

Nano-sensing of the orientation of fluorescing molecules with active coated nano-particles

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Abstract

The potential of using active coated nano-particles to determine the orientation of fluorescing molecules is reported. By treating each fluorescing molecule as an electric Hertzian dipole, single and multiple fluorescing molecules emitting coherently and incoherently in various orientations are considered in the presence of active coated nano-particles. It is demonstrated that in addition to offering a means to determine the orientation of a single molecule or the over-all orientation of the molecules surrounding it, the nature of the far-field response from the active coated nano-particles provides insight into the degree of phase coherence between emitters in specific molecular environments.

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1. Introduction

Fluorescence spectroscopy techniques have become a mainstay in many fields associated with cellular and molecular imaging [1]. They can be used to determine molecular properties down to single-molecule levels [2–6]. The measurements of several single-molecule photo-physical parameters (e.g., distances, emission rates, lifetimes, quantum yields, resonant energy transfer) depend critically on the orientation of

the molecule's absorption- or emission-dipole moment [7–10].

As suggested in the seminal paper by Purcell [11], it is now well known that the lifetime of an excited atomic state is impacted by its local environment. There have been a variety of investigations into how this effect impacts the properties of fluorescence. It was demonstrated that the fluorescence process near an interface can be modeled as the emission of an electric Hertzian dipole (EHD) oriented along the molecule's dipole transition moment in the same local environment [12]. This approach was followed to understand how a single molecule's orientation can be determined from its emission pattern in the presence of glass cover-slip. The resulting measurement techniques rely on the

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presence of a planar substrate on which the effective dipole is located and optical systems to record the emissions [10]. More localized approaches have been sought that are more nano-technology based. For instance, the Purcell-fluorescence connection was further explored in experimental and theoretical studies of the fluorescence rate of a single molecule as a function of its distance from a laser-irradiated gold nanoparticle [13] and in terms of the gold nano-particle acting as an optical nano-antenna [14]. The introduction of other types of optical nano-antennas near fluorescing molecules also has demonstrated the ability to control their emissions [15].

Contrary to these previous works, that rely either on the presence of cumbersome substrates or passive nano-particles, the present work considers the use of coated (core–shell) nano-particles in which gain is encapsulated in their core that act as optical nano-antennas for effective determination of the orientation of fluorescing molecules (which are here modeled as EHDs). It is shown that highly sub-wavelength active nano-particles provide extremely sensitive far-field signatures that highly depend on the orientation, as well as the degree of phase coherence of the surrounding sensed molecules, thereby rendering their use for very effective determination of the orientation of a single-molecule and multiple molecules undergoing fluorescence.

The manuscript is organized as follows. In Section 2 the configuration of a CNP excited by one, two, or four EHDs is described. Section 3 includes the necessary details of the analytical treatment. Section 4 is initiated by a brief account on the material and gain models used in the active CNP configuration. This discussion is followed in Section 5 by a presentation of the highlighted numerical results. Finally, Section 6 summarizes and concludes the present work, and outlines ideas for its extensions. Throughout the manuscript, the time factor $\exp(j\omega t)$, with ω being the angular frequency, and t being the time, is assumed and suppressed.

2. Configuration

The spherical CNP configuration that will be used to sense the molecular orientation is shown in Fig. 1. It consists of a nano-core (radius r_1) covered concentrically with a spherical nano-shell (outer radius r_2), and is immersed in free space. The nano-core and nano-shell have the permittivities, permeabilities, and wave numbers, respectively, denoted by ε_1 , μ_1 , and k_1 , (for the nano-core) and ε_2 , μ_2 , and k_2 (for the nano-shell).

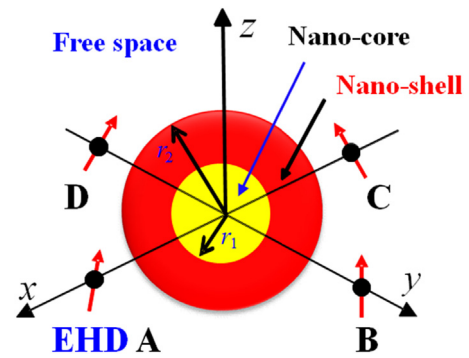


Fig. 1. Multiple electric Hertzian dipoles (EHDs) illumination of a spherical CNP. A gain impregnated SiO₂ core is coated with a Ag shell. The EHDs are represented by the black circles. Three excitation cases are considered: Case 1 (EHD A), Case 2 (EHD A + C) and Case 3 (EHD A + B + C + D). Within each of the excitation cases, the EHDs oriented tangentially or normally, as well as their combinations, with respect to the CNP surface are investigated.

The permittivity and permeability of the ambient free space are ε_0 , μ_0 ; and the corresponding wave number is $k_0 = \omega\sqrt{\varepsilon_0\mu_0}$, and the intrinsic impedance is $\eta_0 = \sqrt{\mu_0/\varepsilon_0}$. As discussed in detail in Section 4, the specific active CNP configuration which will form the basis of this work is a gain impregnated SiO₂ nano-core coated with a Ag nano-shell.

The CNP in Fig. 1 is excited by one or more electric Hertzian dipoles (EHDs), which, following [12], constitute our model of the surrounding molecules. These EHDs are represented by the red arrows overlaid with the black circles and are labeled as A, B, C, and D, in the figure. This is to imply that they can be arbitrarily oriented with the dipole moment of the X th EHD, $X = A, B, C,$ and D , being equal to $\vec{p}_X = \hat{p}_X p_X$, where \hat{p}_X is its orientation and $p_X[\text{Am}]$ is its complex amplitude. A spherical coordinate system (r, θ, ϕ) and the associated rectangular coordinate system (x, y, z) are introduced such that their origins coincide with the center of the CNP.

Three specific excitation cases of the CNP in Fig. 1 are investigated: Case 1, where only EHD A excites the CNP; Case 2, where EHD A and C excite the CNP; and Case 3, where all four dipoles excite the CNP simultaneously. The exciting EHDs are located symmetrically at positions in the xy -plane outside the CNP, i.e., EHDs A and C are centered on opposite sides of the CNP along the x -axis, whereas EHDs B and D are centered on opposite sides along the y -axis. The coordinates of the observation point are (r, θ, ϕ) , and those of the EHDs are $(r_s, \theta_s = \pi/2)$, where $\phi_s = 0$ (EHD A), $\pi/2$ (EHD B), π (EHD C) and $3\pi/2$ (EHD D). Within each of the excitation cases, both tangential and normal EHD orientations with respect to

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