Quantum detection and ranging using exciton-plasmon coupling in coherent nanoantennas

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We utilize interaction of a laser field with a quantum dot-metallic nanoshell system to investigate nanoscale detection and ranging using quantum coherence. We demonstrate that the nanoshell in this system can act as a coherent nanoantenna capable of designating each position in its range with unique space-time field coordinates. This shows that coherent exciton-plasmon coupling in such a system allows the electric field of this antenna generates position-dependent dynamics in molecules and nanostructures in its vicinity, allowing their remote detection. The results are obtained considering the ultrafast polarization dephasing of the quantum dot at elevated temperatures. © 2013 AIP Publishing LLC. [http://dx.doi.org/10.1063/1.4807603]

Interaction of hybrid systems consisting of one semiconductor quantum dot (QD) and one metallic nanoparticle (MNP) with a laser field has been shown to be an appealing host for investigation of many interesting effects and applications. In such systems quantum coherence generated in the QDs can normalize the plasmonic fields of the MNPs, forming significantly different type of fields (called here as coherent-plasmonic fields or CP fields).1–3 Coherent plasmon-exciton coupling in these systems can also lead to a wide range of applications. These include control of the speed of light,4 gating of the energy transfer from QDs to MNPs,5 bistability and hysteresis,6,7 and induction of Rabi frequency.8 Quantum coherence in QD-MNP systems has also been investigated to control energy dissipation in MNPs,1,5,9–13 and even reversing the course of energy transfer, allowing the flow of energy happens from the MNPs to the QDs.14

The objective of this paper is to show the CP field of a system consisting of a QD and a metallic nanoshell (NS) allows the NS to act as a coherent nanoantenna capable of using quantum coherence for detection and ranging of nanoscale biological molecules and nanostructures. We demonstrate theoretically that using such an antenna one can remotely detect the spatial coordinates and movement of such molecules and structures when they are optically active. As schematically shown in Fig. 1, for this we show that the CP field of a QD-NS can designate each point around the NS with unique space-time coordinates (field properties). Therefore, the emission of a nanoscale biological molecule or a nanocrystal, for example, at a given point exhibits a specific time dependency which is unique to that point. We show such space-time field coordinates can be controlled via variation of the intensity and frequency of the laser field responsible for the generation of quantum coherence and by changing the size of the QD. The quantum detection and ranging (QuDAR) introduced in this paper may offer alternative ways for biological sensing applications and presents unprecedented concepts in electromagnetism and fields, in general.

In this paper, we include the effects of ultra-fast dephasing rates (short decoherence time) of QDs, which is the prime requirement for investigation of QuDAR at elevated temperatures. As shown recently by one of us,15 a unique feature of QD-MNP (and QD-NS) systems is that in such systems quantum coherent effects, as those discussed in this paper, can sustain themselves even when the pure dephasing times of the QDs are in the range of couple of hundreds of femtoseconds. The results of this paper show that with creative use of material properties and application of NSs, one can design nanostructure assemblies that can support QuDAR with relatively long range (up to 50 nm) in the presence of ultra-fast dephasing times. Therefore, these results suggest a unique range of quantum coherence effects and nanodevices that can happen or perform at elevated and even room temperatures.

The system considered in this paper is schematically shown in Fig. 1. It includes a QD and an Ag NS (AgNS) with inner and outer radii \( R_1 = 4 \) and \( R_2 = 6 \) nm, respectively, and a core with dielectric constant \( \varepsilon_c = 7 \) (Cu_{2}O). The center-to-center distance between the QD and AgNS is \( R = 9 \) nm. The QD is considered to be made of CdSe-based material with dielectric constant \( \varepsilon_q = 6.25 \) embedded in ZnSe with dielectric constant \( \varepsilon_{0q} = 7.1 \).16,17 We consider the NS is an environment with dielectric constant \( \varepsilon_{0m} = 1.77 \) (as water). The

FIG. 1. Schematic illustration of quantum detection and ranging and coherent nanoantenna. The antenna includes a QD and a metallic NS. \( F(r, t) \) and \( G(r', t') \) refer to two different time-dependent emission intensity patterns of an optically active nanoscale system at two different locations \( (r, t) \) and \( (r', t') \).
In the above equation, the 1–2 transition is assumed to have frequency \( \omega_0 \) and resonantly excited by a laser beam with the frequency \( \omega(t) = E_0(t) \cos(\omega_0 t) \). The amplitude of this laser \( E_0(t) \) is considered to have a step-like rise change and it is polarized along the axis of the QD and the NS. We adopt a semiclassical approach wherein the NS and interaction of the QD-NS system with the laser beam are treated classically, while the QD is addressed quantum mechanically.

Polarization dephasing in the QD is not restricted to real transitions, rather virtual transitions involving elastic exciton-phonon scattering (pure dephasing) can play a dominant role, especially at elevated temperatures. The excitons in the QD structure considered in this paper are strongly confined and their energy separations are large. Therefore, we assume the diagonal electron-phonon interactions mostly determine the pure dephasing process.\(^{18}\) Additionally, although the rate of pure dephasing can change with the laser amplitude,\(^{19}\) in the following we treat it independent of the laser intensity. This approximation is supported by the fact that, as shown in this paper, in QD-NS systems the rate of Forster resonance energy transfer (FRET) from the QDs to the NSs can be significantly high. Therefore, it can be observed, to some extent, the time dependent changes of pure dephasing. Considering these issues, in the following we treat such a dephasing process in a phenomenological way. Under these conditions, the semiclassical Hamiltonian of the QD system in the dipole and rotating wave approximation is given by\(^ {2}\)

\[
H_{\text{sc}} = \sum_{i=1,2} \hbar \omega_i \sigma_{ii} + \hbar \Omega_{\text{eff}} \sigma_{21} e^{-i\omega t} + H.C. \tag{1}
\]

In the above equation, H.C. stands for Hermitian conjugate and \( \sigma_{ii} = |i\rangle\langle i| \) with \( i \) and \( j \) = 1 and 2. \(|1\rangle\) and \(|2\rangle\) refer, respectively, to the ground and excited states of the QD (exciton) with \( \hbar \omega_1 \) and \( \hbar \omega_2 \) energies. The 1–2 transition refers to the transition between these two states and \( \omega_0 = \omega_2 - \omega_1 \). \( \Omega_{\text{eff}} \) refers to the normalized Rabi frequency of the QD given by\(^ {2}\)

\[
\Omega_{\text{eff}} = \frac{\Omega_{12}}{\hbar} = \rho_{21} \eta_{\text{NS}}. \tag{2}
\]

Here, \( \eta_{\text{NS}} = \frac{4 \mu_{\text{NS}}^{2} R_{\text{NS}}}{\hbar \omega_{\text{eff}}^{2} R_{\text{NS}}^{2}} \), where \( \omega_{\text{eff}} = \frac{2 \omega_{1} + \omega_{2}}{3 \omega_{0}} \) and \( \Omega_{\text{eff}}^{2} = -\frac{\mu_{12} E_{0}}{2 \hbar \omega_{\text{eff}}} \left(1 + \frac{2 \beta_{\text{NS}}}{R_{\text{NS}}^{2}}\right) \). \( \beta_{\text{NS}} \) is the polarizability of the NS given by

\[
\beta_{\text{NS}} = \frac{R_{\text{NS}}^{3} \left[\omega_{\text{NS}}(\omega_{\text{NS}} + 2 \omega_{m}) - \omega_{\text{NS}}(\omega_{\text{NS}} - 2 \omega_{m})/2\right] + \omega_{\text{NS}}(\omega_{\text{NS}} - 2 \omega_{m})/2}{2 R_{\text{NS}}^{3} \left[\omega_{\text{NS}}(\omega_{\text{NS}} + 2 \omega_{m}) + \omega_{\text{NS}}(\omega_{\text{NS}} - 2 \omega_{m})/2\right] + \omega_{\text{NS}}(\omega_{\text{NS}} - 2 \omega_{m})/2}. \tag{3}
\]

\( \rho_{21} \) in Eq. (2) refers to the off-diagonal element of the density matrix of the QD. Additionally, the \( \leftrightarrow \) sign in Eq. (1) emphasizes on the self-normalization of the Hamiltonian caused by its dependency on \( \rho \). \( \mu_{12} \) is the dipole moment of the QD transition, considered to be 0.65 \( e \times \text{nm}^{2} \).

Considering Eqs. (1)–(3), the density matrix element in Eq. (2) \( \rho_{21} \) and the other elements can be obtained from the following:

\[
\dot{\rho}_{11} = -2i\hbar [\Omega_{\text{eff}}^{\text{NS}} \rho_{21}, \Sigma_{\text{FRET}}] + \Gamma_{22} \rho_{22} - \Gamma_{11} \rho_{11}, \tag{5}
\]

\[
\dot{\rho}_{22} = 2i\hbar [\Omega_{\text{eff}}^{\text{NS}} \rho_{21}, -\Sigma_{\text{FRET}}] - \Gamma_{22} \rho_{22}, \tag{6}
\]

\[
\dot{\rho}_{21} = -i[\hbar \omega_{0} - \omega_{0}, + i\hbar [\eta_{\text{NS}}] \Sigma_{\text{FRET}] + \gamma_{12}^{\text{dep}} \rho_{21} - \Omega_{\text{eff}}^{\text{NS}} J_{\text{NS}}. \tag{7}
\]

Here, \( \delta = \rho_{11} - \rho_{22} \) and \( \Sigma_{\text{FRET}} = -2i\hbar [\eta_{\text{NS}}] \rho_{21} \) refer to the carrier relaxation induced by the FRET from the QD to the NS, \( \gamma_{12}^{\text{dep}} \) in Eq. (7) refers to the total polarization dephasing rate of the QD given by

\[
\gamma_{12}^{\text{dep}} = \Lambda_{\text{FRET}} + \gamma_{\text{FRET}}^{r} + \gamma_{12}^{\text{dep}}, \tag{8}
\]

where \( \Lambda_{\text{FRET}} = Im[\eta_{\text{NS}}] \delta \) refers to the normalized FRET rate. \( \gamma_{12}^{\text{dep}} = \frac{1}{2}[\Gamma_{1} + \Gamma_{2}] \) is the dephasing rate caused by radiative decay and \( \gamma_{12}^{\text{dep}} \) is the rate of pure dephasing rate, representing the contribution of elastic phonon-exciton scattering.\(^{18,19}\)

To find out the CP field at a given point \( \mathbf{L} \) at the distance of \( z \) from the center of the NS (Fig. 1), we need to calculate the field generated by direct excitation of the plasmons in the NS and the excitons in the QD, and by the plasmons induced by the dipole of the QD. Considering the above discussion, the amplitude of the CP field generated at the point \( \mathbf{L} \) can be obtained from

\[
E_{\text{plas}}(z) = \frac{\beta_{\text{NS}}}{\epsilon_{\text{NS}} e^{3}} \left( E_{0} + \frac{4 \mu_{12}^{2} \rho_{21}}{3 \epsilon_{\text{eff}}^{3} R_{\text{NS}}^{2}} \right) + \frac{2 \mu_{12} \rho_{21}}{\epsilon_{\text{NS}} e^{3} \epsilon_{\text{eff}}^{3} (R + z)^{2}}. \tag{9}
\]

Having this, we can calculate coherent plasmonic field enhancement defined as

\[
P_{\text{coh}}(z) = \frac{|E_{\text{plas}}(z) + E_{0}/2\epsilon_{\text{NS}}|^{2}}{|E_{0}/2\epsilon_{\text{NS}}|^{2}}. \tag{10}
\]

\( P_{\text{coh}} \) represents the ratio of the effective field intensity at the location of \( \mathbf{L} \) in the presence of the NS \( (I_{\text{eff}}) \) to that in the absence of it \( (I_{0}) \), i.e., \( I_{\text{eff}} = P_{\text{coh}} I_{0} \).

To investigate QuDAR, we consider the \( \gamma_{12}^{\text{dep}} = 1 \text{ ps}^{-1} \). This corresponds to a linewidth of about 9 meV. We consider \( \Gamma_{2} = 0.01 \text{ ns}^{-1} \) and the risetime of the laser field, with intensity \( I_{0} \) inside the QD, happens at 100 ns of the time scale (Fig. 2a, inset). The results in Fig. 2 show the intensity of the effective field experienced by the QD \( (I_{\text{eff}}) \) as \( z \) is varied from 7 to 35 nm. Here, we consider \( I_{0} = 26.7 \text{ W/cm}^{2} \) and the laser photon energy \( (h \nu_{\text{L}}) \) is 2.28 eV. We also consider the 1–2 transition energy \( (h \nu_{\text{L}}) \) is the same. Figs. 2a and 2b suggest that at each coordinate \( I_{\text{eff}} \) has a characteristic time dependence. With respect to the applied field, for \( z = 7–11 \text{ nm} \) (Fig. 2a) \( I_{\text{eff}} \) is delayed \( (\tau_{p}) \). After the delay, \( I_{\text{eff}} \) increases significantly, suggesting a strong field enhancement. For larger values of \( z \), the relative values of \( I_{\text{eff}} \) during \( \tau_{p} \) start to increase. As seen in Fig. 2b, for \( z = 15 \text{ nm} \) \( I_{\text{eff}} \) during \( \tau_{p} \) becomes even larger than that after the delay.

Figs. 2c–2f show the corresponding changes in the real and imaginary parts of the electric field (in statvolt/cm) around the NS for the same changes in \( z \). The results suggest that, depending on the value of \( z \), the real part can have negative or positive values, while the imaginary part remains...
always positive (Figs. 2(c) and 2(e)). Additionally, for $z$ around 15 nm the real part of the electric field during $\tau_p$ can become much larger than that after the delay. This trend diminishes for larger values of $z$. These results suggest a significant dependency of the phase and amplitude of electric field around the NS on $z$.

To better visualize coherent designation of time-space field coordinates around the NS, in Fig. 3(a) we plot the normalized field form factor defined as the ratio of $I_{\text{eff}}(z,t)$ to its value at $t = 300$ ns (well after the time delay). The results show that during $\tau_p$ this ratio changes significantly from about 0.05 to about 1.4 (thick solid line) and then reduces to 1 as $z$ becomes large. Therefore, each $z$ has its own specific field time dependency, different from other points. These results show that the range of space-time designation by the CP field can even go beyond 35 nm (dashed-dotted line).

The mode of coherent space-time field designation can be controlled by changing the transition energy of the QD, which can be readily achieved by controlling its size. To see this we consider the same system as that in Fig. 2 but assume $\hbar \omega_0 = 2.1$ eV and the laser field intensity inside the QD ($I_0$) is 4.9 W/cm$^2$. The results in Fig. 4(b) show that with the increase of $z$ the value of $I_{\text{eff}}$ during $\tau_p$ reach its values after this delay, but does not go beyond it. These results also suggest that the impact of space-time designation remains
distinct even when $z = 50 \text{ nm}$ (thick solid line). Similarly, Figs. 4(c)–4(f) show that the electric field (in statvolt/cm) around the NS is quite different than that in the case of Fig. 2. This suggests that by changing the QD size very different field properties can be generated.

The way $I_{\text{eff}}$ is changed with $z$ in Fig. 4 is further clarified in Fig. 3(b) where we show its field form factor. Comparison between the results in this figure and those in Fig. 3(a) highlights the characteristic changes in the space-time field coordinates generated by CP field as the QD transition energy is changed. The distinctive differences between the two modes of coordinate designations shown in Figs. 2 and 4 are illustrated in Fig. 5(a), where we show the ratio of $I_{\text{eff}}$ in the middle of the delay time ($t_p/2$) to that for $t > t_p$ (denoted as field factor ratio), as a function of $z$. This ratio is the key factor showing how coherent designation for each set of parameters happens. The results show that for the case of the system studied in Fig. 4 (squares) this ratio undergoes an exponential-like change with $z$, which could be more suitable for QuDAR application.

Note that the time delay seen in Fig. 2(a) is caused by the laser detuning ($Re[\hbar_{\text{NS}}]\delta$) and the broadening ($\Lambda_{\text{FRET}}$) of the QD transition caused by coherent plasmon-exciton coupling (Eq. (7)).21 In the case where one considers $\omega_0 \neq \omega_i$, $Re[\hbar_{\text{NS}}]\delta$ does not present the whole detuning of the laser from the 1–2 transition. Additionally, our results show that QD-NS systems can support significantly high rates for FRET from the QD to the NS. To see this in Fig. 5(b), we show the results for $\Lambda_{\text{FRET}}$ for the two systems studied above. The results suggest that this rate starts with very high values before the risetime of $I_0$ (thick solid line). Right at the risetime, $\Lambda_{\text{FRET}}$ starts to decline. During $t_p$, it reduces and after $t_p$ it levels off to a relatively small value. These results show how coherent effects can normalize the FRET process, which by itself is an incoherent process.

Note that the time delay ($t_p$) can be changed by varying the laser intensity. Therefore, one can further control the way field changes at each position with time by choosing the value of $I_0$. For the results shown in Fig. 4, for example, as $I_0$ becomes less than 4.9 W/cm$^2$ the value of $t_p$ increases in a nonlinear fashion. To see this and demonstrate variation of FRET from the QD to the NS with time for the system in Fig. 4, in Fig. 6 we show FRET-induced exciton recombination ($\Sigma_{\text{FRET}}$) as $I_0$ is reduced. Note that the value of $t_p$ can ultimately become infinite when $I_0$ reaches a critical value. Additionally, the results show that $\Sigma_{\text{FRET}}$ is only significant during $t_p$. This suggests that FRET happens only during a

![FIG. 5. (a) Variation of ratio of $I_{\text{eff}}$ at $t_p/2$ to that at $t > t_p$ as a function of $z$ for the cases considered in Fig. 2 (circles) and Fig. 4 (squares); (b) shows the variations of $\Lambda_{\text{FRET}}$ with time for these cases. The thick solid line in (b) shows the profile of the applied laser intensity.](image)

![FIG. 6. Variation of FRET-induced exciton annihilation ($\Sigma_{\text{FRET}}$) as a function of time for various $I_0$ (legends in W/cm$^2$). Other specifications are the same as those in Fig. 4.](image)

![FIG. 7. Field form factor for the system considered in Fig. 2 at $z = 15 \text{ nm}$ for different values of $R_1$ (legends in nm). All other specifications are the same as those in Fig. 2.](image)
given period of time, offering gating of the energy flow, despite very large pure dephasing rate.

In this paper, we utilized the prime advantage of the NS that allowed us to adjust its plasmon frequency to match that of the QD transition. For the structure considered in this paper, our calculations showed that for different values of \( R_1 \), as long as this condition remains valid, one can adjust the intensity of the laser to reach similar results. Fig. 7 shows the impact of variation of \( R_1 \) on the field form factor of the system considered in Fig. 2 for \( z = 15 \text{ nm} \). The results suggest that for very different \( R_1 \), the features seen in Fig. 2 do not happen (thin solid and dashed-dotted lines). This is because of the fact the plasmon peak frequencies in these cases are very different from that of the QD transition. One can revive these features by adjusting the parameters of the QD to allow its transition frequency falls around that of the plasmon peak of the NS.

In conclusion, the results suggested that the CP field of the QD-NS system allows the NS to act as coherent plasmonic antenna that can designate each point in its range with specific field properties that are unique to that point. The strongly position-dependent variations of the effective field intensity can determine the optics of the biological molecules and nanocrystals. Therefore, their detection and ranging can be done by detecting variations of their emission patterns as they move.

\[ \text{References} \]