Cooperative emission of light by an ensemble of dipoles near a metal nanoparticle: The plasmonic Dicke effect

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We identify a new mechanism for cooperative emission of light by an ensemble of N dipoles near a metal nanostructure supporting a surface plasmon. The cross talk between emitters due to the virtual plasmon exchange leads to the formation of three plasmonic superradiant modes whose radiative decay rates scale with N, while the total radiated energy is *thrice* that of a single emitter. Our numerical simulations indicate that the plasmonic Dicke effect survives nonradiative losses in the metal.

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Radiation of a dipole near a metal nanostructure supporting a surface plasmon (SP) is attracting renewed interest due to possible biosensing applications [1]. While early studies mainly focused on fluorescence of molecules near rough metal films [2], recent advances in near-field optics and in chemical control of molecule-nanostructure complexes spurred a number of experiments on single metal nanoparticles (NPs) linked to dye molecules [3–8] or semiconductor quantum dots [9]. Emission of a photon by a dipole-NP complex involves two competing processes: enhancement due to resonance energy transfer (RET) from an excited dipole to the SP [10] and quenching due to energy exchange with optically inactive excitations in the metal [11]. These decay channels are characterized by radiative Γ^r and nonradiative $\Gamma^{\rm nr}$ decay rates, respectively, and their balance is determined by the separation d of the emitter from the metal surface [12,13]. The emission is most enhanced at some optimal distance and is quenched close to the NP surface due to suppression of the quantum efficiency $Q = \Gamma^r/(\Gamma^r + \Gamma^{\rm nr})$ by prevalent nonradiative processes. Both enhancement and quenching were widely observed in fluorescence experiments on Au and Ag nanoparticles [3–8]. In recent *single-molecule* measurements [6–8], the distance dependence was in good agreement with *single-dipole-NP* models [12,13], prompting proposals for a NP-based nanoscopic ruler [8].

In this Letter, we identify a novel mechanism in the emission of light by an *ensemble* of dipoles located near a nanostructure supporting a localized SP. A typical setup would involve, e.g., dye molecules [3–5] or quantum dots [9] attached to a metal NP via DNA linkers. We demonstrate that RET between individual dipoles and SP leads to *cross talk* between the emitters. As a result, the emission of a photon becomes a *cooperative* process involving *all* dipoles in the ensemble *and* the NP. This *plasmonic* mechanism of cooperative emission represents an extension, to plasmonic systems, of the Dicke effect for N radiating dipoles in free space, confined within a volume with characteristic size a smaller than the radiation wavelength λ [14–16]. The Dicke mechanism of cooperative emission is photon exchange between the emitters that gives rise to superradiant (SR) states with total angular momentum $l = 1$ and enhanced radiative decay rate $\sim N\Gamma_0^r$, where Γ_0^r is the decay rate of an *isolated* dipole. In plasmonic systems, as we show below, the dominant mechanism is *SP exchange*, i.e., excitation of a virtual SP in a nanostructure by an excited dipole followed by its absorption by another dipole, rather than direct radiative coupling [see Fig. 1]. This SP-induced coupling between dipoles leads to the formation of *plasmonic SR* states that dominate the emission of a photon. Importantly, because SP extends throughout the nanostructure, the latter acts as a *hub* that couples nearby and remote dipoles with about equal strengths, so the SP-induced cross talk is more uniform throughout the ensemble, as compared to the radiation coupling, and leads to a more efficient hybridization and, hence, cooperative emission.

The usual photonic Dicke effect could be inhibited by internal nonradiative transitions in molecules or by their energy exchange with environment. In fact, nonradiative processes are even more pronounced when emitters are located near a metal nanostructure. In particular, along with the optically active ($l = 1$) SP, a dipole also excites dark ($l > 1$) plasmons that dissipate in the metal without converting their energy to radiation, and so the *photon*

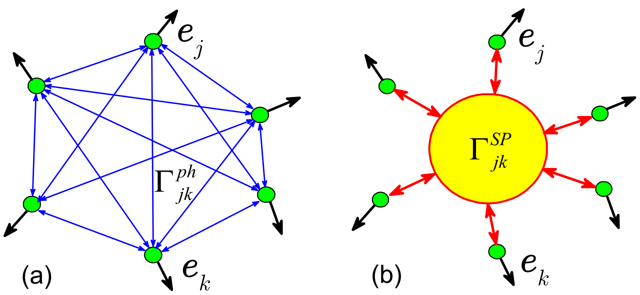

FIG. 1 (color online). (a) Radiative coupling of emitters in free space and (b) plasmonic coupling of emitters near a NP.

exchange processes are largely quenched by Ohmic losses. In contrast, since *plasmon exchange* between dipoles involves *both* bright and dark plasmon modes, the plasmonic Dicke effect is *not* significantly affected by nonradiative transitions up to very small dipole-NP distances and thus provides the main mechanism for cooperative emission in plasmonic systems. Specifically, we show that, for an ensemble of N dipoles distributed around a metal NP, there are *three* plasmonic SR states (with total angular momentum $l = 1$) that dominate the emission, with radiative decay rates $\gamma^r_\mu \sim N\Gamma^r/3$. Furthermore, in a wide range of dipole-NP distances, their *nonradiative* decay rates also scale as $\gamma^{\mathrm{nr}}_\mu \sim N\Gamma^{\mathrm{nr}}/3$, so that the *SR quantum efficiencies* are essentially the *same* as those of individual dipoles near a NP. As a result, the total energy radiated by an ensemble W is only *thrice* that radiated by a single dipole near a NP W_0:

$$W \simeq 3(\hbar kc/4)Q = 3W_0, \tag{1}$$

where k and c are the wave vector and the speed of light, and the remaining energy is dissipated in the NP via subradiant states. The fact that, in plasmonic systems, radiated energy of an ensemble is nearly independent of its size could allow unambiguous determination of single-emitter decay rates in common situations when a large but uncertain number of emitters participate in radiation.

Theory.—We consider a system of N emitters, e.g., fluorescing molecules, with dipole moments $\mathbf{d}_j = d_j\mathbf{e}_j$, where d_j and $\mathbf{e}_j$ are their magnitudes and orientations, respectively, located at positions $\mathbf{r}_j$ around a spherical NP of radius R in a dielectric medium with its center at origin. We assume incoherent emission, i.e., molecules initially excited by a laser pulse subsequently relax through internal transitions before emitting a photon, and adopt the classical model of Lorentz oscillators with random initial phases. The frequency-dependent electric field $\mathbf{E}(\mathbf{r}, \omega)$, created by all dipoles in the presence of a NP, satisfies Maxwell's equation

$$\frac{\epsilon(\mathbf{r}, \omega)\omega^2}{c^2}\mathbf{E}(\mathbf{r}, \omega) - \boldsymbol{\nabla}\times\boldsymbol{\nabla}\times\mathbf{E}(\mathbf{r}, \omega) = -\frac{4\pi i\omega}{c^2}\mathbf{j}(\mathbf{r}, \omega), \tag{2}$$

where dielectric permittivity $\epsilon(\mathbf{r}, \omega)$ is that of the metal inside NP $\epsilon(\omega)$, for $r < R$, and that of the outside dielectric ϵ_0, for $r > R$. Here $\mathbf{j}(\mathbf{r}, \omega) = -i\int_0^\infty e^{i\omega t}\mathbf{j}(t)dt$ is the Laplace transform of dipole current $\mathbf{j}(t) = q\sum_j \dot{d}_j(t)\mathbf{e}_j\delta(\mathbf{r} - \mathbf{r}_j)$, where dipole displacements are driven by the electric field at dipoles positions

$$\ddot{d}_j + \omega_0^2 d_j = \frac{q}{m}\mathbf{E}(\mathbf{r}_j, t)\cdot\mathbf{e}_j, \tag{3}$$

with the initial conditions $\mathbf{d}_j = d_0\mathbf{e}_j\sin\varphi_j$, $\dot{\mathbf{d}}_j = \omega_0 d_0\mathbf{e}_j\cos\varphi_j$, and $\mathbf{E} = \mathbf{0}$ for $t = 0$ (the dot stands for time derivative). Here ω_0, q, m, and φ_j are oscillator frequency, charge, mass, and initial phase, respectively ($\omega_0 = \hbar/2md_0^2$). Closed equations for $d_j(\omega)$ are obtained by Laplace transforming Eq. (3) with the above initial conditions and then eliminating $\mathbf{E}$ from Eqs. (2) and (3) [17]. The latter can be expressed via normalized displacements $v_j(\omega) = d_j(\omega)/d_0 - i(\omega_0/\omega^2)\cos\varphi_j - \omega^{-1}\sin\varphi_j$ and $v_{0j} = -i(\omega_0^3/\omega^2)\cos\varphi_j - (\omega_0^2/\omega)\sin\varphi_j$, as

$$\mathbf{E}(\mathbf{r}, \omega) = \frac{4\pi d_0 q\omega^2}{c^2}\sum_j \mathbf{G}(\mathbf{r}, \mathbf{r}_j, \omega)\cdot\mathbf{e}_j v_j, \tag{4}$$

where $\mathbf{G}(\mathbf{r}, \mathbf{r}', \omega)$ is the electric field Green dyadic in the presence of a NP. For the photon frequency close to those of dipoles $\omega \approx \omega_0$, we arrive at the following system:

$$\sum_k[(\omega_0 - \omega)\delta_{jk} + \Sigma_{jk}]v_k = \frac{v_{0j}}{2\omega_0} = \frac{-i}{2}e^{-i\varphi_j}, \tag{5}$$

where the complex *self-energy matrix* Σ_{jk} is given by

$$\Sigma_{jk}(\omega) = -\frac{2\pi q^2\omega_0}{mc^2}\mathbf{e}_j\cdot\mathbf{G}(r_j, r_k; \omega)\cdot\mathbf{e}_k. \tag{6}$$

The system (5) determines *eigenstates* of N emitters coupled to each other via radiation field *and* electronic excitations in a NP. For $|\mathbf{r}_j - \mathbf{r}_j| \ll \lambda$, we can use the near-field expansion of the Mie theory Green dyadic $\mathbf{G}(r_j, r_k; \omega)$ [13] for calculation of Σ_{jk}. The details will be given elsewhere [18]. The self-energy is dominated by its imaginary part that contains a SP pole, and the *decay matrix* $\Gamma_{jk} = -\mathrm{Im}\Sigma_{jk}$ is a sum of radiative and nonradiative terms $\Gamma_{jk} = \Gamma^r_{jk} + \Gamma^{\mathrm{nr}}_{jk} + \delta_{jk}\Gamma^{\mathrm{nr}}_0$, where

$$\begin{aligned}\Gamma^r_{jk} &= \Gamma^r_0[(\mathbf{e}_j\cdot\mathbf{e}_k) - \alpha'_1(K^{(1)}_{jk} + \mathrm{H.c.}) + |\alpha_1|^2T^{(1)}_{jk}],\\ \Gamma^{\mathrm{nr}}_{jk} &= \frac{3\Gamma^r_0}{2k^3}\sum_l \alpha''_l T^{(l)}_{jk},\end{aligned} \tag{7}$$

and Γ^{nr} accounts for internal molecular transitions (δ_{jk} is the Kronecker symbol). Here $\Gamma^r_0 = 2q^2d_0^2k^3/3\hbar\epsilon_0$ is a dipole radiative decay rate ($k = \sqrt{\epsilon_0}\omega/c$ is a wave vector), $\alpha_l(\omega) = \alpha'_l(\omega) + i\alpha''_l(\omega) = \frac{R^{2l+1}[\epsilon(\omega)-\epsilon_0]}{\epsilon(\omega)+(1+1/l)\epsilon_0}$ are l-pole nanoparticle polarizabilities, and matrices $T^{(l)}_{jk}$ and $K^{(l)}_{jk}$ are

$$\begin{aligned}T^{(l)}_{jk} &= \frac{4\pi}{2l+1}\sum_{m=-l}^{l}[\mathbf{e}_j\cdot\boldsymbol{\psi}_{lm}(\mathbf{r}_j)][\mathbf{e}_k\cdot\boldsymbol{\psi}^*_{lm}(\mathbf{r}_k)],\\ K^{(l)}_{jk} &= \frac{4\pi}{2l+1}\sum_{m=-l}^{l}[\mathbf{e}_j\cdot\boldsymbol{\psi}_{lm}(\mathbf{r}_j)][\mathbf{e}_k\cdot\boldsymbol{\chi}^*_{lm}(\mathbf{r}_k)],\end{aligned} \tag{8}$$

respectively, where $\boldsymbol{\psi}_{lm}(\mathbf{r}) = \boldsymbol{\nabla}[r^{-l-1}Y_{lm}(\hat{\mathbf{r}})]$ and $\boldsymbol{\chi}_{lm}(\mathbf{r}) = \boldsymbol{\nabla}[r^lY_{lm}(\hat{\mathbf{r}})]$, $Y_{lm}(\hat{\mathbf{r}})$ being spherical harmonics.

Radiative and nonradiative decay matrices in Eq. (7) describe two different types of coupling in plasmonic systems. The former, Γ^r_{jk}, can be viewed as a *SP-enhanced photon exchange*, similar to Dicke radiative coupling but with local-field enhancement of dipole moments, while the latter, $\Gamma^{\mathrm{nr}}_{jk}$, represents nonradiative coupling of dipoles via all plasmon modes. From diagonal elements, single-dipole-NP rates can be easily recovered for normal ($s = \perp$) and parallel ($s = \parallel$) orientations with respect to the NP surface [12]: $\Gamma^r_s = \Gamma^r_0|1 + a_s\alpha_1/r_0^3|^2$ and

$\Gamma_s^{\mathrm{nr}} = (3\Gamma_0^r/2k^3)\sum_l b_s^{(l)}\alpha_l''/r_0^{2l+4}$, where $a_\perp = 2$, $b_\perp^{(l)} = (l+1)^2$ and $a_\parallel = -1$, $b_\parallel^{(l)} = l(l+1)/2$.

Radiated energy in the unit frequency interval is obtained by integrating the spectral intensity over the solid angle $dW/d\omega = (c\epsilon_0/4\pi^2)\int|\mathbf{E}(\mathbf{r},\omega)|^2 r^2 d\Omega$ and averaging the result over the initial phases of oscillators φ_j. Here the *far-field* $\mathbf{E}(\mathbf{r},\omega)$ is given by Eq. (4), where v_j is the solution of Eq. (3) and $\mathbf{G}(\mathbf{r},\mathbf{r}_j,\omega)$ is the *large* r asymptotics of the Mie Green dyadic [13]. The result reads [18]

$$\frac{dW}{d\omega} = \frac{1}{4\pi}\,\mathrm{Tr}\Big[\frac{\sqrt{\epsilon_0}\hbar\omega_0}{(\omega-\omega_0-\hat{\Sigma})(\omega-\omega_0-\hat{\Sigma}^\dagger)}\hat{\Gamma}^r\Big]. \quad (9)$$

In the *absence* of coupling, i.e., for purely diagonal $\Sigma_{jk} = -i\delta_{jk}\Gamma$ and $\Gamma_{jk}^r = \delta_{jk}\Gamma^r$, the frequency integration recovers radiated energy of N isolated dipoles near a NP $W = N(\sqrt{\epsilon_0}\hbar\omega_0/4)Q = NW_0$.

To illustrate the role of SP coupling, first consider N dipoles randomly distributed in a solid angle around a NP at the *same* distance $d \gtrsim R$ from its surface, with normal or parallel orientations. At such distances, the high-momenta ($l > 1$) terms in Eq. (7) are negligible, and decay matrices take the simple form $\Gamma_{jk}^r = \Gamma_s^r A_{jk}$ and $\Gamma_{jk}^{\mathrm{nr}} = \Gamma_s^{\mathrm{nr}} A_{jk}$, where $A_{jk} = \mathbf{e}_j \cdot \mathbf{e}_k$ is the cosine matrix and Γ_s^{nr} includes only the $l = 1$ term ($s = \perp, \parallel$). We now introduce *cooperative decay* matrices as $\gamma_{\mu\nu}^r = (N\Gamma_s^r/3)B_{\mu\nu}$ and $\gamma_{\mu\nu}^{\mathrm{nr}} = (N\Gamma_s^{\mathrm{nr}}/3)B_{\mu\nu}$, where $B_{\mu\nu} = (3/N)\sum_j e_{j\mu}e_{j\nu}$ is a 3×3 matrix in coordinate space with $\mathrm{Tr}\hat{B} = 3$. Note now that, since $\mathrm{Tr}\hat{A}^n = \mathrm{Tr}(N\hat{B}/3)^n$ for any integer n, the $N \times N$ matrices $\Gamma_{jk}^{r,\mathrm{nr}}$ have only *three nonzero eigenvalues* coinciding with those of matrices $\gamma_{\mu\nu}^{r,\mathrm{nr}}$. Therefore, only these eigenvalues contribute to the spectral function

$$\frac{dW}{d\omega} = \frac{\sqrt{\epsilon_0}\hbar\omega_0}{4\pi}\sum_{\mu=1}^{3}\frac{Q_\mu\gamma_\mu}{(\omega-\omega_0)^2+\gamma_\mu^2}, \quad (10)$$

$$Q_\mu = \frac{\gamma_\mu^r}{\gamma_\mu} = \frac{\Gamma_s^r}{\Gamma_s^{\mathrm{nr}} + \Gamma_s^r + (3\Gamma_0^{\mathrm{nr}}/N\lambda_\mu)}, \quad (11)$$

where $\gamma_\mu^r = \lambda_\mu N\Gamma_s^r/3$ and $\gamma_\mu = \lambda_\mu N(\Gamma_s^r + \Gamma_s^{\mathrm{nr}})/3 + \Gamma_0^{\mathrm{nr}}$ are the radiative and total decay rates, respectively, of plasmonic SR states, Q_μ are their quantum efficiencies, and $\lambda_\mu \sim 1$ are eigenvalues of $B_{\mu\nu}$. Note that there are three SR states with total angular momentum $l = 1$ because dipole orientations are not uniform in space. Importantly, *both* radiative and nonradiative rates of SR states are enhanced by factors $\sim N/3$ (for each degree of freedom). However, these factors effectively *cancel out* in the quantum efficiencies Q_μ. Furthermore, the contribution of Γ_0^{nr} in the denominator of Eq. (11) is suppressed by the factor $N^{-1} \ll 1$; i.e., Q_μ are not sensitive to intramolecular relaxation processes. Not too far from a NP, when $\Gamma_0^{\mathrm{nr}} \ll (\Gamma_s^r + \Gamma_s^{\mathrm{nr}})$, SR and single-molecule efficiencies essentially *coincide*: $Q_\mu \simeq Q$. Integrating Eq. (10) over frequency, we obtain Eq. (1).

Numerical simulations and discussion.—Although Eqs. (1), (10), and (11) were derived for moderate distances $d \gtrsim R$, these results apply even close to a NP surface, as demonstrated in our numerical simulations below. In Fig. 2, we show the eigenvalue distribution of full decay matrix $\Gamma_{jk}^r + \Gamma_{jk}^{\mathrm{nr}}$ for 30 emitters with dipoles moments normal to a NP surface that are randomly distributed in a solid angle around the Au NP of radius $R = 16$ nm in an aqueous medium ($\epsilon_0 = 1.77$). We assumed $\Delta = 10\%$ fluctuations around the average NP-dipole distance d to account for stretching and folding of linker molecules [3–5]. NP polarizabilities $\alpha_l(\omega)$ were calculated with the experimental bulk Au complex dielectric function, and angular momenta up to $l = 30$ in Eq. (7) were included in $\Gamma_{jk}^{\mathrm{nr}}$. Calculations were carried at SP energy 2.31 eV, size-dependent Landau damping was incorporated, and we used the value of $\Gamma_0^{\mathrm{nr}} = 1.08 \times 10^9\ \mathrm{s}^{-1}$ for Cy5 dye as the intramolecular nonradiative rate. For $d = 20$ nm, there are only *three* nonzero eigenvalues corresponding to SR states, in agreement with Eq. (10). With decreasing d, the remaining $N - 3$ subradiant states start emerging ($d = 10$ nm), and, at small distances ($d = 5$ nm), all system eigenstates acquire a finite decay rate. Note that similar results hold for any dipole orientations, e.g., for random orientations in a tangent plane.

The above behavior for small NP-dipole distances indicates the crucial distinction between photonic and plasmonic Dicke effects that comes from nonradiative coupling in the latter. For dipoles in free space, the system eigenstates are eigenvectors of *radiative* decay matrix $\hat{\Gamma}_0^r$ and represent super- or subradiant modes characterized by the strength of their coupling to radiation. Similarly, in the presence of a NP, these modes are defined as eigenvectors of the *plasmon-enhanced* radiative decay matrix $\hat{\Gamma}^r$

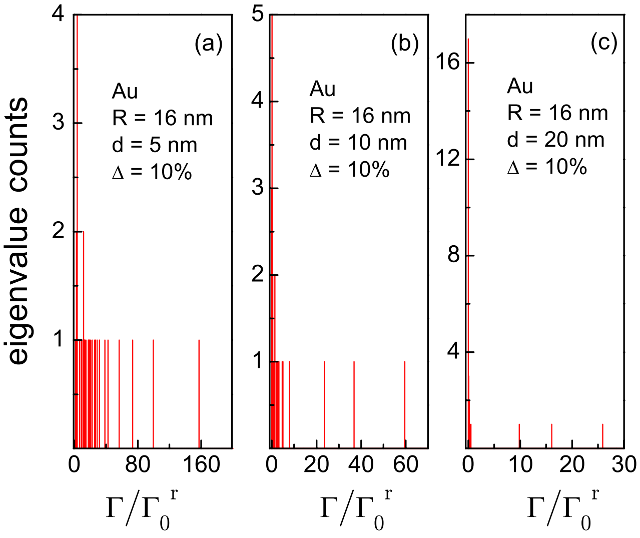

FIG. 2 (color online). Distribution of decay rates for 30 dipoles around a Au NP at several average (with 10% fluctuations) distances to its surface.

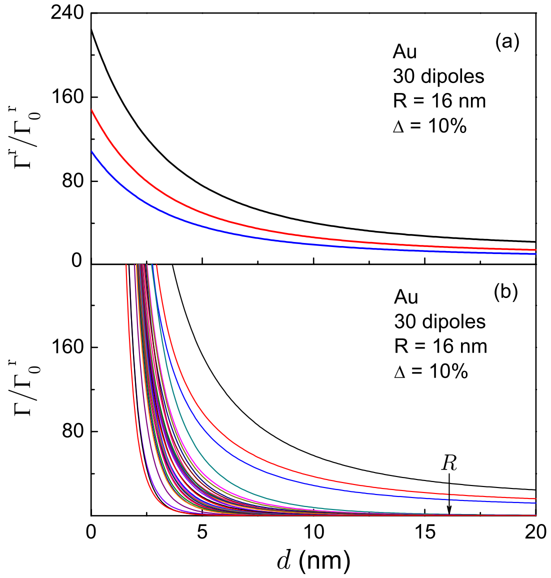

FIG. 3 (color online). Eigenvalues of (a) radiative Γ_{jk}^r and (b) full Γ_{jk} decay matrices vs the average distance to a NP surface for 30 dipoles randomly distributed around a Au NP.

[Eq. (7)] *regardless* of dipole-surface separation [see Fig. 3(a)]. However, the *true* system eigenstates $|j\rangle$ are described by the *full* decay matrix $\hat{\Gamma}^r + \hat{\Gamma}^{\mathrm{nr}}$ with eigenvalues Γ_j, and their radiative decay rates are given by expectation values $\Gamma_j^r = \langle j|\hat{\Gamma}^r|j\rangle$, with quantum efficiencies $Q_j = \Gamma_j^r/\Gamma_j$. Then, close to the NP, nonradiative coupling via high-l excitations leads to the *mixing* of SR and subradiant modes [see Fig. 3(b)], giving rise to a finite decay rate for all eigenstates. However, except for very small distances, this mixing is *weak* and cooperative behavior largely persists. In Fig. 4, we compare the distance dependence of *combined* ensemble quantum efficiencies $Q_{\mathrm{ens}} = \sum_j Q_j$ for 30, 60, and 100 molecules to the single-molecule Q near a $R = 16$ nm Au NP. For distances $d \gtrsim R/2$ (8 nm), all Q_{ens} collapse into a single curve with amplitude $3Q$, indicating that the emission is dominated by SR modes. Even closer to a NP surface, up to $d \approx 5$ nm, the emission remains cooperative although deviations from $3Q$ behavior appear. For smaller d, the eigenstates are no longer SR and subradiant modes, and cooperative emission is destroyed.

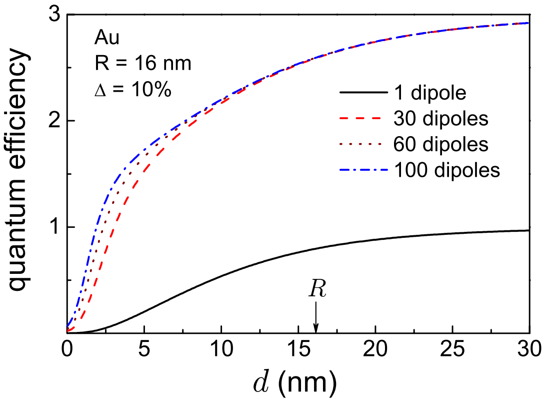

FIG. 4 (color online). Combined quantum efficiencies for ensembles of 30, 60, and 100 dipoles compared to that for an isolated dipole near a Au NP.

The above analysis applies under the standard condition $a \ll \lambda$, which also allowed us to use the long-wave approximation for the Mie Green dyadic. We also disregarded the real part of self-energy with direct dipole-dipole interactions since, in contrast to the photonic Dicke effect [15], here the imaginary part is dominant due to SP resonance, while weak frequency shifts [19,20] have no significant effect on SR modes [17]. Finally, the plasmonic Dicke effect could be observed in experiments with controllable separation of emitters from the NP surface. In recent experiment on Cy5 dyes linked to a Au NP [5], a systematic study of distance dependence for the ensemble fluorescence was performed. Even though the number of emitters was increasing with their separation from a NP surface, a fast saturation of quantum efficiency was observed, consistent with our prediction in Fig. 4.

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