Quantum theory for plasmon-assisted local field enhancement

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HIGHLIGHTS

- We model plasmon-induced local field enhancement near gold nanoantenna.
- Bio-inspired asymmetric geometry of nanoantenna is considered.
- Three asymmetrically placed near nanoantenna detectors allow us to resolve the polarization of incoming photons.

ARTICLE INFO

ABSTRACT

We applied quantum theory for nonlocal response and plasmon-assisted field enhancement near a small metallic nanoscale antenna in the limit of weak incoming fields. A simple asymmetric bio-inspired design of the nanoantenna for polarization-resolved measurement is proposed. The spatial field intensity distribution was calculated for different field frequencies and polarizations. We have shown that the proposed design the antenna allows us to resolve the polarization of incoming photons.

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1. Introduction

There is a strong demand for a new generation of optical detectors with a high spatial, frequency and polarization resolution. For many applications, including secure fiber-optical communication and quantum information processing [1], satellite imaging and thermal imaging [2], DNA fluorescence and biochemical sensing [3] or sensing and imaging through biomedical tissue [4], it is necessary to detect a limited number of photons. Single photons are difficult to detect and characterize. A usual way to enhance the interaction between a single photon and matter is to place the photon between two mirrors, creating a high-quality optical cavity [5]. By making multiple round trips the photon is much more likely to interact, for example, with an atom in the cavity. A resonant microcavity with a high quality factor Q, and a mode confined in volume V enhances the density of photon states by a factor known as the Purcell factor Q/V [6]. Creating microcavities with a high Purcell factor is the central target in quantum optics. But there is a limitation: the resonance cavity cannot be made less than the wavelength λ of the photon, which is for visible light is about 400–700 nm.

An alternative and very promising way to facilitate a strong coupling regime between matter and a quantized electromagnetic field is to use plasmonics [7–10]. The interaction of light with metal nanostructures at certain frequencies may result in the formation of collective excitations-plasmons. Plasmons were recently recognized as the key in breaking down the diffraction limit of conventional optics [9,10]. A highly confined electromagnetic field associated with the response of the conduction electrons in a metal nanostructure is not limited by the photon wavelength, and may localize the electric field in a volume of just several cubic nanometers. This results in enormous enhancement of the field–matter interaction assisted by the resonant plasmon excitation [11–13], which may significantly improve the chances of single photon detection. The local-field enhancement was also suggested for waveguide amplifiers in optical integrated circuits to compensate for signal losses [14].

In this work we develop theory for local field enhancement in the presence of a very small metallic nanostructured antenna. We give the quantum mechanical description of both electron and photon degrees of freedom, which is necessary in the case of very small antennas and extremely weak fields. We show that the presence of even a tiny nanostructure, containing several hundred of conduction electrons, one can observe a significant increase of the local field intensity. We have found that placing more than one

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detector near the nanoantenna can facilitate the resolution of the polarization of the incoming photons. A smaller antenna size (under 5 nm) may allow a stronger localization of the incoming photons, however, for the nanostructure sizes less than 1 nm, when the amount of conduction electrons becomes of the order of 100 and less, the plasmon response may become more damped. Therefore, there is an optimal size of the nanoantenna, and for smaller sizes the efficiency of the field localization would drop. For even smaller sizes of the nanoantenna the plasmon peak may split into several molecular-like resonances due to the Landau damping [15].

2. Quantum theory of nonlocal plasmon excitation in quantized electromagnetic field

Let us consider a small metal antenna shown in Fig. 1. The antenna may consist of three or more parts placed under some angle to each other. The design of the antenna is inspired by the shape of phycobilisome, which is a light harvesting antenna of photosystem II in cyanobacteria, red algae and glaucophytes [16]. It is known that the efficiency of the energy transfer by phycobilisome exceeds 95% [17].

Photon detection near the antenna can be realized using either dye molecules [18], nanoscaled quantum dots [19], or donors impurities [20]. The positions of the detectors are shown in Fig. 1 by the circles. The asymmetry of the placement of the detectors is intentional, which will be explained below. An incoming photon can excite a plasmon that will generate a stronger and more localized induced field, which can then be successfully detected. Therefore, the nanoantenna results in a larger effective cross-section of the detectors. But this is not the only advantage. The asymmetric placement of the detectors makes the local field enhancement very sensitive to the polarization of the incoming photon. By comparing the magnitude of the detected photocurrents from each detector one, in principle, may resolve the incoming photon polarization with a high precision. In this work we proposed that a fully functioning antenna–detector complex can be made on the scale under 5 nm. An antenna of this size cannot be manufactured with the currently available mass production lithography methods. One can estimate that with the current pace of the technological progress this spatial resolution will be achieved in the next decade [21]. Using the currently available Surface Tunneling Microscope technique [22], it is still possible to assemble sample nanoantenna devices to perform the measurements.

For ultrasmall nanostructures the dielectric response becomes different from the bulk, and one needs to take into account the nonlocal nature of the dielectric function $\epsilon(\mathbf{r}, \mathbf{r}', \omega)$. To do this let us assume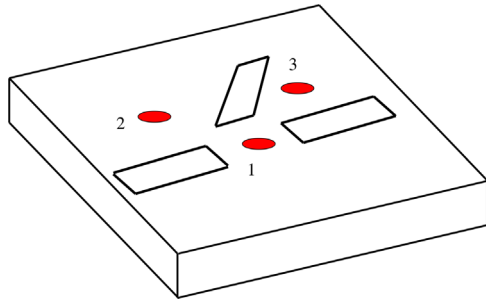 that the conduction electrons in the nanoantenna are confined in an effective trapping potential of the positive background $U_{eff}(\mathbf{r})$ and are interacting with quantized electromagnetic field modes. Using standard second quantization procedure the Hamiltonian of the system can be written as

$$H = H_{el} + H_F + H_{int}, \tag{1}$$

where the Hamiltonian of the electron subsystem H_{el} reads

$$H_{el} = \sum_n E_n c_n^\dagger c_n + \frac{1}{2} \sum_{nmlk} V_{nmlk} c_n^\dagger c_m^\dagger c_l c_k. \tag{2}$$

Here $c_n^\dagger$ (c_n) is the creation (annihilation) electron operator, and V_{nmlk} is the Coulomb matrix element in the eigenbasis $\{\psi_k(\mathbf{r})\}$ of an electron in the trapping potential $U_{eff}(\mathbf{r})$. The Hamiltonian for the electromagnetic field is

$$H_F = \sum_i \hbar\omega_i(a_i^\dagger a_i + 1/2), \tag{3}$$

where $a_i^\dagger$ and a_i are the creation and annihilation photon operators in the mode i with the frequency ω_i and the wavevector $\mathbf{Q}_i$. Under the assumption that for relatively weak fields one may neglect multiphoton processes, the interaction between the electrons and the field is given by

$$H_{int} = \sum_{nm,i} \phi_{nm,i}^{ext} a_i c_m^\dagger c_n + H.\, c. \tag{4}$$

Here the coupling constant $\phi_{nm,i}^{ext}$ is determined by

$$\phi_{nm,i}^{ext} = \left[\frac{\hbar}{2V\omega_i}\right]^{1/2} \frac{ie\hbar}{mc} \int \psi_m^*(\mathbf{r})(e^{i\mathbf{Q}_i\mathbf{r}}\nabla)\psi_n(\mathbf{r})\, d\mathbf{r}, \tag{5}$$

where V is the field quantization volume, m and e are the electron mass and the charge, and c is the speed of light. The gradient operator ∇ takes the usual form: $\nabla = (\frac{\partial}{\partial x}, \frac{\partial}{\partial y}, \frac{\partial}{\partial z})$.

Now let us introduce the density matrix ρ of the total system. It is natural to assume that the system is initially in the product state $\rho(0) = \rho^E(0) \otimes \rho^F(0)$ in terms of the electronic and the field degrees of freedom. The electron quantum subsystem is described by a set of the equilibrium occupations of the electron levels: $\{f(E_n)\}$, where f is the Fermi distribution function and E_n is the correspondent electron energy. As for the initial state of the electromagnetic field, the most general state may be represented in the photon number state basis $\rho_F(0) = \sum_{\{n_i\}} \rho_F^{\{n_i\}\{n_i\}}|\{n_i\}\rangle\langle\{n_i\}|$, where we adopted the notation used in [23] $|n_1, n_2, \ldots n_i, \ldots\rangle = |\{n_i\}\rangle$. For example, the state $|n_1, n_2, \ldots n_i, \ldots\rangle$ has n_1 photons in the 1st mode, n_2 photons in the 2nd mode, and so on.

If the coupling constant $\phi_{nm,i}^{ext}$ is relatively small, one may expand the density matrix of the system as a series $\rho(t) = \rho(0) + \rho_1(t) + \cdots$, and within linear response theory the perturbed density matrix satisfies

$$i\hbar\frac{\partial}{\partial t}\rho_1 = [H_{el} + H_F, \rho_1] + [H_{int}, \rho(0)]. \tag{6}$$

Note that under this approximation one neglects the entanglement between the electron and photon subsystems and possible induced entanglement between different photon modes n_i.

We introduce the perturbed electron density matrix, which can be obtained by applying the trace operation over the field variables $\rho_1^E = \text{Tr}_F\{\rho_1\}$. Applying the trace operation and evaluating the first commutator in Eq. (6), we note that $\text{Tr}_F([\hbar\omega_l a_l^\dagger a_l, \rho_{1nikj}]) = 0$, $\text{Tr}_F([E_p c_p^\dagger c_p, \rho_{1nikj}]) = [E_p c_p^\dagger c_p, \rho_{1nk}^E] = (E_n - E_k)\rho_{1nk}^E$ and $\text{Tr}_F\left(\left[\frac{1}{2}\sum_{nmlk} V_{nmlk} c_n^\dagger c_m^\dagger c_l c_k, \rho_{1piqj}\right]\right) = \left[\frac{1}{2}\sum_{nmlk} V_{nmlk} c_n^\dagger c_m^\dagger c_l c_k, \rho_{1pq}^E\right]$. The latter commutator with the Coulomb term can be calculated using the standard Random Phase Approximation (RPA) for a product of electronic operators [24]: $c_k^\dagger c_q^\dagger c_m c_n \approx c_k^\dagger c_n \langle c_q^\dagger c_m \rangle +$

Fig. 1. The antenna and single photon detectors placed on a dielectric substrate. Incoming photons can lead to plasmon excitation and the local field enhancement next to the asymmetrically placed antenna parts. The local field can be registered by detectors, shown by the circles near the antenna.