\gamma_0$.