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Part I

FUNDAMENTALS

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1 From near-field optics to optical antennas

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1.1 The near-field

The birthplace of a photon is an excited and decaying atom or molecule. It behaves like a point-like oscillating dipole or an EHD, in radio-engineering language. The photon has to travel a distance of at least $\lambda/2\pi$ to become a real photon with well-defined momentum and energy. Before, the electric ($\vec{E}$) and magnetic ($\vec{H}$) fields of the corresponding wave undergo severe changes. This is obvious from the well-known equations for an EHD with complex amplitude $p = p_0 \exp(\mathrm{i}\omega t)$, where p_0 denotes the dipole strength. In spherical coordinates (r, θ, ϕ) with dipole orientation along $\phi = 0$, the non-zero components of fields write [1]

$$E_r = 2p_0 \left(\frac{1}{r^3} + \frac{\mathrm{i}k}{r^2} \right) \cos\theta, \tag{1.1}$$

$$E_\theta = p_0 \left(\frac{1}{r^3} + \frac{\mathrm{i}k}{r^2} - \frac{k^2}{r} \right) \sin\theta, \tag{1.2}$$

$$H_\phi = p_0 \left(\frac{\mathrm{i}k}{r^2} - \frac{k^2}{r} \right), \tag{1.3}$$

and $k = \omega/c$. The first terms in Eqs. (1.1) and (1.2) go with r^{-3}. They are the dominant terms for $r \to 0$ and typical for a quasi-static dipole. For large r, only the r^{-1} terms are left over, producing the well-known far-field wave. The two extremes of the dipolar wave are almost orthogonal to each other. The terms with r^{-2} are significant in the transition zone between near- and far-field. The borderline between near- and far-field can be drawn at

$$r_\mathrm{B} = \frac{1}{k} = \frac{\lambda}{2\pi}, \tag{1.4}$$

where the energies of the different contributions are balanced.

For visible radiation, r_B amounts to roughly 100 nm – a distance which can accommodate up to $\sim$ 300 atoms in condensed matter, all of them potentially waiting to swallow (absorb) or modify the "proto-photon." Indeed, quite a few things happen on this scale in nature and, more and more, also in technology. Near-field *microscopy* looks at this region in detail. The wealth of light/matter interactions allows for a variety of space-resolved spectroscopies. They provide information not only on the elemental composition of a sample, but also on its chemical organization and structure. Furthermore, optical techniques are unsurpassed with respect to power and dynamic response. However, *near-field optics* is a science with more (potential) than mere microscopy. Optical antennas have enlarged its field of research and applications, as will be emphasized in Sec. 1.7.

1.2 Energies and photons

For $r < r_\mathrm{B}$, an EHD creates the electrical and magnetic energy density

$$\mathcal{E}_\mathrm{E} \sim \epsilon|\vec{E}_\mathrm{B}|^2 \sim r^{-6}, \tag{1.5}$$

$$\mathcal{E}_\mathrm{M} \sim \mu|\vec{H}_\mathrm{B}|^2 \sim r^{-4}, \tag{1.6}$$

where $\vec{E}_\mathrm{B}$ and $\vec{H}_\mathrm{B}$ are respectively the electric and magnetic fields at r_B. The magnetic energy is negligible here because of the power laws (see Eqs. (1.5) and (1.6)). Note that this would be different for a magnetic dipole source. The r^{-6} dependence of Eq. (1.5) also indicates that the energy in the near-field is large compared to that of the far-field, which is proportional to r^{-2}. The excess energy does not radiate into the far-field but goes back and forth around the origin, resulting in a Poynting vector

$$\vec{S} \sim \vec{E}_\mathrm{B} \times \vec{H}_\mathrm{B} \sim r^{-5}, \tag{1.7}$$

which is to be compared to the r^{-2} dependence of the far-field. It is not an emission ($\langle S_r \rangle = 0$), but it goes back and forth around the EHD, giving the near-field an evanescent character. Quantum mechanically, it corresponds to a swarm of photons emitted and immediately reabsorbed by the atom or molecule. They are often called "virtual" photons which disappear when the "real" photon has left the near-field zone.

These "virtual" photons are absolutely real in the near-field. An example is the Förster effect [2]: a solution of two certain types of fluorescent molecules, say a green and a red fluorescing species, is excited by light of a frequency absorbed only by the "green" molecules. If the solution is dilute with respect to "red" molecules, only green fluorescence is seen; if, however, the concentration of "red" molecules is so large that they penetrate the near-field zone of the "green" ones, transfer of green fluorescent energy to the "red" molecules is dominant, observed by fluorescence of the latter.

Evanescence is obvious also from the field behind a sub-microscopic aperture, which is limited to its diameter $a \ll \lambda/2\pi$. The respective Fourier components are centered on $k_x = 2\pi/a \gg k$. The wave equation hence demands imaginary values of the respective longitudinal components of the wave vector. Imaginary values, however, mean a damped field instead of a propagating one [3].

Generally speaking, interactions of "virtual" photons or "evanescent" waves are at the center of near-field optics. They gain increasing scientific and even industrial attention in a world which is concerned with nanometer entities.

1.3 Foundations of near-field optical microscopy

Intuitively, the most obvious configuration of a near-field optical microscope is a screen with aperture as essential element. It had already been proposed in 1928 [4], but later forgotten. There is a second configuration though, complementary to the aperture in some aspects, which produces similarly confined electromagnetic fields. It is the NP, known from Mie scattering. When illuminated, it produces an EHD field to first order that is influenced by its environment. The scattered radiation is inherently more intense than that of a same-sized aperture.

The strong attenuation by sub-microscopic apertures can be imagined as a short-circuit phenomenon: the potential of the wall around the aperture is almost constant because of its small dimensions. The field representing a light wave consequently has to go to zero near the rim of the aperture. The corresponding voltage between center and rim is small, resembling that of a magnetic dipole. The advantage of the scattering NP is compensated, however, by a strong background from the primary radiation.

The sub-microscopic aperture or NP with diameter a ($a \ll \lambda/2\pi$) has to fulfill three requirements:

- Finite transmission or scattering
- Confinement of light at the exit side to an area $\sim a^2$
- Scanning in near-field proximity with the object.

The development of an aperture *at the apex of a transparent metal-shielded tip* met these requirements and enabled sub-microscopic imaging for the first time [5–13]. The scattering NP as near-field probe came up a little later; it allows for a variety of experimental configurations (see, for instance, Refs. [14–16]).

There are a number of developments relating to near-field optical microscopy. In the 1940s Bethe and Bouwkamp calculated the transmission in the context of the electromagnetic showers following an atomic bomb explosion [17, 18]. It was much lower than expected from the geometrical size. Later, Lewis *et al.* observed experimentally the transmission of light through 50 nm diameter apertures and pointed at their potential use for super high-resolution microscopy [19].

Fischer and Zingsheim did contact imaging of small metal disks and latex beads [20], and observed enhanced fluorescence next to sub-microscopic apertures [21]. Scattering NPs were at the beginning of Plasmonics, another important part of near-field optics research (see, for instance, Ref. [22]). It should be noted that holes can also cause plasmonic phenomena [23], which may blur the output spot and decrease resolution.

1.4 Scanning near-field optical microscopy

The essential part of a SNOM is the narrow aperture or the NP. These scattering centers have to be located at the apex of a tip to allow scanning over samples with

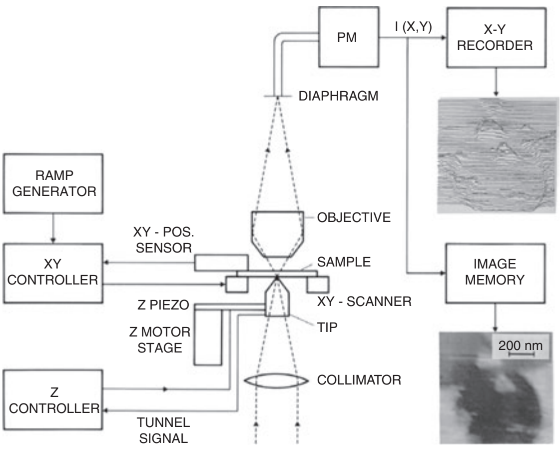

Figure 1.1 The "optical stethoscope," later called a SNOM. Schematic setup, record of an opaque metal film with small holes. (Reproduced with permission from Ref. [6]. Copyright (1986). American Institute of Physics.)

arbitrary shape. A small distance to the object is maintained by tunneling [5–7] or interfacial forces [11]. Light, usually from a laser, is focused onto the aperture or scattering center. The object optical properties in its near-field modulate the transmitted or scattered radiation. Both amplitude and phase variations have an influence. The interpretation of SNOM images is therefore often more complex than that of classical micrographs, but it is also richer in information.

The first SNOM [5] emitted light through a narrow aperture at the apex of a transparent quartz tip (see Fig. 1.1). It was called an "optical stethoscope" to make plausible the subwavelength imaging. It had turned out that "resolution beyond the diffraction limit" was too strange an idea for some conventional microscopists to even think about. In medicine, a stethoscope transmits the beating of the heart to the doctor's ear. Its strength allows determination of its position to a few centimeters' accuracy, although its low frequency implies meters of wavelength.

The optical stethoscope was a prototype delivering only line scans, but they already showed high resolution. With the next version, two-dimensional images of holes were demonstrated with about 30 nm resolution [6, 7].

Different aspects of near-field optical microscopy were investigated in the years following, e.g. aperture in collection [8], reflection [9], and interferometric [13] modes, distance dependence [12], scattering NP with plasmonic effects [14], use of metallic scattering tips [15], or the combination of aperture and tip [16]. Betzig *et al.* in particular demonstrated the considerable potential of SNOM [10–12] (see Fig. 1.2). Their setup with a fiber optical tip and shear force regulation has become a standard.

The experimental efforts stimulated the theory of near-fields and numerical simulations. This resulted in various new phenomena in near-field optics, such as the discovery of increased forces [25] and heat transfer [26] between two bodies at close distance, or the role of LSPRs [27].

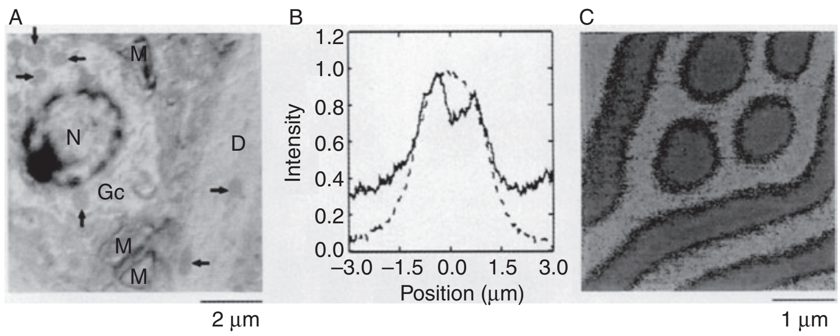

Figure 1.2 Applications of SNOM, each panel with a different contrast mechanism. (a) Absorption within section from the monkey hippocampus. D, pyramidal cell dendrite; Gc, glia cell; M, myelinated axon; N, nucleus; arrows point at mitochondria. (b) Simultaneous refractive index (solid line) and luminescence detection (dashed line) of core and erbium doping distribution within an optical fiber [24]. (c) Polarization imaging of magnetic domains within a bismuth-doped, yttrium–iron–garnet film. (Reproduced with permission from Ref. [12]. Copyright (1992). American Association for the Advancement of Science.)

1.5 Problems of near-field optical microscopy

The near-field optical microscopy developed up to now essentially suffers from two drawbacks: low efficiency of transmission (aperture) or small contrast (scattering NP) on the one hand, and vulnerability of the aperture or scattering NP on the other.

The first drawback of aperture SNOM has three origins, two of these being fundamental:

- The principally low transmissivity of a small aperture (see above).
- The metal walls around the aperture must be several times the penetration depth in thickness to be opaque. Hence, the aperture is the exit of a tiny over-damped waveguide.
- Backward reflections inside the tip

This generates an enormous attenuation for the laser radiation resulting in low data acquisition rates. An inverse situation exists when a scattering object, for instance a sharp tip, is used. A light beam can easily be focused on such a structure, resulting in strong scattering. However, the light beam also hits the object causing considerable background. As a result, the signal/noise level is roughly the same for both implementations.

Vulnerability is caused by the scanning process. It is difficult to avoid eventual encounters with the object surface. The respective forces can be kept small by regulation, but the tiny size of the tip still results in destructive pressures. These are responsible for wear of the scattering center, e.g. widening of the aperture or rupture of the tip apex. These shortcomings are a challenge for future technically gifted microscopists!

1.6 From near-field optical microscopy to optical antennas

The idea of the "optical antenna" has been around for a long time [28–32], but its relevance for the optical near-field was only recognized in recent years. It promises a much better concentration of electromagnetic radiation than the submicroscopic aperture. There are two reasons for this:

- Efficient antennas allow for finite voltage differences in a narrow gap ($\ll \lambda$) between its two arms, in contrast to an aperture of the same dimensions.
- The antenna can be made resonant by adjusting its dimensions.

Scaling from RF to optical frequencies may not give the correct antenna length, since metals can no longer be treated as ideal conductors: their electrons cannot follow the rapid oscillations of the optical field. As a consequence, plasmonic resonances interfere with geometric ones.

One expects a resonant antenna to collect radiation energy within an area of roughly $(\lambda/2)^2$ and concentrate it in the gap. Hence, the energy density inside the gap is increased by a factor

$$f_{\text{enh}} = g_{\text{ant}} \frac{\lambda^2}{4wd}. \tag{1.8}$$

Here w and d are the dimensions of the gap in the plane of the antenna. g_{ant} is a correction factor specific to the antenna shape and material. These influence the loss within the antenna and the resonances. It is an interesting question as to which values of w and d Eq. (1.8) holds – would a single molecule between the arms of the antenna be sufficient to represent a "gap"? What about field enhancement in the tunnel regime? Some of these issues have been recently addressed by means of first-principles studies of NP dimers and of their interaction with a single molecule (see Chapters 5 and 11).

1.7 Optical antennas

The optical antenna with dimensions in the order of $\lambda/2$ has become a realistic option through the increased sub-micrometer structuring capability of the last decade. Three main questions had to be addressed:

- Where are the resonances of an optical, i.e. plasmonic antenna?
- How strong is the field enhancement at resonance?
- How localized is the field enhancement?

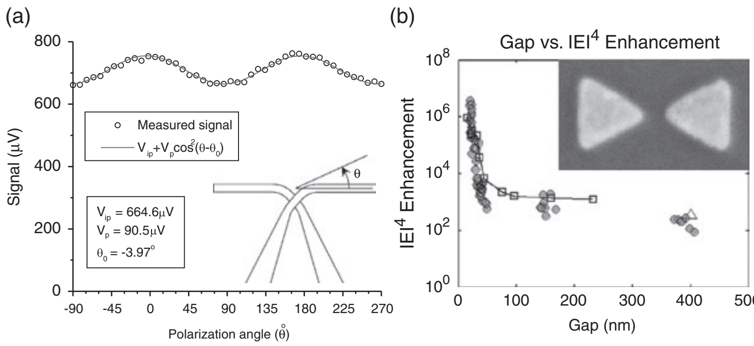

Figure 1.3 (a) Detection of visible light with an IR antenna. (b) $|\vec{E}|^4$ enhancement versus gap width. ((a) reproduced with permission from Ref. [30]. Copyright (1999). Optical Society of America. (b) reproduced with permission from Ref. [33]. Copyright (2005). American Physical Society.)

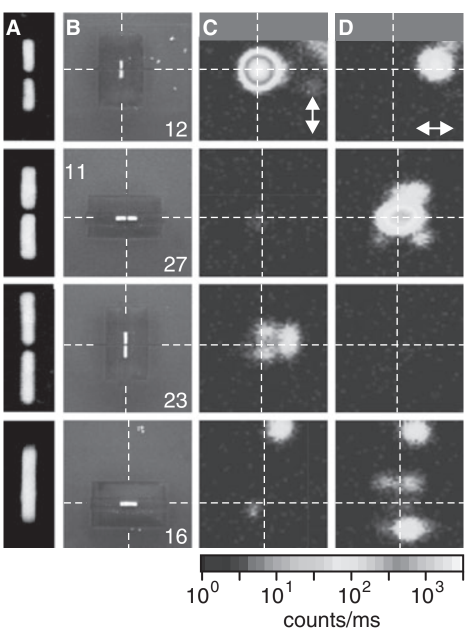

Figure 1.4 Optical antennas. (a,b) SEM pictures of example resonant (12,27) and off-resonance (23) dipoles and a stripe without gap (16). (c,d) Confocal scan images, 830 nm illumination, observation in the range 450–750 nm. (Reproduced with permission from Ref. [34]. Copyright (2005). American Association for the Advancement of Science.)

Several groups, e.g. at Stanford University [29] and at the University of Central Florida [30], had studied antennas for the IR before. Even visible light could be detected with such a long IR antenna (see Fig. 1.3a). The first experiments with "optical antennas," i.e. antennas with dimensions in the order of $\lambda/2$ were published in 2005 [33–35]. TPL demonstrated the strong gap width dependence of the enhancement by Au dipole/bowtie antennas (see Fig. 1.3b) [33].

The analogy of optical antennas with RF antennas was first demonstrated by dipole antennas designed to be resonant at the wavelength of light [34]. They were prepared by EBL and FIB (see Figs. 1.4a–b). They differed in length,

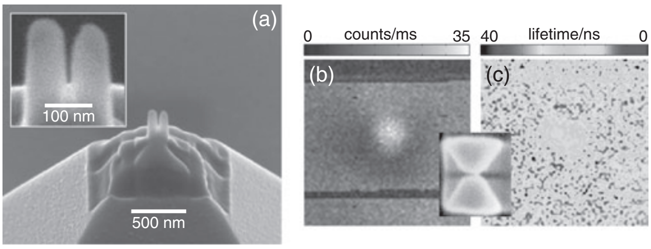

Figure 1.5 (a) Optical antenna at the apex of an AFM tip; (b) Antenna to scale, count rate; (c) exciton lifetime. (Reproduced with permission from Ref. [35]. Copyright (2005). American Physical Society.)

orientation and the existence of a gap. Picosecond laser pulses were focused onto the antennas for excitation. It was assumed that the radiation would cause a large electromagnetic field at the laser frequency in the antenna gap, similar to the situation in a RF receiver antenna. This, in turn, would give rise to nonlinear effects within the gap material. The latter would cause radiation at frequencies larger than the original one. All this proved to be the case, but only if the Au NPs representing the antennas had a gap, were oriented in the field direction and had a certain length (see Figs. 1.4c–d). A resonance was found at an antenna length clearly shorter than $\lambda/2$.

The gap as the place of strong enhancement was confirmed in a second experiment [35]. An approximately resonant bowtie antenna, prepared on an AFM tip (see Fig. 1.5a) was scanned over an illuminated, excitonically fluorescent nanocrystal. The fluorescence was maximal at centered position, i.e. when the gap of the nanoantenna is over the nanocrystal. The excited state lifetime, however, was a minimum at this position.

The results of the two experiments not only prove the gap to be the primary enhancement center, but also demonstrate the electromagnetic coupling of nanoantenna with the fluorescence of the nanocrystal and opportunities in nonlinear optics with antennas.

1.8 Conclusions and outlook

Optics traditionally has been considered a field of physics although a light wave is described by the same Maxwell equations as at RF. Traditional optics, however, was more concerned with phenomena like fluorescence, Raman effect or picosecond pulse generation than with impedance of optical waveguides or antenna matching. This has changed with the coming of near-field optics. Many phenomena may be usefully described either in the language of electrical engineering or of optics. The ambivalence may create new insight and novel applications.

The experiments described above have shown that optical antennas are feasible and offer interesting properties. Their further research may possibly open novel applications in fields like (tele-) communication, (quantum-) computing, or (solar-) energy conversion.