Sección Especial en Óptica Cuántica / Special Section on Quantum Optics

Optical antennas for quantum emitters

Antenas ópticas para emisores cuánticos

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ABSTRACT:

Optical antennas are nanoscale versions of radio antennas and have emerged in recent years as an enabling technology for nanophotonic devices. In particular, they open interesting avenues to control and enhance the emission of single quantum emitters such as molecules and quantum dots. The ability to modify transition rates, polarization and angular radiation patterns of fluorescence could improve the performance of single-photon sources and ultrasensitive biosensors. In this contribution, we review the state of the art of optical antennas, focusing especially on the activities of the Molecular Nanophotonics group at ICFO. We describe the possible resonant modes of nanorod optical antennas, address the controlled coupling of single emitters to a nanoantenna with two different experimental approaches, and show the realization of optical monopole and Yagi-Uda antennas to direct the emission of single emitters.

Keywords: Optical Antennas, Nano-Optics, Single Molecules, Quantum Dots.

RESUMEN:

Las antenas ópticas, equivalente nanométrico de las antenas de radiofrecuencia, se han postulado recientemente como un componente básico para futuros dispositivos nanofotónicos. Concretamente, ofrecen nuevas posibilidades para controlar y mejorar la emisión de emisores cuánticos individuales como moléculas o puntos cuánticos. La capacidad para incrementar las tasas de transición o manipular la polarización y el patrón angular de la fluorescencia de estos objetos cuánticos podría beneficiar el funcionamiento de fuentes de fotones individuales o sensores bioquímicos ultrasensibles. En este artículo, repasamos el estado de desarrollo de la tecnología de antenas ópticas, con especial atención a las actividades desarrolladas en el grupo de Nanofotónica Molecular en ICFO. Describimos los modos de resonancia de antenas formadas por nanohilos y el acoplamiento controlado de emisores individuales y nanoantenas mediante dos métodos experimentales diferentes. La realización de antenas monopolo y de Yagi-Uda permite redirigir la emisión de fotones.

Palabras clave: Antenas Ópticas, Nano-Óptica, Moléculas Individuales, Puntos Cuánticos.

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1. Introduction

Optical antennas are the counterparts of conventional radio-frequency and microwave antennas, operating in the visible regime at a million times higher frequency of ~ 500 THz [1] [2]. These antennas convert propagating radiation (far field) to localized energy (near field), and vice versa. Therefore the history and development of optical antennas is closely related to near-field optical microscopy, since a metal nanoparticle or nanohole can concentrate optical fields to sub-diffraction-limited volumes, surpassing the capabilities of regular bulk optical components. Indeed, like conventional antennas also optical metallic antennas can be much smaller than the wavelength of the electromagnetic wave they receive or emit, which is of direct interest for nanoscale photonic applications. Currently optical antennas are subject of active studies: a rapidly growing number of researchers is exploring opportunities in directions such as nonlinear light-matter interaction, superresolution microscopy, enhancement of photodetection, light emission, sensing and spectroscopy among others [3]. Particularly advances in nanofabrication made it possible to extend the frequency of operation of antennas first to the infrared, where antenna structures are used to improve the performance of photodetectors [4,5]. Now further miniaturization, requiring fabrication accuracies of a few nanometers, is leading to the establishment of the concepts and designs of antenna technology for even shorter light wavelengths, enabling the communication with molecules and quantum dots.

The interfacing of light and matter at the single quantum emitter level is generally achieved by a variety of optical approaches, such as mirror cavities, lenses, optical fibers, photonic crystals or dielectric microcavities, with Q factors surpassing 10^6. In comparison to these conventional components, nanoantennas are relatively poor resonators with Q-factors of only 10-100, yet in compensation antennas do exhibit a subwavelength mode volume. This combination results in a high enhancement of excitation and decay rates, comparable to that of dielectric microcavities with superior Q-factors, however larger volumes. Furthermore, plasmonic nanoantennas offer intrinsic additional advantages such as a much smaller footprint, strongly localized electromagnetic field enhancement, broadband response, and suitability for single-molecule studies.

Nanoscale optical sources come in a large variety such as organic fluorescent molecules, atoms, ions, semiconductor nanocrystals or quantum dots, and vacancy centers in diamond. These quantum emitters are key elements not only in quantum optics but also in biosensing. Both excitation and emission of these emitters can be enhanced by interaction with an optical antenna [6-9]. For example, gold nanoparticles can be regarded as primitive dipole antennas and were shown to enhance the emission of organic fluorophores [6,7], due to an enhancement of the excitation rate and an increase in the radiative transition rate. Especially for poor emitters with low intrinsic quantum efficiency, the antenna can increase the radiation efficiency (external quantum efficiency) [8] thus compensating for the losses due to intrinsic non-radiative decay channels, such as internal conversion. A drawback of emitter-metal interactions is quenching of fluorescence, an increase in the non-radiative rate at very close proximity to the metal surface. Thus in practice one needs to control the distance for an optimal balance.

It should be noted that the local field enhancement achievable with resonant metal nanoparticles is particularly interesting in nonlinear optics. For example higher-harmonic generation in a gas assisted by bow-tie antennas has been demonstrated [10]. The metal itself has a strong nonlinear susceptibility which dominates at the nanoscale [11,12] and such nonlinear optical processes can be exploited to determine the resonant lengths [13] and mode profiles [14] of antenna structures.

Yet another rapidly developing application of the subwavelength local fields around metal nanoantennas is the optical trapping of small dielectric objects, which was demonstrated with gap antennas [15]. Phased arrays have also been proposed for near-field focusing of surface plasmons [16] and freely propagating light [17].

Finally the functionality of antennas is not restricted to enhancing rates or subwavelength focusing. Depending on the specific antenna design one can gain control over the emission or excitation spectrum of quantum emitters, their polarization, and even the directionality of the emitted photons, which is the main subject of this article.

2. Optical antennas in interaction with single quantum emitters

2.a. Metal nanowires as one-dimensional resonators

The operation of antennas commonly relies on resonant electron motion at the surface of the constituting metallic elements. At optical frequencies the metals are non-perfect conductors; the charge oscillation extends into the metal, resulting in a reduced effective wavelength [18,19]. Here one speaks of “plasmons”, collective oscillations of the free electron gas of a metal concomitant with a bound electromagnetic wave. More specifically, metal nanowires exhibit Fabry-Pérot resonances and act as one-dimensional cavity resonators [20]. Their resonances follow a simple linear relation with length that depends on material and geometrical parameters. Modes of increasing resonance order are characterized by an increasing number of longitudinal oscillations in the near field of the structure. Consecutive modes are roughly separated by one half of the effective wavelength of the plasmon in the nanowire. Hence, the first resonance can be found at half the effective wavelength and is called half-wave dipolar mode. The resonant lengths can be sensitively shorter than the free-space wavelength due to the finite skin depth of metals in the visible regime. The sub-wavelength intensity distributions around the nanostructures can only be visualized by near-field scanning optical microscopy [21] [22] or electron beam assisted methods, such as cathodoluminescence and electron energy loss spectroscopy.

Recent progress in nanoscience and nanotechnology has made available several methods for the fabrication of metallic nanowires. Top-down schemes, such as focused ion beam (FIB) milling or electron-beam lithography, offer control on the position and size of individual nanoparticles with 10-20 nm resolution. Alternatively bottom-up colloidal chemistry methods have the advantage of providing spheres and wires of crystalline structure with higher quality optical properties.

2.b. Coupling of a photon emitter to the antenna mode

The interest on single emitters in interaction with optical antennas is twofold. First, the emitters can be enhanced, manipulated and studied thanks to the antenna; second they can be used to drive the antenna modes, as nanoscale local light sources or probes. To feed energy into an optical antenna mode no low-loss coaxial cable is available, as is customary at low (radio) frequencies. As a result, the driving of an antenna must be accomplished either from the far field via scattering of a diffraction limited laser focus, or by near-field coupling of a local emitter. The antenna modes that can be accessed with one method or the other are not necessarily the same. Clearly the near field approach is more specific and allows exploration of all possible modes. Here the challenge is to position a single emitter in the vicinity of the nanoantenna. To this end, we have explored two main approaches: a dynamic approach based on the reversible scanning of the antenna in close proximity over the emitter; and a static approach consisting of dedicated positioning of the emitters on the antennas, however with no reversibility. Both methods offer the possibility of exciting a nanostructure at designated positions as opposed to far field driving of the whole structure at once.

Following the dynamic coupling approach, our group investigated a monopole optical antenna mounted on the tip of a metal-coated scanning probe [23] (Fig. 1a). Monopole antennas are only one quarter of a wavelength long as they are placed perpendicular above a conductive ground plane. Using single fluorescent molecules embedded in a thin polymer film as point probes of the local field amplitude and direction, a geometrical resonance at λ=514 nm was found for an aluminium antenna length of 80 nm. Such an experiment demonstrated the relevance of optical antennas for super-resolution optical microscopy, approaching 25 nanometer resolution (Fig. 1b).

Regarding the static coupling approach, often emitters are randomly spin-cast on the sample, where afterwards one has to search for a suitable emitter-antenna combination [24]. Clearly nanofabrication methods are preferred in order to position the emitters relative to the antenna in a permanent fashion with nanometer accuracy. To this end, at ICFO we have developed a process to locally deposit quantum dots at designated high optical mode density positions. The method relies on a combination of two steps of electron-beam lithography and site-selective chemical functionalization.

Regardless of the practical implementation of the positioning mechanism, the coupling of a dipolar transition to an optical antenna can be calculated analytically with a simple one-dimensional cavity model [20]. This coupling can be summarized in a phase-matching condition between the far field waves and the local plasmonic cavity mode. This model allows us to describe completely the emission by quantifying the radiative decay rate, the quantum efficiency, and the angular emission pattern of the emitter-antenna system. Moreover one gains direct insight in the evolution of the antenna modes as the material parameters evolve from a perfect conductor to a bounded plasmonic mode. As such the model is important to guide any experimental attempt.

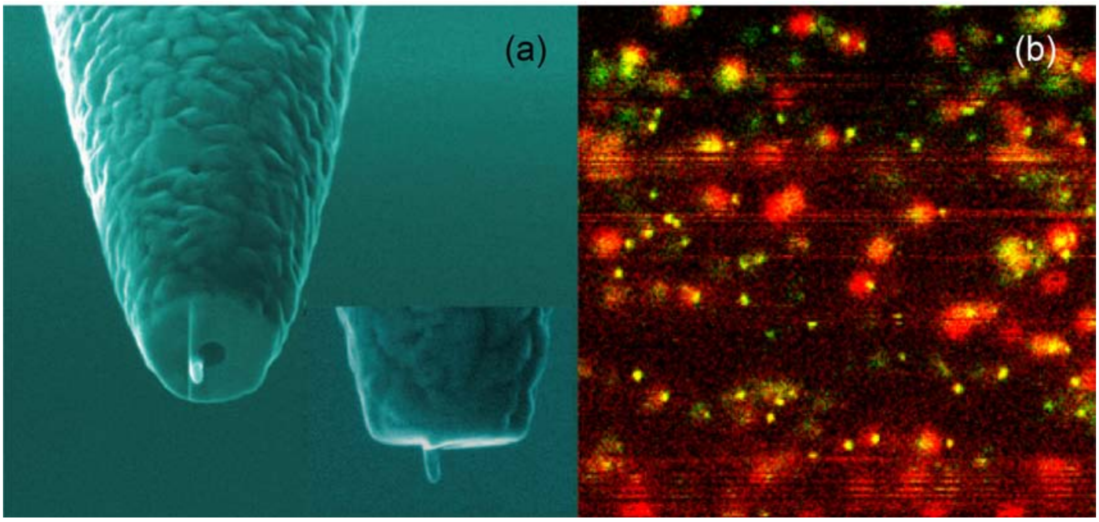

Fig. 1: Optical monopole antenna on a scanning near-field microscope tip. (a) Scanning electron microscopy image of a structure fabricated by focused ion-beam milling. (b) Fluorescence image (4 μm x 4 μm) showing numerous single molecules with 25 nm spatial extent (FWHM) due to the localized excitation field at the tip of the monopole antenna, demonstrating sub-diffraction-limited resolution of λ/20.

2.c. Control of directionality and polarization of a single quantum emitter

Depending on the strength of the emitter-antenna coupling a dramatic modification of the original polarization and angular emission pattern can be expected. Indeed, with the monopole antenna on a scanning probe, our group demonstrated control of the angular emission pattern of individual molecules using polarization-resolved confocal microscopy [25]. By reversible coupling of the emitter to the antenna, it could be proven that the angular emission of the coupled system was fully determined by the antenna mode. Depending on the relative position and orientation of antenna and emitter, both polarization and radiation pattern converted to a dipole oriented parallel to the antenna, perpendicular to the glass substrate. A full rotation by 90 degrees of the polarization or angular pattern could be achieved. It should be noted that such strong redirection of emission can alter the collection efficiency in this kind of experiments, where a high numerical-aperture objective collects the fluorescence of the coupled system. As a result, such changes can have a significant impact on the interpretation of experimental results of metal-enhanced fluorescence, particularly for objectives with a low numerical aperture [26].

One of the most recognizable characteristics of an antenna is its directed emission and reception. Beaming of light from subwavelength objects [27] is highly desirable for applications such as bright single-photon sources for quantum optical technologies or biosensors. However, the radiation pattern of a quantum emitter is typically that of a dipole, a “butterfly” pattern with an angular distribution covering a very wide solid angle. Efficient detection or excitation of a single quantum emitter is thus generally inefficient. Even for the monopole antenna presented before, the angular emission is dipolar and the fluorescence had to be collected with a high-NA objective. Again, Optics can find inspiration in conventional antenna technology to solve this problem; Yagi-Uda antennas are commonly used for directional television reception. These antennas consist of an actively driven feed element surrounded by a set of detuned, parasitic elements: reflector and directors. By coupling a dipolar emitter to the resonant feed element, as proposed theoretically by several authors [28-30], the emission can be directed to a single angular lobe. Therefore, the radiation pattern could be more easily contained within the collection aperture of a reduced numerical aperture lens system. The antenna needs to be tuned with respect to the emission wavelength of the emitter. To this end, 3D numerical calculations (Finite Integration Technique) can be performed [28]. The results clearly show that the interaction of a single quantum emitter with the electromagnetic field is directed by a nano-optical Yagi-Uda antenna, enhancing both excitation and emission rates. The directivity is further increased by the presence of a dielectric substrate.

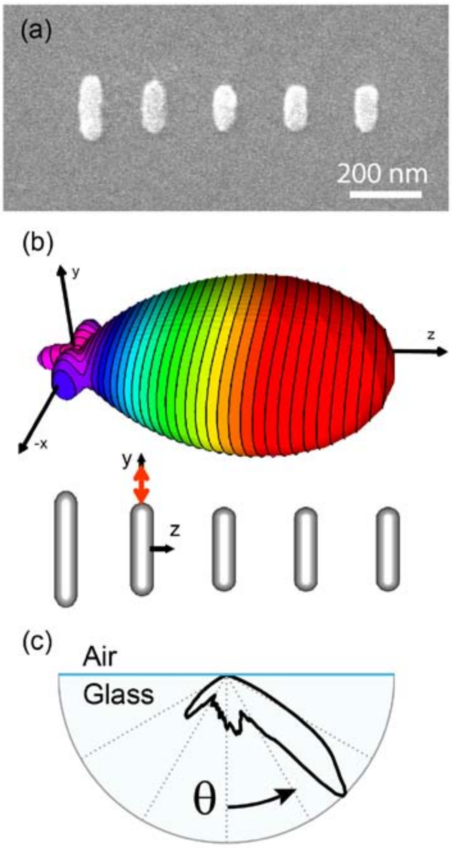

Fig. 2: Optical Yagi-Uda antenna driven by a single emitter. (a) Scanning electron microscopy image of a 5-element gold Yagi-Uda antenna tuned to operate at a wavelength of 800 nm. A quantum dot is deposited at the end of the second, feed element. (b) Numerical simulation of the three-dimensional far-field angular radiation pattern of an aluminum Yagi-Uda antenna in air driven by an oscillating dipole (red arrow). (c) Experimental angular radiation pattern of the structure in a) showing unidirectional emission of a quantum dot by coupling to the mode of the antenna. The directed emission lobe is bent as a result of the presence of a glass substrate.

This type of antenna was first realized experimentally in far-field scattering mode, working with the scattering of a laser beam and separating contributions from different polarizations [31]. The experimental realization of this multi-element optical antenna driven by a quantum emitter requires local driving of the feed element alone, such that the correct phase between the scattering contribution of each element is satisfied. Quantum dots were chosen as emitters, due to their superior photostability. We employed the static approach described previously to position quantum dots at the end of the feed element of Yagi-Uda nanoantennas [32]. In this case, the angular radiation patterns of the antennas were directly imaged in the back-focal plane of the collection objective, where the Fourier space information is readily accessible. With this characterization and fabrication techniques, we demonstrated unidirectional emission of a single emitter by coupling to an antenna. Indeed the resulting quantum-dot luminescence appeared enhanced, strongly polarized, and most importantly directed into a forward lobe.

3. Conclusions

Optical antennas are an attractive tool to help control light-matter interaction at the single-emitter level with nanoscale resolution in a variety of different, innovative ways. We have presented the experimental realization of monopole and Yagi-Uda antennas coupled to a single emitter, showing control over emission direction and polarization. The perspectives for the study of existing radiowave antenna designs translated into the optical regime and the development of structures more suited to the peculiarities and applications of nanoantennas surely offer exciting opportunities for future research. For example, femtosecond phase-controlled excitation of single molecules [33] could lead to an improved control over the photodynamics of such nano-emitters by coupling to metal nanostructures [34]. The ability to fully engineer the electromagnetic environment of a quantum emitter could render visible optical transitions that are forbidden by conventional selection rules. Likewise, the enhanced local electromagnetic fields surrounding optical antennas could lead to strong coupling between an emitter and a metal nanoparticle. On the other hand, the development of broadband near-field concentrators could contribute to the improvement of the efficiency-cost ratio in thin film photovoltaics. These few examples illustrate the opportunities opened by optical antennas for the realization of novel optoelectronic devices.

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