Optical Antennas for Enhanced Light-Matter Interactions

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Abstract

Similar to radiowave and microwave antennas, the purpose of *optical antennas* is to convert the energy of free propagating radiation to localized energy, and vice versa. Optical antennas exploit the unique properties of metal nanostructures, which behave as strongly coupled plasmas at optical frequencies. Optical antennas are being explored for increasing the efficiency of light-matter interactions in applications such as light-emitting devices, photovoltaics, and spectroscopy.

1. Introduction

Electromagnetic antennas are a key enabling technology for devices like cellular phones and televisions. They are mostly employed in the radiowave or microwave regime of the electromagnetic spectrum, but their optical implementation is non-existent in today's technology, which is primarily associated with their small scale. In optical science and engineering, light is commonly controlled by re-directing the wave fronts of propagating radiation by means of lenses, mirrors, and diffractive elements. This type of manipulation relies on the wave nature of electromagnetic fields and is therefore not amenable to controlling fields on the subwavelength scale. In contrast, radiowave and microwave technology predominantly makes use of antennas to manipulate electromagnetic fields, controlling them on the subwavelength scale and interfacing efficiently between propagating radiation and localized fields. Recent research in nano-optics and plasmonics has generated considerable interest in the *optical antenna* concept and several studies are currently focused on how to translate established radiowave and microwave antenna theories into the optical frequency regime. The introduction of the antenna concept into the optical frequency regime will provide access to new technological applications. Optical antennas will likely be employed to enhance absorption cross-sections and quantum yields in photovoltaics, to release energy efficiently from nanoscale light-emitting devices, to boost the efficiency of photochemical

or photophysical detectors, and to increase spatial resolution in optical microscopy.

The word *Antenna* most likely derives from the prefix *an-* “up” and the Indo-European root *ten-* “to stretch” [1, 2]. Therefore, from an etymological perspective, an *antenna* is *that which stretches or extends up* [3]. Today, we are accustomed referring to an electromagnetic transmitter or receiver as an antenna, but these were originally called *aerials* in English [4]. In 1983, IEEE defined the antenna as a means for radiating or receiving radio waves (IEEE Std 1451983) [5]. While radio antennas were developed as solutions to a communication problem, the invention of optical antennas was motivated by microscopy. In analogy to its radiowave and microwave counterparts, we define the *optical antenna* as *”A device designed to efficiently convert free-propagating optical radiation to localized energy, and* vice versa*.”* In the context of microscopy, an optical antenna effectively replaces a conventional focusing lens or objective, concentrating external laser radiation to dimensions smaller than the diffraction limit.

In a letter dated 22 April 1928, Edward Hutchinson Synge describes to Albert Einstein a microscopic method in which the field scattered from a tiny particle could be used as a light source [6] (c.f. Fig. 1). The particle would convert free-propagating optical radiation into a localized field that would interact with a sample surface. Thinking of the surface as a receiver, the particle can be viewed as an optical antenna. Synge’s was likely inspired by the development of dark-field microscopy, a technique invented at the turn of the twentieth century by the Austrian chemist Richard Adolf Zsigmondy [7]. In 1988 Ulrich Ch. Fischer and Dieter W. Pohl carried out an experiment similar to Synge’s proposal [8]. Instead of a solid metal particle, they used a gold-coated polystyrene particle as a local light source. Fischer and Pohl imaged a thin metal film with 320nm holes and demonstrated a spatial resolution of ~50nm. Later, laser-irradiated metal tips were proposed as optical antenna probes for near-field microscopy and optical trapping [9, 10] and since then various antenna geometries have been studied (rods, bowties, etc.).

Optical antennas are strongly analogous to their RF and microwave counterparts, but there are crucial differences in their physical properties and scaling behavior. Most of these differences arise because metals are not perfect conductors at optical frequencies, but are instead strongly correlated plasmas described as a free electron gas. Optical

antennas are also not typically driven with galvanic transmission lines – localized oscillators are instead brought close to the feed point of the antennas, and electronic oscillations are driven capacitively [11]. Moreover, optical antennas can take various unusual forms (tips, nanoparticles, etc.) and their properties may be strongly shape and material dependent due to surface plasmon resonances.

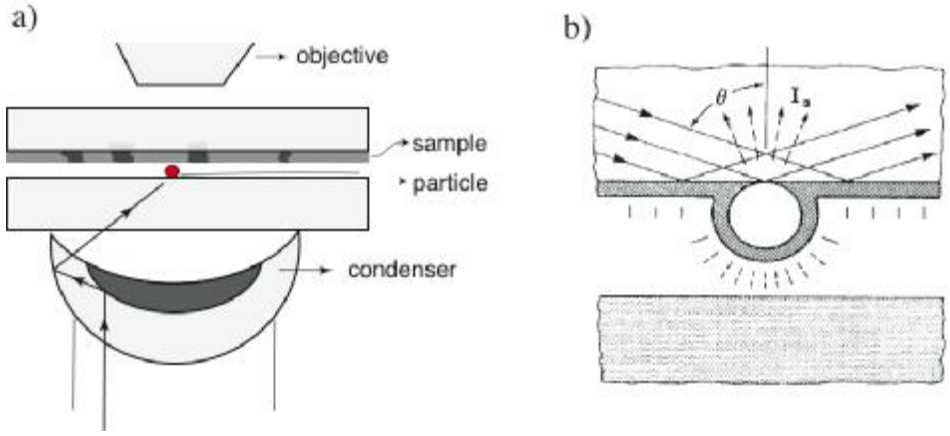

Figure 1. (a) Synge's original proposal of near-field optical microscopy based on using the scattered light from a particle as a light source. Adapted from Synge's letter dated 22 April 1928 sent to Einstein [6]. (b) 1988 experiment by Fischer and Pohl. The near-field probe consists of a gold-coated polystyrene particle [8].

Typically, an optical antenna interacts with a receiver or transmitter in the form of a discrete quantum system, such as an atom, molecule, or ion. The antenna enhances the interaction between the receiver or transmitter and the radiation field. It therefore provides the prospect of controlling the light-matter interaction on the level of a single quantum system. The presence of the antenna modifies the properties of the quantum system, such as its transition rates and, in the case of a strong interaction, even the energy-level structure. Likewise, the antenna properties depend on those of the receiver/transmitter, and it becomes evident that the two must be regarded as a coupled system. The efficiency of the interaction can be expressed in terms of established antenna terminology, such as antenna gain, efficiency, impedance, directivity, and aperture [12].

2. Radiation Enhancement with Nanoparticle Antennas

A spherical nanoparticle is probably the simplest model antenna [13-16]. This antenna geometry is not very efficient, but it allows for a quantitative comparison with simple analytical solutions [16]. As shown in the inset of Fig. 2a, we consider a transmitter in the form of a single fluorescent molecule, optically pumped by external laser radiation. For weak excitation intensities the radiation rate Grad can be expressed as

$$\Gamma_{rad} = \Gamma_{exc}\, \eta_{rad} \qquad (1)$$

where Γ_{exc} is the excitation rate and η_{rad} is the quantum yield. Both Γ_{exc} and η_{rad} depend on the antenna properties and the separation z between antenna and molecule. η_{rad} corresponds to the radiation efficiency and the rates Γ_i can be expressed in terms of powers as $P_i = \Gamma_i\, h\nu_i$ with $h\nu_i$ corresponding to the atomic transition energy.

Fig. 2a shows the experimentally recorded photon emission rate of a single dye molecule as a function of its separation from a 80nm silver nanoparticle. The superimposed red curve is a theoretical calculation according to a simple electromagnetic model, which treats the molecule as a classical oscillating dipole [16]. The data demonstrates that as the silver particle is brought closer to the molecule the fluorescence emission rate first increases and then gets suppressed. The initial fluorescence enhancement is due to the antenna effect of the silver particle. The excitation rate Γ_{exc} increases because of the enhanced local fields near the nanoparticle. However, for separations shorter than z = 10nm, the radiation efficiency hrad starts to decrease rapidly because more and more of the energy is being absorbed in the silver nanoparticle. At a distance of z ~ 3nm the fast decrease of η_{rad} wins over the increase of Γ_{exc} and the fluorescence of the molecule gets quenched. Hence, there is an optimum separation between molecule and antenna.

Fig. 2b shows a near-field fluorescence image of single dye molecules dispersed on a flat glass surface. The fluorescence emission rate has been recorded pixel-by-pixel while the dye sample was raster scanned underneath a laser-irradiated nanoparticle antenna. The latter was held fixed at a distance of z ~ 5nm above the sample surface by means of a shear-force feedback mechanism [17, 14, 18]. The resolution achieved in this type of near-field imaging is determined by the antenna size. With a 80nm silver or gold

particle we typically achieve resolutions of ~ 65nm. Notice the different fluorescence patterns in Fig. 2b. The different patterns are due to different orientations of the molecular transition dipole axis [19, 20].

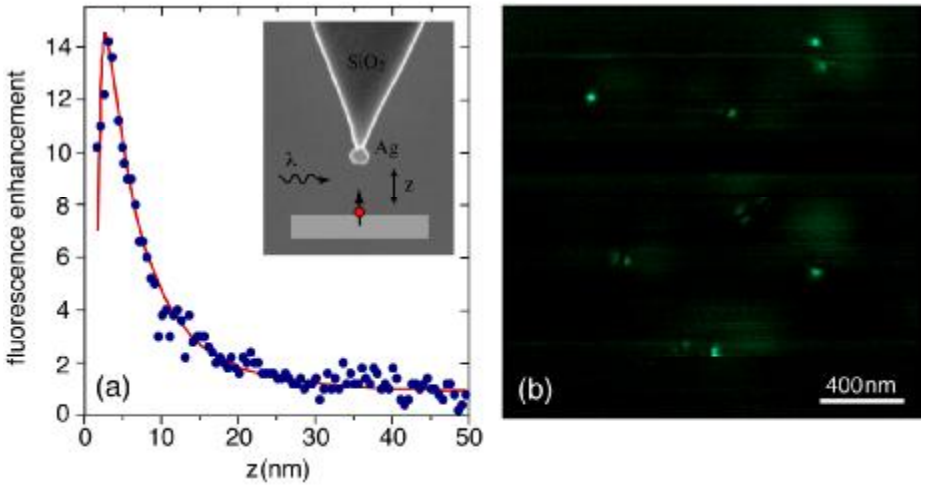

Figure 2. Enhancement of the radiation rate of a single molecule with a silver nanoparticle antenna. (a) Normalized fluorescence rate as a function of antenna-molecule separation. Dots are data and the curve is the result of a theoretical calculation. Inset: SEM image of a nanoparticle antenna. The particle is held by a dielectric tip. λ=488nm. (b) Fluorescence rate image recorded by raster scanning a sample with dispersed dye molecules in a plane $z \approx 5$nm underneath a nanoparticle antenna. The different fluorescence patterns are due to different orientations of the molecular transition dipole axis.

Similar experiments have been performed with other quantum systems, such as quantum dots or carbon nanotubes, and the findings are consistent with the results discussed here for single fluorescent molecules. An important finding is that for systems with weak intrinsic quantum efficiency (η_i) the radiation efficiency can be enhanced by the optical antenna. In the example discussed here we assumed $\eta_i = 1$ and hence the antenna can only *decrease* the radiation efficiency. However, for poor emitters, such as carbon nanotubes, the antenna can increase the radiation efficiency by more than a factor of 10 [21]. In general, the lower η_i the more the antenna increases the overall efficiency, an effect that was observed already in 1983 by Wokaun *et al.* [22]. The possibility of increasing the quantum efficiency of weak emitters holds promise for

boosting the efficiency of organic light emitting devices (OLEDs), silicon based lighting, and solid state lighting (SSL) in the yellow and green spectral region [23, 24].

The nanoparticle antenna serves as a model antenna and its predictions have been tested in various recent experiments. However, much higher efficiencies can be achieved with optimized antenna designs, such as the optical half-wave antenna.

3. Near-field Raman Scattering

The hallmark of optical antennas, their ability to influence light on the nanometer scale, leads naturally to nanoimaging applications. In the context of nanoscale imaging, an optical antenna represents a near-field optical probe used to locally interact with an unknown sample surface. To acquire a near-field optical image, the optical antenna is guided over the sample surface in close proximity and an optical response (scattering, fluorescence, antenna detuning, ..) is detected for each image pixel.

The vibrational spectra provided by Raman scattering define a unique chemical fingerprint for the material under study. Raman scattering involves the absorption and emission of photons almost identical in energy, and a nearby antenna can amplify *both* the incoming field and the outgoing field. The total Raman scattering enhancement is therefore proportional to the *fourth* power of the field enhancement [25].

In tip-enhanced Raman scattering (TERS) optical antennas such as metal tips are employed for point-by-point Raman spectroscopy [27, 26, 28], similar to the original idea of Wessel [29]. Raman enhancements achieved with tips are typically in the range of 10^4 – 10^8, corresponding to field enhancements of 10 - 100. Our TERS studies are focused on localized states in carbon nanotubes. These localized states are due to defects and dopants and can be studied with resolutions of 10 - 20nm by near-field Raman scattering [30, 31]. Fig. 3 shows the simultaneously recorded topography (a) and near-field Raman image (b) of a single-walled carbon nanotube sample. The image contrast in the near-field Raman image is defined by the intensity of the G' line (vibrational freuency of $\nu = 2615\ \mathrm{cm}^{-1}$) highlighted in the spectrum (c).

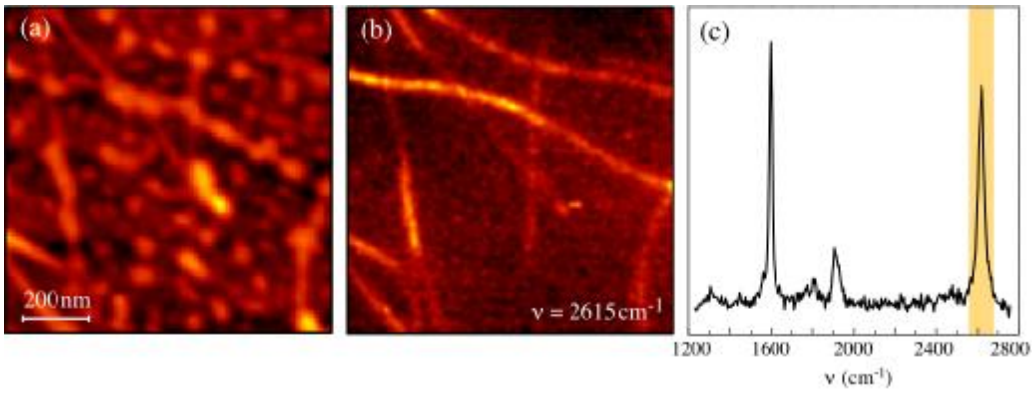

Figure 3: Near-field Raman imaging of a single-walled carbon nanotube sample. (a) Topography showing a network of carbon nanotubes covered with small droplets. (b) Raman image of the same sample area recorded by integrating, for each image pixel, the photon counts that fall into a narrow spectral bandwidth centered around n *= 2615* cm^{-1} *(indicated by the yellow stripe in c). (c) Raman scattering spectrum recorded on top of the nanotube. From [26].*

4. Wavelength Scaling

At optical frequencies electrons in metals have considerable inertia and cannot respond instantaneously to the driving fields. Typically, the skin depth is in the order of tens of nanometers, comparable to the dimensions of the antenna. Traditional design rules that prescribe antenna parameters only in terms of an external wavelength are thus no longer valid. Rigourously treating the metal as a strongly coupled plasma is required, which leads to a reduced effective wavelength seen by the antenna [32]. This effective wavelength λ_{eff} is related

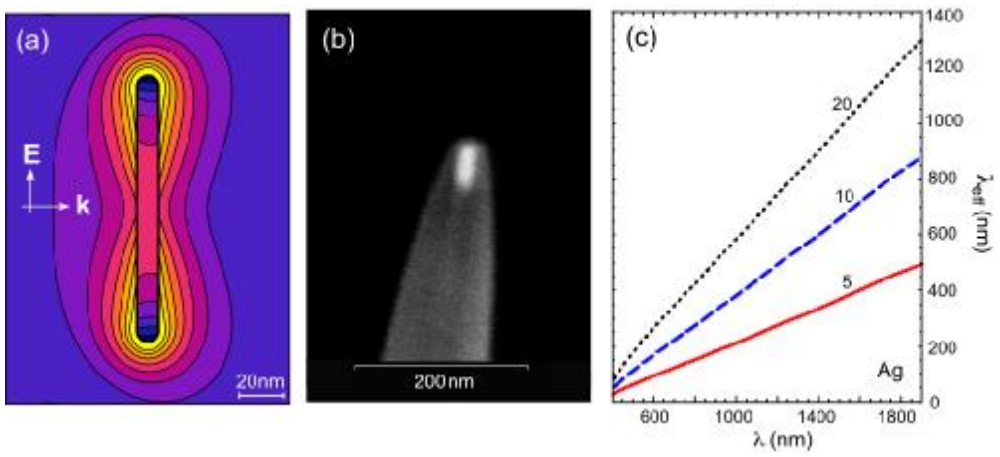

Figure 4: Effective wavelength scaling for linear optical antennas. (a) Intensity distribution (E^2, factor of 2 between contour lines) for a gold half-wave antenna irradiated with a plane wave (l = 1150nm). (b) SEM image of a half-wave antenna resonant at l = 650nm, fabricated by placing a gold nanorod of length ~65nm into the opening of a quartz nanopipette. (c) Effective wavelength scaling for silver rods of different radii (5, 10, and 20nm).

to the external (incident) wavelength λ by the surprisingly simple relation

$$\lambda_{eff} = n_1 + n_2 [\lambda / \lambda_p] \qquad (2)$$

where λ_p is the plasma wavelength of the metal and n_1 and n_2 are constants that depend on the geometry and dielectric parameters of the antenna. leff is roughly a factor of 2 to 6 shorter than the free space λ for typical metals (Au, Ag, Al) and realistic antenna thicknesses [32, 33].

The wavelength shortening from λ to λ_{eff} has interesting implications. For example, it implies that the radiation resistance of an optical half-wave antenna is of the order of just a few Ohms [34, 32, 35]. To see this, we note that the radiation resistance of a thin-wire antenna is roughly $R_{rad} = 30\ \pi^2 (L/\lambda)^2$, with L being the antenna length. For a half-wave antenna at RF frequencies $L = \lambda/2$ and $R_{rad} \sim 73\Omega$. However, for an optical half-wave antenna, $L = \lambda_{eff}/2$ and hence $R_{rad} = (30/4)\ \pi^2 (\lambda_{eff}/\lambda)^2$. In other words, the radiation resistance at optical frequencies is a factor of $(\lambda_{eff}/\lambda)^2$ smaller than at RF frequencies. For $\lambda_{eff} = \lambda/5$ we find $R_{rad} = 3\ \Omega$. Fig. 4a shows the intensity distribution near

a gold half-wave antenna of length L = 110nm and radius R = 5nm resonantly excited at λ = 1170nm. The effective wavelength is λ_{eff} = 220nm. The induced current density $\mathbf{j} = i\omega\varepsilon_o[\varepsilon(\omega) - 1]\mathbf{E}$ evaluated along the axis of the antenna is found to be nearly 180 ^{o} out of phase with respect to the exciting field.

The notion of an effective wavelength can be used to extend familiar design ideas and rules into the optical frequency regime. For example, the optical analog of the $\lambda/2$ dipole antenna becomes a thin metal rod of length $\lambda_{eff}/2$. Since λ_{eff} for a silver rod of radius 5nm is roughly $\lambda/5.2$ (c.f. Fig. 4c), this means that the length of a "$\lambda/2$" dipole antenna is surprisingly small, about $\lambda/10.4$. One can similarly construct antenna arrays like the well-established Yagi-Uda antenna developed in the 1920s for the UHF/VHF region [32, 36].

5. Enhanced Light-Matter Interactions

The localized fields near an optical antenna open up new interaction mechanisms between light and matter, such as higher-order multipole transitions or momentum-forbidden transitions. These interactions are inaccessible in free space and have the potential to enrich optical spectroscopy, and provide new strategies for optical sensing and detection. In free space, the momentum of a photon with energy E is $p = E/c$. On the other hand, the momentum of an unbound electron with the same energy is two to three orders of magnitude larger than the photon momentum and the photon momentum can be neglected in electronic transitions. However, near optical antennas the photon momentum is no longer defined by its free space value. Instead, the localized optical fields are associated with a photon momentum that is defined by the spatial confinement D, which can be as small as 1-10nm. Thus, in the optical near field the photon momentum can be drastically increased and comparable with the electron momentum, especially in materials with small effective mass m^*. Hence, localized optical fields can give rise to "diagonal" transitions in an electronic band diagram thereby increasing the overall absorption strength, which can be useful for devices such as silicon solar cells. The increase of photon momentum in optical near-fields has been discussed in the context of photoelectron emission [37] and photoluminescence [38].

The strong field confinement near optical antennas also has implications on

selection rules in atomic or molecular systems. Usually, the light-matter interaction is treated in the dipole approximation where the spatial variation of the fields is much weaker than the spatial variation of the quantum states (wavefunctions). However, the localized fields near optical antennas give rise to spatial field variations of a few nanometers and hence it may no longer be legitimate to invoke the dipole approximation. This is especially the case in semiconductor nanostructures where the low effective mass gives rise to quantum orbitals with large spatial extent.

6. Conclusions and Outlook

Research in the field of optical antennas is currectly driven by the need for high field enhancement, strong field localization, and large absorption cross sections. This includes antennas for high-resolution microscopy and spectroscopy, photovoltaics, light emission, and coherent control. In one way or another, optical antennas are used to make processes more efficient or to increase the specificity of gathered information.

As in canonical antenna theory, there is no universal antenna design. Instead, optical antennas have to be optimized separately for each application. However, to achieve the best efficiency the internal energy dissipation of any antenna must be minimized. For a quantum emitter such as an atom, molecule, or ion, a good antenna yields a low nonradiative decay rate.

New ideas and developments are emerging at a rapid pace and it is clear that the optical antenna concept will provide new opportunities for optoelectronic architectures and devices. Today, the building blocks for optical antennas are plasmonic nanostructures. These can be fabricated from the bottom-up by colloidal chemistry or top-down with established nanofabrication techniques, such as electron beam lithography or focused ion-beam milling. It is also conceivable that future optical antenna designs will draw inspiration from biological systems such as light harvesting proteins in photosynthesis.

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