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Enhancement of High Order Multipole Fields in Optical Nanoantennas

Upendra Kumar

Author's Affiliations:

Upendra Kumar Department of Physics, G.D. College, Begusarai, Bihar 851101, India.

Corresponding author: **Upendra Kumar,** Department of Physics, G.D. College, Begusarai, Bihar 851101, India.
E-mail: upendra.physics@yahoo.in

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ABSTRACT We have studied a mathematical framework of higher-order multipole fields in the vicinity of optical nanoantennas. We have exemplified at suitably chosen Nanoantennas i.e. a Nano antenna which strongly enhances the quadruple field. The modification of excitation rates in quantum mechanical systems due to this quadrupolar enhancement was studied. The hybrid system consisting of an optical Nano antenna and a quantum mechanical three level system was studied in detail. A properly designed Nano-antenna can excite dipole for bidden transitions in three level systems due to the enhanced higher order multipole fields. The dynamics of the system is strongly altered by the presence of the nanoantenna and cannot be done by the quadrupolar enhancement alone. This can be achieved by enhancing higher order multipole fields near the antenna. A quadrupole transition as the dominant excitation channel in a three level system was considered. It was found that the enhancement of this transition significantly intensify subsequent emission processes with respect to altered emission characteristics. The effects depend on geometrical parameters; the properties of the optical nanoantenna can be tailored and hence allowed for direct implementations in spectroscopies schemes. The obtained results were found in good agreement with previously obtained results.

KEYWORDS Multipole fields, Vicinity, Optical Nanoantenna, Quadrupole Field, Enhancement, Three Level System, Forbidden Transition, Excitation Channel.

1. INTRODUCTION

Nanoantenna is made from a noble metal which supports localized surface plasmon polaritons at distinct frequencies in the visible and near infrared spectral range. Localized surface plasmon polaritons are excited when the electromagnetic radiation is reasonably coupled to the charge density oscillation in the metal. The hot spot intensities exceed the intensity of the external illumination by orders of magnitude. Nanoantennas for controlling and improving the interaction of far field light with other nanoscopic building blocks such as quantum dots, atoms, molecules [1-4]. Ferry et al. [5] and Rockstuhi et al. [6] studied and found their observations pivotal examples in the field of Bio-sensing and photovoltaics. Muskens et al. [7] studied that considered individual hybrid systems, the most important finding consisted in showing that optical nanoantennas can modify the radiative decay rate of emitters. Filter et al. [8] studied the interaction of light with such hybrid systems for highly symmetric optical nanoantennas. Curto et al. [9] studied emission characteristics of hybrid systems entirely governed by the optical Nano-antenna and strongly deviate from the radiation pattern of emitters in free space. The new engineering possibilities pave the way for the development of highly directed single photon source and other applications [10]. Karaveli et al. [11] and Kern et al. [12] studied the interaction of optical nanoantennas with quantum mechanical system by resorting to more or less appropriate approximations. Apart from few exceptions [13], the effect of higher order multipole fields in the vicinity of the antenna has been restricted to the pure electric dipole field. Although experimental indications for the influence of high order multipole fields have been reported by Moskovits et al. [14] and Kawazol et al. [15]. Tojo et al. [16] and Deguchi et al. [17] presented that nondipolar transitions often termed as forbidden tremendously

enhanced due to large higher order field components. Stockman et al. [18] showed that higher order components are used to excite dark modes in plasmonic systems using strong field gradients. Vogel et al. [19] presented that the emission in the vicinity of nanoantennas is also strongly modified with respect to free space.

2. METHOD

The electric field of a plane wave varies spatially as $\exp\left[i\left(\mathrm{k}\cdot\mathrm{x}\right)\right]\approx 1+i\left(\mathrm{k}\cdot\mathrm{x}\right)$ in the limit of $\mathrm{k}\cdot\mathrm{x}\to 0$.Where k is the wave vector that can be assumed as $\left|\mathrm{k}\right|\approx 10^{7}\,\mathrm{m}^{-1}$ for visible light and the characteristic spatial extent of the atomic system being $\langle 1\times 1\rangle = Za_0 \approx 10^{-10} m$ for hydrogenlike atoms. This approximation is reasonable since the first order term in Taylor expansion is three orders of magnitude larger than the second term, which is attributed to both the electric quadrupole and magnetic dipole fields. Hence for the given spectral domain and the usually considered spatial extent of the atomic system, the excitation rates induced by the local quadrupole field of a plane wave are orders of magnitude smaller than those transitions induced by the electric dipole field. Quadrupole transitions are usually to be inaccessible i.e. they are forbidden. Components of octupolar or higher order are even weaker. In the presence of an optical nanoantenna, the situation changes. Such plasmonic structures support highly localized near fields that are characterized by huge gradients. Using a multipole expansion in special coordinates, the electric field in a coordinate system with origin r_0 can be expressed as

$$E\left(\mathrm{x},\omega\right)=\sum_{m,n}\Big[\,p_{mn}\left(\omega;\mathrm{r}_0\right)N_{mn}\left(\mathrm{x}-\mathrm{r}_{0,\omega}\right) +q_{mn}\left(\omega;\mathrm{r}_0\right)M_{mn}\left(\mathrm{x}-\mathrm{r}_{0,\omega}\right)\Big]$$

Where N_{mn} and M_{mn} are spherical harmonics. Multipole expansion in spherical coordinates with p_{mn} and q_{mn} being the complex electric and magnetic multipole coefficients. The order $n=1$ corresponds to electric and magnetic dipoles, where as $n=2$ corresponds to quadrupoles and so on. To simplify the subsequent result, the more familiar Cartesian quadupole components Q_{ij} have been used. They can be obtained from the quadrupole coefficients p_{m2} using linear transformations. Optical nano-antenna supports strong local electric quadrupole fields, an optical nanoantenna consisting of two strongly coupled silver nanospheres, sometimes termed as dimer. Two silver nano-spheres have a radius of 30 nm and are separated by either 3 or 10 nm. With such a separation, quantum effects and / or possible nonlocal material properties of the silver nano-spheres may only constitue a minor contribution. The assumed geometry is in reach of state of the art fabrication techniques and can even be scaled to large arrays of strongly coupled nanospheres. The scattering cross section of such a dimer structure exhibits a strong quadrupole contribution for a plane wave illumination direction parallel to the connecting line of the nanospheres. The enhancement of local quadrupole fields as probed by the quantum mechanical system was found to be strongest if the two nanospheres are illuminated perpendicular to the connecting line with a polarization of the electric field parallel to it. The quadrupole contribution to the far field is negligible for this illumination scenario. Considering plane wave illumination in the given coordinate system, the only non vanishing component of the related quadrupole tensor in free space. This coefficient is linked to the multipole expansion coefficient as used via

$$Q_{xz}=\frac{i}{\sqrt{6}}\left(p_{-12}-p_{12}\right).$$

The local quadratic enhancement of a quadrupole field may be as the ratio

$$\eta_{ij}^{loc}\left(r_0\right)=\left|Q_{ij}^{na}\left(r_0\right)/Q_{xz}^{fs}\left(r_0\right)\right|^2.$$

The integrated enhancement factor

$$\eta_{ij}=\int_{\Omega}\eta_{ij}^{loc}\left(r_0\right)dV$$

with respect to a certain domain Ω. For enhancements obeying certain symmetries, it is convenient to regard a lower dimensional integration. The super scripts fs and na designate the free space and the nanoantenna scenario.

3. RESULTS AND DISCUSSION

Figure (1) (a) shows η_{xz} as a function of excitation wavelength for two different separations of the nanospheres. The dimer is shown in Figure (1) (b). It is seen that for a separation of 3nm, η_{xz} has a maximum of 1.6×10^{6} at 437 nm. Figure (1) (a) also shows η_{xz} for a nanosphere separation of 10 nm. The integrated enhancement factor is approximately two orders of magnitude less than compared to the 3-nm separation. It is found that the enhancement of the electric quadrupole contribution with respect to the near field stems from the strong coupling of the two nanospheres, which critically depends on the separation. All other components of η_{ij} are order of magnitude less η_{xz} and be neglected. Figure (1) (b) shows $\eta_{xz}^{loc}\left(r_0\right)$ in the xz plane for a 3-nm separation at a wavelength of 437nm. The enhancement of the electric quadrupole contribution takes place in a narrow spatial domain between the surfaces of the two nanospheres.

Within the applied framework, the action of an electromagnetic field on an electron with momentum operator p is given by the minimal coupling interaction potential

$$V_I = -\frac{e}{m}A\cdot p + H.c.$$

The weak field approximation has been applied and hence the ponderomotive potential $\frac{e^2}{2m}A^2$ has been neglected. The electromagnetic fields are related to the vector potential A by B = Curl A and E = $i\omega A$- gradU. Choosing the coulomb gauge where $U\equiv 0$ be achieved, the electric field is given by $E = i\omega A$.

A decomposition of the interaction potential yields

$$V_{\mathrm{I}} = -\frac{ie}{\hbar}\left\{\left[H_0, A\cdot \mathrm{x}\right]+\left[A, H_0\right]\mathrm{x}\right\}\cdot$$

Considering three level system which is excited through an electric quadrupole transition and decays via two consecutive electric dipole transitions is shown in Figure 2(a). Only the quadrupolar transition i.e. the transition is dipole forbidden. It was also found that the emission characteristics of the entire system are modified in the presence of nanoantenna.

The dynamics of the three level system is governed by the rate equations. They describe the population of the three energy levels i.e.

$$\dot{n}_0 = \gamma_{10}\cdot n_1 - \Gamma_{02}\cdot n_0, \qquad \dot{n}_1 = \gamma_{21}\cdot n_2 - \gamma_{10}\cdot n_1,$$
$$\dot{n}_2 = \Gamma_{02}\cdot n_0 - \gamma_{21}\cdot n_2.$$

Where γ_{ij} denote the spontaneous decay rates from the *i*th to the *j*th level, Γ_{02} is the excitation rate of the quadrupole transition. The quadrupole transition takes place at a wavelength of $\lambda_{02} = 437$ nm, for which optical nanoantenna provides the maximal enhancement of an electric quadrupole field as shown in Figure (1) (a). The enhancement of the efficiency of the quadrupole transition due to the nanoantenna, the amount of light spontaneously emitted at $\lambda_{10} = 500$ nm per uit time serve as a figure of merit. Thus the rate equations are solved in equilibrium. In this regime luminescence is gen by $\dot{n}_1^{rad} = \gamma_{10}^{rad} n_1$, where γ_{10}^{nonrad} describes the nonradiative decay rate. In free space $\gamma_{10}^{fs} = \gamma_{10}^{rad.fs}$ and $\gamma_{10}^{nonrad.fs} = 0$. In the presence of the nanoantenna, $\gamma_{10}^{nonrad.na}$ is merely determined by the absorption in the metal. Since the quadrupole enhancement of the electric fields varies spatially the same holds for Γ_{02}. The presence of nanoantenna alters the local density of states. Thus the spatial

dependence for the spontaneous decay rates γ_{ij} also has been considered. In the subsequent results $\dot{n}_1^{rad}$ has been chosen as the figure of merit, assessing how efficiency the emission at λ_{10} can be raised due to the enhancement of the electric quadrupole fields. Figure (2) (b) shows $\dot{n}_1^{rad.na} / \dot{n}_1^{rad.fs}$. It was found that $\dot{n}_1^{rad.na}$ can be enhanced by over four orders of magnitude relative to free space. The obtained results were compared with previously obtained results of theoretical and experimental works and were found in good agreement.

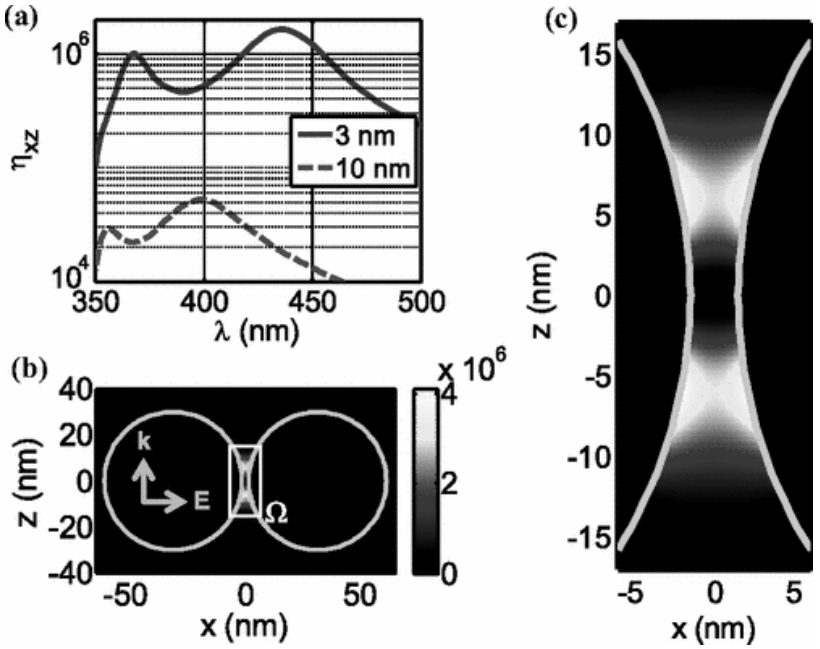

Figure 1: Integrated enhancement factor η_{xz} as a function of excitation wavelength for two separations of the dimer nanospheres under plane-wave illumination.

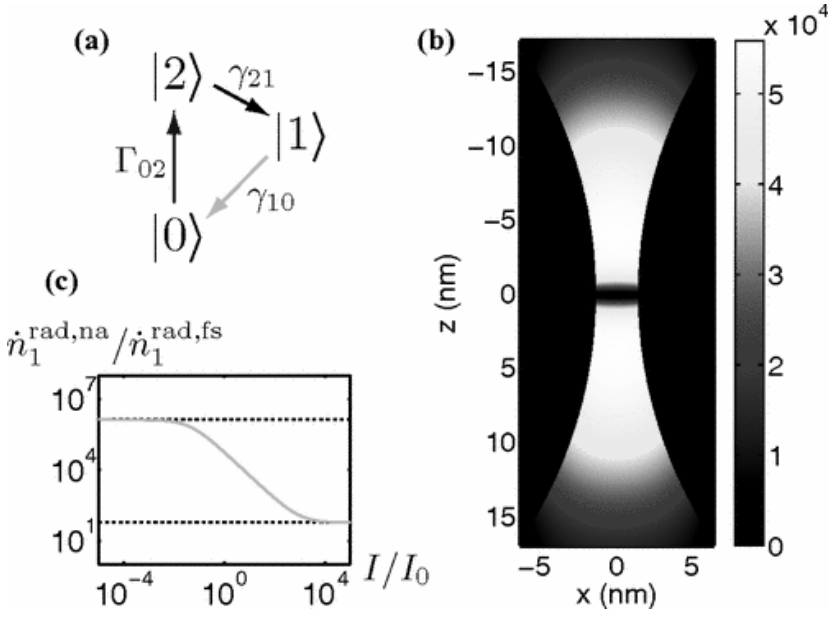

Figure 2: Scheme of the quantum mechanical system that is placed in the vicinity of the nanoantenna.

4. CONCLUSION

We have studied the enhancement of high order multipole fields in optical nanoantennas. The antennas were tailored such that their near field contained sufficiently strong contributions of higher order multipole moments. The strengths of thse moments exceeded their free space analogs by several orders of magnitude. The impact of such excitation enhancement was exemplarity found by studying the dynamics of a three level system. It decays upon excitation by an electric quadrupole transition via two electric dipole transitions. Such self consistent treatment of excitation, emission and internal dynamics as developed in this contribution is the key to predict any observable quantity. It was found that optical nanoantennas effectively enhance higher order multiple transitions. This was achieved by enhancing higher order multiple fields near the antenna. It was also found the enhancement of this transition significantly intenstified subsequent emission processes with respect to altered emission characteristics. The obtained results were found in good agreement with previously obtained results of theoretical and experimental works.

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