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Possible plasmonic acceleration of LED modulation for Li-Fi applications

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Here plasmonic effect specifically on excited state(s) decay rate is studied in terms of radiative and nonradiative decay rates for a dipole near a metal spherical nanoparticle (NP) taking contribution from scattering into account. Proximity of a metal nanobody promotes faster radiative and non-radiative decay which are shown to be reasonably balanced to get with Ag NPs near 100-fold enhancement of excited state(s) decay rate for a blue LED either without loss in overall LED efficacy or even with gain in LED intensity. Gain in intensity occurs for devices with intrinsic quantum yield $Q_0 < 1$ being higher for lower Q_0 . With silver, rate enhancement can be performed through the whole visible and can be applied for the blue LED electroluminescence as well as for the color conversion by phosphors used in white LEDs. Since phosphors rely on photoluminescence (rather than electroluminescence), local field enhancement factor enables more than 50-fold intensity rise that can be used to compensate possible intensity loss from nonradiative decay. In this case 30-fold rate enhancement is feasible for phosphors without loss in efficacy. Gold NP are found to be efficient in the red only enabling 40-fold rate enhancement for 600 nm and 30-fold one for 650 nm without considerable loss in efficiency. The results are believed to stimulate extensive experimental implementations towards advanced high-speed LEDs for visible light communication systems (Li-Fi). ©2016 American Institute of Physics, [doi XXXXXXXXXX]

Wireless radiofrequency (RF) communication within restricted rooms and confined areas can possibly be replaced by visible light communication (VLC) based on white LED lighting systems. This concept introduced first in 2000 [1,2] has nowadays gained an important notion known as light fidelity, “Li-Fi” in short, after well-known wi-fi [3]. The Li-Fi paradigm easily breaks RF bandwidth restriction, enables security against tapping from outside the room/area covered by a specific Li-Fi hub, inhibits undesirable noising and cross-talk issues and therefore not only can be readily integrated in residential communication services but also in special interference-sensitive areas like hospitals or aircrafts[4]. To make efficient use of white LEDs in Li-Fi networks all possible ways of high-speed modulation should be examined. A typical commercial white LED consists of a blue LED (the most common case is $\lambda_{\text{max}} = 450$ nm emission wavelength maximum) and phosphors converting a portion of blue light into green and red radiation. Therefore, in case that the whole visible is expected to carry on optical data, both blue LED electron-hole pair recombination rate and phosphor active component decay rate may become the critical parameters defining the data processing rate limit. If the phosphor emission is filtered at the receiver end, then only

the blue electroluminescence matters for high-speed optical modulation.

In this paper, we suggest plasmonic enhancement of decay rate for both LED and phosphor in the context of high-speed Li-Fi source. Plasmonic enhancement is well established for photoluminescence (PL) [5-10]. PL intensity enhancement is defined by interplay of local incident field enhancement, and radiative and nonradiative decay rates enhancement promoted by proximity of a metal nanobody. Recently a number of groups have reported on LED intensity enhancement using plasmonic resonances of Ag and Au nanoparticles (NPs) including both quantum well based LEDs^{11,12} and colloidal quantum dot based LEDs,¹³⁻¹⁵ the latter being of special interest in the context of the colloidal nanophotonics platform¹⁶ since metal colloidal NPs can be integrated therein. Enhancement of radiative and nonradiative decay rates promoted by metal nanoparticles means that modulation rate of data transfer systems can be accelerated accordingly. It is important to look for favorable balance of radiative versus nonradiative processes enhancement to enable modulation rate speed-up without undesirable noticeable loss in overall efficiency since dominating enhancement of nonradiative process will result in fall of the intrinsic emission quantum yield Q .

We examine plasmonic effects on decay rates based on the model of light emission by a dipole near a metal spherical NP. The model accounts for the scattering that has been shown¹⁰ to be crucial for NPs bigger than 20 nm, which is the case for the typical experimental situations. It was shown to provide reasonable explanation for above mentioned experimental implementations¹²⁻¹⁴ of LED intensity enhancement¹⁶. Here the multi-parametric problem is examined including the material type (Au and Ag), NP size, metal-emitter spacing, emission wavelength, and intrinsic quantum yield. The results obtained show that plasmonic enhancement of decay rates both for blue LED and color-converting phosphors in white LEDs as well as for green and red LEDs in case of making LED clusters for white light generation are feasible without noticeable loss in overall efficiency or even along with its increase.

We use the model^{10,18} based on a modified radiative decay rate of a dipole emitter near a metal nanobody by means of computing its modified dipole transitions probability. The calculation scheme has been described elsewhere.¹⁸ The most favorable case is considered of a dipole moment oriented along the line connecting an emitter with the NP center. Change in the total decay rate $\gamma = \gamma_0 + \gamma_{\text{int}}$ breaks into plasmonic effect on radiative and nonradiative process,

$$\frac{\gamma^*}{\gamma} = \frac{\gamma_r + \gamma_{nr}}{\gamma_0 + \gamma_{\text{int}}} = Q_0 \frac{\gamma_r + \gamma_{nr}}{\gamma_0}, \quad Q_0 = \frac{\gamma_0}{\gamma_0 + \gamma_{\text{int}}} \quad (1)$$

where γ^* is the modified total decay rate in the presence of a nanobody, γ_0 is the intrinsic radiative spontaneous decay rate, γ_{int} is the intrinsic nonradiative decay rate describing internal losses of an emitter, γ_r and γ_{nr} are the radiative and nonradiative decay rates of an emitter in the presence of a metal nanobody, respectively, and Q_0 is the intrinsic quantum yield of the emitter. For the dipole orientation chosen the required values read,^{2,10,18}

$$\frac{\gamma_r}{\gamma_0} = \frac{3}{2} \sum_{p=1}^{\infty} p(p+1)(2p+1) \left| \frac{\psi_p(k_0 r_0 n)}{(k_0 r_0 n)^2} + A_p \frac{\zeta_p(k_0 r_0 n)}{(k_0 r_0 n)^2} \right|^2, \quad (2)$$

$$\frac{\gamma_r + \gamma_{nr}}{\gamma_0} = 1 + \frac{3}{2} \sum_{p=1}^{\infty} p(p+1)(2p+1) \text{Re} \left\{ A_p \left(\frac{\zeta_p(k_0 r_0 n)}{(k_0 r_0 n)^2} \right)^2 \right\}, \quad (3)$$

where $\psi_p(x) = x j_p(x)$ and $\zeta_p(x) = x h_p^{(1)}(x)$ are Ricatti-Bessel functions, $r_0 = a + \Delta r$ is distance for an NP center to an emitter, k_0 is the wave number in vacuum, n is the medium refractive index, $j_p(x)$ and $h_p^{(1)}(x)$ are the spherical Bessel functions, and

$$A_p = - \frac{\sqrt{\varepsilon} \psi_p(k_0 a \sqrt{\varepsilon}) \psi'_p(k_0 a n) - n \psi'_p(k_0 a \sqrt{\varepsilon}) \psi_p(k_0 a n)}{\sqrt{\varepsilon} \psi_p(k_0 a \sqrt{\varepsilon}) \zeta'_p(k_0 a n) - n \psi'_p(k_0 a \sqrt{\varepsilon}) \zeta_p(k_0 a n)}$$

is one of the Mie coefficients for the field reflected from a NP surface, primes denote derivatives, a is NP radius, and ε is the metal complex dielectric permittivity. When an emitter is moved far from the metal NP, i.e. when $\Delta r \rightarrow \infty$ holds, one has $\gamma_r / \gamma_0 \rightarrow 1$ and $(\gamma_r + \gamma_{nr}) / \gamma_0 \rightarrow 1$.

Fig. 1. Calculated modified radiative decay rate for an emitter near an Ag nanoparticle with diameter 20, 40, 60, 80, and 100 nm and spacing 5, 10, 20, and 30 nm.

Calculations were made for 20...100 nm Ag and Au NPs with $\varepsilon(\lambda)$ function according to Ref.¹⁹ with $n = 1.5$ which is typical for polymers. Notably, Eqs. (1)–(3) show that plasmonic effect on total decay rate does not depend on the absolute values of intrinsic radiative γ_0 and nonradiative γ_{int} decay rates, i.e. it is independent of intrinsic emitter lifetime. Acceleration factor of the total decay rate is entirely defined by Q_0 , emission spectrum, metal dielectric permittivity, nanoparticle size and emitter–metal spacing.

Fig. 2. Calculated extinction spectral of Ag nanoparticles

Acceleration in radiative decay component (Fig. 1) features pronounced size dependence from so-called the morphological resonance arising from light scattering for sized higher than 20 nm. This size-dependent enhancement

spectra remarkably correlate with size-dependent nanoparticles extinction spectra (Fig. 2.).

Fig. 3. The same as in Fig. 1 but for modified nonradiative decay rate.

Enhancement for nonradiative component also features size dependence but it exhibit fast decrease with wavelength and distance. For wavelengths >440 nm it vanishes at spacing >15 nm whereas enhancement of radiative decay measures up to 10-15 at the same conditions. This difference in radiative and nonradiative rate behavior allows for efficient enhancement of the total decay rate without noticeable loss in efficiency. Therefore for every emitter independently of intrinsic quantum yield Q_0 modulation rate at this conditions will rise without efficiency loss since quantum yield will not fall down. For $Q_0 < 1$, nonradiative decay enhancement may become not very valuable for efficiency since it will be added to the existing intrinsic nonradiative rate, and then even higher acceleration of modulation rate can be expected using closer position of the emitter to the metal. E.g., if we consider $Q_0 = 0.5$, then for 450 nm wavelength, 60 nm NP diameter and 5 nm distance one has total decay rate enhancement about 100 times and quantum yield rise up from 0.5 to 0.7. Thus for this example chosen, not only modulation rate will speed up but intensity will rise as well.

Figs. 1-4 show that enhancement is also possible for green, yellow and red LEDs that can be used in case of designing white LED as a cluster consisting of 3-4 discrete LEDs to get options for desirable emission spectrum depending on a consumer's demand.

Fig. 4. The same as in Fig. 1 but for the full decay rate.

For more commonly used white LEDs based on a blue LED together with a color-converting phosphor, metal effect on phosphor photoluminescence should be examined. Enhancement for the total decay rate is feasible for the phosphor spectrum (500-650 nm) by a factor of 20...30 mainly owing to radiative component enhancement, i.e. without overall intensity loss. Additionally, one should bear in mind that for phosphors optical excitation is the case. Therefore the overall photoluminescence intensity I_0 rises to I owing to the blue LED incident local field $\mathbf{E}$ enhancement [10] in accordance with

$$\frac{I}{I_0} = \frac{|\mathbf{E}|^2}{|\mathbf{E}_0|^2} \frac{\gamma_r}{\gamma_r + \gamma_{nr}}. \quad (4)$$

Fig. 5 presents intensity enhancement maps for phosphor excitation by a blue LED. One can see 50 nm silver NPs enable more than 60-fold increase in intensity over the whole phosphor spectrum. This allows for higher overall rate enhancement factors by using closer distance where nonradiative transition rates rises considerably or even dominate over radiative rate. Based on data presented in Figs. 4,5, 30 to 40 rate enhancement factors can be foreseen for phosphor in white LEDs without loss in efficacy or even with simultaneous efficiency enhancement. It is noteworthy that superior photoluminescence enhancement excited at 450 nm inherent for 50- and 60-nm particles remarkably correlates with their extinction maxima very close to the above excitation wavelength (see Fig. 2). Higher enhancement factors can be foreseen for elongated NPs that can be developed by means of colloidal techniques²⁰. Shaping of silver NPs beyond spherical ones will result in morphological resonances where radiative decay enhancement overtakes nonradiative one^{21,22}.

Fig. 5. Enhancement factor for intensity of a phosphor photoluminescence near an Ag nanoparticle with diameter 50 nm (left) and 80 nm (right) as function of emission wavelength and spacing, excitation wavelength is 450 nm. Intrinsic quantum yield $Q_0 = 1$, and ambient medium refractive index $n_m = 1.5$.

Gold NPs are found to be much less efficient in Li-Fi applications (Fig. 6). First, blue LED rate enhancement is not possible at all. Only green and red emitters can be affected by gold NPs, and color converting phosphor rate in white LEDs can also be affected. Second, acceleration strongly changes across green-red spectrum (550-650 nm). Third, in most of cases nonradiative decay dominates and thus modulation acceleration will occur at the expense of losses in quantum yield. For these reasons only a few representative graphs are plotted for Au NPs in Fig. 6. For 550 nm enhancement of nonradiative decay dominates and will cause undesirable efficiency loss. For 600 nm 40-fold total decay rate enhancement is possible with equally enhanced radiative and nonradiative decays. That means, for $Q_0 = 1$ quantum yield will drop to 0.5 but for $Q_0 < 1$ the quantum yield will not change. For 650 nm nearly 30-fold enhancement of the total decay rate is possible without considerable loss in the quantum efficiency. One can see that actually only in the red Au NPs offer rate enhancement without considerable drop of quantum efficacy. It is noteworthy that, unlike silver, shaping of gold nanoparticles is not efficient for the visible since morphological resonance(s) will shift outside the visible to the infrared.

Fig. 6. (Top) radiative, (middle) nonradiative, and (bottom) total decay rate modification for AU nanoparticles at (left) 10 and (right) 20 nm spacing.

In conclusion, the calculations predict considerable acceleration of blue LED decay rate up to 100 times using silver nanoparticles without essential loss in efficacy. Green and yellow emission rate can be enhanced by a factor of 20-30 with loss in efficiency which can be compensated in the case of optically pumped phosphor within a white LED by local incident field enhancement. Red emission rate can be enhanced both with silver and gold nanoparticles by a factor of 30-40. Silver nanoparticles are found to be useful within the whole visible whereas gold ones are efficient mainly in the red. These findings indicate the prospect for the essential improvement of white LED performance in the context of data transfer rate in Li-Fi set-up.

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