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Editorial

Optical Antennas

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1. Introduction

This special issue features contributing and invited papers from leading groups in the field of optical antennas. Fourteen papers address some of the most challenging aspects of these photonic components aimed to increase at a nanometer scale and at room temperature the light/matter interaction.

Optical antennas feature the property to strongly enhance light matter interaction at a nanometer scale, at room temperature, and over spectrally broad bandwidths. Interaction between localized light emitters and optical antennas has been deeply investigated in order to (i) enhance the radiative time constants and radiative efficiency of emitters and also (ii) to redirect the electromagnetic radiation. Reciprocally, when illuminated from the far field, optical antennas can focus light into tiny spots which permits to probe the local environment with a high resolution. Consequently, optical antennas have been embraced by many researchers for improved spectroscopy techniques but also to enhance photovoltaic devices, or to realize single-photon nonlinearities in quantum optics.

All these progresses have been permitted on the one hand by an emerging dialogue between physicists and electrical engineers that provides a new understanding of nanooptics, and on the other hand by the recent advances of top-down (FIB, e-beam lithography) and bottom-up nanofabrication technologies. In this context, it is expected that chemical functionalization and self-assembled nanoantennas will play a key role in the near future to realize new nano-antenna designs.

This special issue addresses some of the most exciting challenges of this research field. It contains 14 papers combining experimental and theoretical works from leading groups in the field. These contributions are tentatively classified in four sections surveying the different aspects of the topic: (i) antenna efficiencies, (ii) antenna devices, (iii) coupled antennas and (iv) antenna enhanced spectroscopy.

2. Coupled Antennas

Realizing the full power of plasmonic antennas for any linear or nonlinear application in spectroscopy, light generation or detection naturally requires that we understand coupled plasmonic systems. Firstly, only upon combining single plasmonic scatterers in coupled oligomers do we obtain full control over near-field enhancements, polarization response and far-field directivity in scattering and emission. Secondly, controlling and understanding the coupling of plasmonics to molecular light sources or nanoscale detectors is of the essence to realize the promised enhanced functionalities. The valuable contributions in this section focus on strategies to fabricate coupled systems controllably on the smallest scale and on how to quantify the coupling in optical measurements.

A. Lereu et al. present a study of plasmonic dimer antennas in which high field enhancements are obtained as a consequence of the high field in a narrow gap of a dimer coupled mode. Their study focuses on how to fully quantify the field enhancement in nano-antennas, firstly by linear and nonlinear spectroscopy of the luminescence that especially Au metal antennas generate intrinsically. Secondly they show how a very dilute random sprinkling of single fluorescent molecules on such antennas can be used as ultimate vectorial nonperturbative probes. By collecting statistics on emission polarization, spectra, and lifetime, one can obtain statistical information on antenna performance beyond the diffraction limit and ensemble averaging intrinsic to traditional studies with dense fluorescent films. Finally they show how nanolithography could be used to truly localize single fluorophores.

Some of the most exciting proposals to reach ultimately high field enhancements rely on the fabrication of dimers, trimers and self-similar chains of metal particles of different size and with ultrasmall gaps. At the typical desired dimensions where gaps are just nanometers in size and particles below 40 nm are desired, only bottom-up fabrication strategies can be employed. S. Bidault et al. describe the realization of exactly such a strategy, in which gold particles are assembled in dimers and trimers using a simple one-step water-based scheme. Via appropriate selection of a short dithiol linker molecule, the spacing between particles is reduced to the nanometer scale. The controlled fabrication of dimers and trimers with spacings of just 1.0 to 1.5 nm shows large promise for SERS applications.

S. Suck et al. present experiments and simulations that explore the distance regime in which plasmonic coupling reaches outside the familiar quasistatic limit. For very small coupled antennas well below $\lambda/2\pi$ in size, the characteristic near-field $1/r^3$ coupling is often assumed, without any phase retardation over the object. However, in actual structures this quasistatic assumption usually does not hold. Firstly, phase retardation modifies the coupled eigenmodes, and secondly, quasistatics intrinsically ignores all interaction with the far field. Thus retardation determines scattering. However, exactly where and how the cross-over from quasistatic to retarded behaviour occurs is a matter of debate. S. Suck et al. present measurements of the radiation strength and directionality for the hybridized modes of plasmon disk dimers. Importantly, retardation in their structure with a coupling distance below $\lambda/4$ is directly evident from the fact that the supposedly dark mode is in practice as bright as the nominally bright mode.

Application of plasmonic antennas to detection brings the challenge of coupling optical field enhancements with electrically connected detection elements. H. Chen et al. describe the integration of plasmonic bow tie antennas for large field enhancements with electrically connected carbon nanotubes that act as the actual photodetectors. As the nanotube diameter is comparable to the antenna gap width, these structures form a perfect match. Importantly, H. Chen et al. report an order-of-magnitude photocurrent enhancement as compared to bare carbon nanotube photodetectors at wavelengths in the near infrared. Their structure could thus be a promising candidate for future nanoscale detectors.

3. Antenna Efficiencies

Metallic optical antennas resonantly interact with incident light through the excitation of localized surface plasmon polaritons. This electromagnetic resonance offers moderate quality factors due to the intrinsic metallic losses but permits to confine light into very small volumes. The absorption losses of a single spherical metallic particle can be analytically derived but E. Castanié et al. highlight in their paper the fundamental role played by the environment. They use a rigorous scattering theory based on the Green dyadic function formalism to show that the absorption of light by lossy metallic particles is not an intrinsic property of the metallic particle but depends on its surrounding environment. More precisely, they demonstrate that an increase of the local density of photonic states around the particle permits to decrease the absorption losses. When a particle is excited by incident light, the electron gas oscillates and its relaxation can take place either through the scattering of light (radiative decay rates) or through electron-phonon collisions (non-radiative channels). An increase of the local density of photonic states permits to privilege the relaxation through radiative channels. They illustrate this result by considering a single metallic particle coupled with a perfect mirror. They plot on the same graph the local density of states and the absorption losses of the particle when varying the particle/mirror distance and nicely reveal the link between those parameters.

The efficiency of optical antennas is also questioned by J. Wenger when dealing with fluorescent molecules. The author shows that the fluorescent enhancement factors offered by an optical antenna is not a trivial issue and depends on several parameters such as the antenna geometry and the spectral overlap of its resonance with the emission spectrum of the molecule. Emphasise is placed in this paper on collection efficiency and also emitter's quantum yield. J. Wenger shows how the fluorescence enhancement can be highly modulated by tuning the setup of the experiment but without modifying the design of the plasmonic antenna. He finally gives the ingredients that will offer high fluorescence enhancements but also draws our attention to the fact that high fluorescence enhancement is not synonymous of bright photon sources.

It is now well known that spontaneous emission is not an intrinsic property of quantum emitters and strongly depends on the surrounding environment. G. Colas des Francs et al. derive the Purcell factor of quantum emitters coupled with a single metallic particle. In a first part, they clearly formulate, in the case of an electric dipolar assumption, the quality factor of the plasmon resonance, before deriving the spontaneous emission decay rates when the electric dipolar transition moment is normal to the surface of the particle. Identification with the Purcell factor gives the effective volume of the metallic particle dipolar mode. In a second part, this work is extended and generalized to the case of multipolar modes, and explicit formulations of the quality factors and effective volumes are given as a function of the multipolar order n. The last part of the paper addresses the coupling strength between a quantum emitter and a given mode n compared with all the other deexcitation channels.

4. Antenna Devices

Optical antennas are often considered as essential units of the photonics toolbox for controlling light-matter interaction.

Major research efforts are focused on the development of functionalities at the nanoscale in order to realize optical devices or advanced investigation tools. In their paper, M. Abb et al. report on an all-optical mechanism enabling control over the spectral response of an optical gap antenna. Their approach consists in loading the optical antenna with a nonlinear material. The authors experimentally showed that Indium Tin Oxide (ITO), a common material traditionally used as a thin conductive layer in electron-beam lithography, features a Kerr nonlinearity governed by the free carrier concentration. Optically depleting the material in a pump-probe configuration induces a local change of the refractive index which consequently shifts the antenna resonance. Potential exploitation of this ultrafast antenna switch is discussed.

Another important functionality is approached by M. Klemm. Very much like their radiofrequency counterparts, optical antennas are increasingly used to control the emission diagram of nanoscale light sources. In this contribution, the author describes a novel antenna design with a computed directivity better than state-of-the-art optical Yagi-Uda structures. The concept, based on the travelling wave notion, could enable efficient intrachip wireless optical transmission providing that the emitting and receiving antennas are embedded in the same optical medium.

The role of an optical antenna is essential to interface near-field information to a detectable far-field signal. Scanning near-field optical microscopy is largely taking advantage of antenna characteristics to increase lateral resolution, enhance weak signals, and map optical fields. Because of geometrical considerations, metal tip antennas are usually sensitive to field polarization aligned with the main axis of the tip, limiting thus the detectable information. In their article, Y. Saito et al. demonstrate that, with proper polarization conditions and controlled tip shape, efficient scattering of in-plane optical fields can occur. They demonstrate their approach by mapping the focal point of azimuthal polarized laser beam using different tips and quantified their in-plane efficiencies.

5. Antenna-Enhanced Spectroscopy

Surface modes, in particular those sustained by noble metals, as a result of the excitation of polaritons and plasmons in their surface regions, have played a fundamental role in the so-called surface-enhanced spectroscopies. A myriad of materials and morphologies have emerged in an effort to understand the role of surface excitations and to further exploit the observed enhancement in the applications of molecule-metal substrate systems. The nanoscale curvature and roughness associated with these morphologies yield electromagnetic scattering properties that can elegantly be embraced by the concept of optical nanoantennas. As a receiver, the large number of available modes at the optical frequencies possessed by these antennas facilitates flexible collective electronic excitation. As an emitter, the near- and far-field patterns of the antennas offer a superb platform for nanooptics investigations. Here in an effort to elucidate the enhancement effect of nanoantennas in spectroscopic studies, the authors have treated a series of morphologies with emphasis on their plasmonic properties. Particularly, dipole antennas (A. Ahmed et al.), semicontinuous films (K. Seal et al.), nanocrosses (J. Ye et al.), and nanospheres (V. Le Nader et al.) all support plasmon excitation and as such have been implicated in a form of enhancement process. Typically, either the molecules of interest are brought into the vicinity or contact with the nanostructure making up the antenna, or the antenna is brought near the molecules as in the so-called tip enhancement effect. Consequently, the responses of both nanoantennas and molecules couple to create new scattering properties.

Directing the radiation to the molecules optimally can greatly enhance the signal regardless of what may be provided by surface enhancement due to plasmonic processes. By designing a set of auxiliary structures to serve as reflectors, A. Ahmed et al. fabricated nanoantennas with improved directivity that was quantified employing Raman spectroscopy. Apart from directivity, a desired capability is the flexibility of the spectral placement of the plasmonic resonances of the antenna toward targeting specific vibrational transitions during surface enhanced spectroscopies. To this end J. Ye et al. propose a four-piece gold nanostructure in form of a cross where two of the arms are allowed to vary in length to provide spectral tunability. In doing so, they numerically predict useful levels of enhancement delivered at two resonance positions, a relevant feature in the context of Raman spectroscopy. The utility of nanoantennas in spectroscopy can also be appreciated in structures with less stringent a priori design parameters. The first example herein capitalizes on the controlled deposition of metals below a threshold (typically below 10 nm), which results in the formation of islands. By capturing the near-field of a series of discontinuous silver films with varying surface coverage but random distribution, K. Seal et al. discuss localized as well as delocalized plasmon excitation in describing the measured field distribution, which is of importance in spectroscopic measurements of adsorbates residing on such substrates. In a second example, V. Le Nader et al. describe a chemical immobilization technique for attaching gold nanoparticles from a colloidal solution at the apex of a tapered optical fiber. The sharp optical probe fabricated is subsequently shown to randomly host a number of gold nanoparticles. Owing to their plasmonic properties the particles can resonantly deliver localized energy to a Raman active sample of carbon nanotubes. This tip-enhanced spectroscopy then permits the study of the various Raman bands as a function of the nanoantenna-nanotube distance.

Acknowledgments

The guest editors thank all the authors for their valuable contribution to this special issue. The high quality papers united in this special issue address some of the most important issues in optical antenna theory and design. All the papers are open access which we anticipate will stimulate

the rapid dissemination and wide application of these state-of-the-art findings.

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