The History of Near-field $Optics^{\perp}$

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Abstract

This article provides a review of early work and developments in the field of near-field optics. The roots trace back to the letters exchanged between Edward Hutchinson Synge and Albert Einstein in 1928 and, because of the analogy to antenna theory and lightning rods, the origins project back to the time of Benjamin Franklin who discovered *the* wonderful Effect of Points both in drawing off and throwing off the Electrical Fire. The modern interest was mainly inspired by the invention of scanning probe microscopy and by the first optical near-field measurements by Dieter W. Pohl and co-workers at the IBM Research Laboratory in Rüschlikon, Switzerland, and also by parallel developments of other groups. Near-field optics received inspiration from the fields of surface enhanced spectroscopy and from studies of energy transfer. While optical near-fields were extensively exploited for overcoming the diffraction limit in optical imaging the study of their physical aspects revealed unique properties which cannot be imitated by free propagating radiation.

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1 Introduction

Near-field Optics is the study of non-propagating inhomogeneous fields and their interaction with matter. Optical near-fields are localized to the source region of optical radiation or to the surfaces of materials interacting with free radiation (secondary sources). In many situations, optical near-fields are explored for their ability to localize optical energy to length scales smaller than the diffraction limit of roughly $\lambda/2$, with λ being the wavelength of light. This localization is being explored for ultrasensitive detection (Fischer, 1985, 1986; Levene, 2003) and for high-resolution optical microscopy and spectroscopy (Novotny and Stranick, 2006, e.g.). Optical near-fields can have physical properties that are drastically different from their free-propagating counterparts. Examples are spatial and temporal coherence (Carminati and Greffet, 1999; Apostol and Dogariu, 2003; Roychowdhury and Wolf, 2003), the polarization state (Setälä, 2002; Ellis, 2005), thermal energy density (Shchegrov, 2000), and the very nature of the light-matter interaction (Carniglia and Mandel, 1971; Agarwal, 1975; Keller, 2002; Zurita-Sanchez, 2004; Henkel, 2005).

An angular spectrum representation of electromagnetic fields yields a decomposition into homogeneous plane waves and inhomogeneous evanescent waves. This decomposition depends on the particular choice of a reference axis (optical axis). The near-field region is generally characterized by the region in space where the evanescent waves cannot be neglected. The most elementary near-field is the one associated with a single evanescent wave as generated, for example, by total internal reflection at the surface of a dielectric material. Mathematically, the evanescent wave is a solution of the Helmholtz equation in free-space. However, because of its exponential distance dependence the evanescent wave would yield an infinite energy at a distance far away from its mathematical origin. Therefore, on physical grounds, the evanescent wave cannot exist in free-space and is restricted to material boundaries making it impossible to decouple the evanescent wave from its source. Consequently, an evanescent wave cannot exist in the absence of other waves in space. This property makes a theoretical understanding of light-matter interactions in the optical near-field challenging. For example, evanescent waves cannot be quantized because they do not form an orthonormal set of solutions. Only when the evanescent waves are complemented by other solutions (e.g. exciting and reflected planes waves) a quantization can be accomplished (Carniglia and Mandel, 1971). Unlike free propagating radiation, evanescent waves are not purely transverse ($\nabla \cdot \mathbf{E}$ is not zero everywhere) and hence longitudinal fields enter the light-matter interaction in the near-field. For example, already in 1891 Paul Drude and Walther Nernst investigated experimentally the excitation of fluorescence by a standing evanescent wave (Drude and Nernst, 1891). The study confirmed that maxima of the field coincide with the maxima of the fluorescence yield, i.e. with the light-matter interaction strength. While today this study might seem trivial other fundamental aspects of optical near-fields still pose a great theoretical challenge. For example, because an evanescent field is coupled to its source the light-matter interaction in the near-field influences the very nature of the source. When two atoms A and B interact over a short distance their physical properties become coupled and causality makes it impossible to define either of them as the source of the interaction (Power and Thirunamachandran, 1997; Keller, 2002).

Near-field optics deals with optical interactions on a subwavelength scale. In this sense, nonradiative interactions are of key interest. However, nonradiative interactions are found in so many different fields of study that it is rather difficult to incorporate them consistently into the field of near-field optics. For example, optical near-fields are key ingredients in Van der Waals attraction and in Förster energy transfer. The importance of near-fields was also realized in Arnold Sommerfeld's analysis of dipole radiation over lossy ground (Sommerfeld, 1909) and in Jonathan A. W. Zenneck's and Demetrius Hondros' study of guided electromagnetic waves on metal surfaces (Zenneck, 1907; Hondros, 1909). It would be a long haul to describe these developments in detail. We will touch on them only marginally in order to bring near-field optics can be found in the 1993 proceedings of the first conference on near-field optics held in Arc-et-Senans (Pohl, 1993). Another historical review written by Dieter W. Pohl summarizes the developments of the decade 1984–1994 (Pohl, 2004).

Research in the field of near-field optics was vitally important for the advance of the more general field of nano-optics (Novotny and Hecht, 2006) and the now independent fields of single molecule spectroscopy (Xie and Trautman, 1998) and nanoplasmonics (Xia and Halas, 2005). Over the past ten years, developments in near-field optics and near-field optical microscopy have been summarized in several review articles and books (Girard and Dereux, 1996; Paesler and Moyer, 1996; Fillard, 1996; Fischer, 1998; Dunn, 1999; Ohtsu and Hori, 1999; Courjon, 2003; Kawata, 2002; Richards and Zayats, 2004; Wiederrecht, 2004; Hong, 2004; Prasad, 2004; Keller, 2005; Novotny and Stranick, 2006; Novotny and Hecht, 2006; Bouhelier, 2006). As with all review articles and historical perspectives, it is not possible to account for all the individual contributions in the field. The purpose of this article is to provide a rough chronological outline of developments that lead to the field of near-field optics as it is known today. More recent developments are touched upon only peripherially.

This article is organized as follows: Following this short introduction I shortly review the classical diffraction limit and its consequences, and I discuss the basic ideas behind near-field optical microscopy. Section 3 then outlines the very first ideas behind nearfield microscopy which date back to the prophetic letters of Edward Hutchinson Synge sent in 1928 to Albert Einstein. Over the years, Synge's ideas were reinvented several times and his papers resurfaced only in the mid of the 1980s. After reviewing these very early proposals, Section 4 concentrates on the first experimental developments starting with acoustical experiments performed in 1956 by Albert V. Baez, followed by the microwave experiments of Ash and Nicholls in 1972. A section on surface plasmons and surface enhanced Raman scattering (SERS) reviews research in the 1970s and the early 1980s related to the development of a theoretical understanding of the SERS effect. The theoretical models developed in the SERS community brought important inspiration to the field of near-field optics as evidenced by the 1985 paper by John Wessel. With a similar purpose, Section 6 touches upon studies of energy transfer processes and reviews the ideas of Hans Kuhn of making use of optical near-fields for contact printing. First experimental developments of near-field *optical* microscopy are the subject of Section 7. Starting with the 1982 IBM patent by Dieter W. Pohl and the first measurements by his group in the same year we review independent parallel developments such as the one by Ulrich Ch. Fischer and Hans Kuhn at the Max-Planck-Institute in Göttingen and the one by the group of Aaron Lewis at Cornell University. The following years are characterized by technical improvements and first applications of near-field optical microscopy. New modalities, such as the photon scanning tunneling microscope (PSTM), were developed and it was realized that the interpretation of the contrast in the recorded images is not an easy task. Section 8 shortly reviews early theoretical studies, outlines the challenges associated with light-matter interactions in the near-field, and discusses the problem of inverse scattering. Scattering-based methods as well as tip-enhancement methods are then reviewed in Section 9. For metal tips and scattering particles, the similarity between near-field optics and antenna theory is particularly evident and this analogy is discussed in Section 10. In the same section, I also shortly reflect on Benjamin Franklin's invention of the lightning rod and its ability to localize static fields. The article is concluded with some final remarks in Section 11.

2 The Diffraction Limit

Near-field optics has its origin in the effort of overcoming the diffraction limit of optical imaging. At the end of the nineteenth century, Abbe and Rayleigh derived a criterion for the minimum distance Δx between two point sources at which they can still be unambiguously distinguished as two separate sources (Abbe, 1873; Rayleigh, 1896). Abbe's

criterion is illustrated in Fig. 1a and reads as^2

$$\Delta x = 0.61\lambda/NA . \tag{1}$$

Here, $NA = n \sin \Theta_{max}$ is the numerical aperture, a property of the optical system. n is the index of refraction of the surrounding medium and Θ_{max} is the maximum collection angle of the optical system. The best possible NA is NA = n which, for optical glasses, is $NA \approx 1.5$ and hence $\Delta x \approx \lambda/3$. The resolution can be increased further by the use of two opposing objectives as demonstrated by Hell and co-workers (Schrader and Hell, 1996).

The diffraction limit is often associated with Heisenberg's uncertainty principle (Vigoureux and Courjon, 1992). From Fig. 1a we recognize the maximum transverse wavenumber as $Max[k_{\parallel}] = \pm k \sin \Theta_{max}$ which defines the bandwidth of spatial frequencies as $\Delta k = 2 \operatorname{Max}[k_{\parallel}] = 4\pi N A \lambda$. Inserting into Eq. 1 we obtain $\Delta x \Delta k = 0.61 4\pi$. In agreement with the uncertainty principle this product is larger than 1/2. At first sight, the uncertainty product associated with the diffraction limit could be increased by a factor of $\approx 4\pi$. To accomplish this, the Airy function defining Eq. 1 would need to be replaced by the minimum uncertainty function, i.e. a Gaussian function. However, a Gaussian function has a Gaussian spectrum and hence involves evanescent waves which do not propagate through the optical system. It is the hard cut-off of spatial frequencies which gives rise to the Airy function and to a weaker uncertainty product. It is important to note that Abbe's and Rayleigh's criteria make *no* use of any information that is available on the properties of the two emitters. Furthermore, these criteria assume that the light-matter interaction is *linear*. However, by taking into account knowledge about the excitation properties or the spectral properties of the sample it is possible to stretch the resolution limit beyond the diffraction limit (Toraldo di Francia, 1955). For example, the distance between a red emitting molecule and a green emitting molecule can be measured with nanometer accuracy by using spectral filters to only pass the radiation from one molecule at a given time. In this case, resolution is reduced to the problem of position accuracy (Novotny and Hecht, 2006). Recent work has demonstrated that resolution can be extended beyond Abbe's limit by driving the light-matter interaction into saturation (Klar, 2000; Willig, 2006) or by recording higher harmonics of the sample's spatial frequencies (Gustafsson, 2005).

In near-field optics, the bandwidth of spatial frequencies is no longer limited by $\Delta k = 4\pi N A/\lambda$. Instead, the dependence on the wavelength λ is replaced by a dependence on a characteristic length d (e.g. aperture diameter or tip diameter) of a local probe. As illustrated in Fig. 1b,c, in near-field optical microscopy the local probe ensures a confined

²This result is based on a scalar paraxial approximation ($\sin \Theta_{max} \approx \Theta_{max}$) but it provides satisfactory accuracy even for high *NA* systems (Novotny and Hecht, 2006).

photon flux between the probe and the sample surface. The probe is raster-scanned over the sample surface and for predefined positions (x,y) of the probe a remote detector acquires the optical response. In this way an optical contrast image can be recorded. A reproduction of an early near-field optical scan trace is shown in Fig. 2. It was recorded on 22 October 1982 and is an excerpt from the laboratory book of Winfried Denk then working with Dieter Pohl at the IBM Research Laboratory.

3 Synge and Einstein

It is well known that the original idea of using a tiny aperture in an opaque screen for performing near-field optical imaging has been published by Edward Hutchinson Synge in 1928 (Synge, 1928), an Irishman living in Dublin. Synge's contributions and his later life are summarized in a beautiful article by McMullan (McMullan, 1990). Here, we shall



Figure 1: Schematic comparison of (a) diffraction-limited optical microscopy and (b,c) near-field optical microscopy. In (a) the point-spread function is defined by the wavelength λ and the numerical aperture $NA = n \sin \Theta_{max}$ whereas in (b,c) it depends on the size and proximity of a local probe (e.g. aperture or particle). Near-field optical imaging is a sampling technique, i.e. the sample is probed point-by-point by raster-scanning the local probe over the sample surface and recording for every image pixel a corresponding optical signature. The local probe functions as an optical antenna, converting localized energy into radiation, and vice versa.

revisit Synge's key ideas and outline their relationship to the developments in the field of near-field optics. In his 1928 publication, Synge writes "The idea of the method is exceedingly simple, and it has been suggested to me by a distinguished physicist that it would be of advantage to give it publicity, even though I was unable to develop it in more than an abstract way." Today, we know that the distinguished physicist to whom Synge was referring to is Albert Einstein.

In a letter dated 22 April 1928 Synge describes to Einstein a microscopic method in which not the field penetrating through a tiny aperture is used as a light source but the scattered field from a tiny particle. Synge states that "By means of the method the present theoretical limitation of the resolving power in microscopy seems to be completely removed and everything comes to depend upon <u>technical</u> perfection." In the same letter, Synge provides a sketch of the apparatus. Fig. 3a shows an adaptation of this sketch. Synge writes "If a small colloidal particle, e.g. of gold, be deposited upon a quartz slide placed above a Zeiss cardioid condenser of NA 1.05, then, all rays of light from the condenser which reach the surface of the slide will be totally reflected by the surface, except those which strike the surface at the base of the particle. These will be scattered in all directions and if the objective of a microscope is suitably arranged above the slide, a proportion of the rays so scattered will come to a focus in the eye of an observer, or upon a photographic plate, or a photo-electrical cell suitably placed." Synge describes here what is called today dark-field *microscopy*, a technique invented at the turn of the twentieth century by the Austrian chemist Richard Adolf Zsigmondy. In his letter, Synge then proposes to place a very thin stained biological section onto a quartz cover glass and to raster scan it in close distance



Figure 2: Early near-field optical scan trace recorded with an aperture-type probe. The data is from the laboratory book of Winfried Denk then working with Dieter Pohl at the IBM Research Laboratory in Switzerland.

over the irradiated particle. He argues that the amount of light received from the particle and collected by the objective will depend upon the relative opacity of the different parts of the section. In the remainder of the letter, Synge addresses different technical difficulties, describes the scanning process, and also proposes to embed the particle into the end face of the quartz slide. He also realizes a potential difficulty: "I am not sure how near the biological section could be brought to the surface of the quartz plate without impairing the totality of the reflection." He thinks this is the only potential theoretical limitation and that everything else depends only on perfecting the technique. Synge also writes that there is no institution in his country that could carry out the necessary experiments and he suggests that the experiments could be undertaken by the Institute of Physics in Berlin.

On 3 May 1928 Einstein replies with a short letter written in German and sent from Berlin. He states that he believes that Synge's basic idea is correct but that his particular implementation seems to be of no use ("prinzipiell unbrauchbar"). He argues that if the distance between the sample surface and the surface supporting the particle becomes small then there will be considerable light leakage (frustrated total internal reflection) and the total image field will become bright. Instead, Einstein suggests of using the light that penetrates through a tiny hole in an opaque layer as a light source. He also states that he couldn't read many parts of Synge's letter. This could be related to Synge's challenging handwriting (which improved in Synge's second letter).

Only five days later, on 9 May 1928 (imagine the efficiency of the postal service at that time), Synge replies to Einstein's letter and states "It was my original idea to have a very small hole in an opaque plate, as you suggest, and it was in this form that I had mentioned it to several people." In the same letter Synge suggests what later became the most standard way of fabricating aperture probes used in near-field optical microscopy: "A better way could be, if one could construct a little cone or pyramid of quartz glass having its point P brought to a sharpness of order 10^{-6} cm. One could then coat the sides and point with some suitable metal (e.g. in a vacuum tube) and then remove the metal from the point, until P was just exposed. I do not think such a thing would be beyond the capacities of a clever experimentalist." In a following paragraph Synge states that he is "sure that some idea of the kind will be made use of ultimately, but it obviously requires to drop into the brain of an experimental genius." A couple of decades later this prophecy was indeed verified.

Einstein's reply from 14 May 1928 does not address the new thoughts of Synge. Instead, Einstein refers to the original idea of using the scattered light from a tiny particle as a light source and he reiterates the problem of total internal reflection. Einstein states that he believes that it is practically impossible to generate a tiny light source through total internal reflection and scattering. He adds that he neither believes in the promise of the other proposals for generating a tiny light source and that it makes him hesitant to recommend it to an experimentalist. Einstein suggests that Synge publishes the idea in a scientific journal and that he highlights the technical difficulties associated with a practical implementation. This encouragement forms the decisive impetus that led Synge to publish his idea of aperture-based near-field optical microscopy (Synge, 1928). On 29 August 1928 Synge writes his last letter to Einstein. In this letter he proposes an arrangement for an X-ray interferometer and he mentions that he contacted Sir W. Bragg but that he couldn't get a satisfactory response. There appears to be no record of a response from Einstein.



Figure 3: (a) Synge's original proposal of near-field optical microscopy based on using the scattered light from a particle as a light source. The figure is adapted from Synge's letter dated 22 April 1928 sent to Einstein. (b) Wessel's proposal from 1985 showing the following elements: (a) particle, (b) sample surface, (c) optically transparent probe-tip holder, and (d) piezoelectric translators (Wessel, 1985). (c). 1988 experiment by Fischer and Pohl. The near-field probe consists of a gold coated polystyrene particle (Fischer and Pohl, 1989). (d) Figure 6 from Ueyanagi's 2001 patent (Ueyanagi, 2001): (1) optical head, (2b) parallel laser beam, (2c) convergent light, (5) objective lens, (6) transparent condensing medium, (6a) incident surface, (6b) light condensed surface, (8) micro metal member, (9) light spot, (10) near-field light, (12) disk, (120) disk-like plastic plate, (121) recording medium.

Synge's original proposals to overcome the diffraction limit of light microscopy were reinvented over the years and form the basis for both 'aperture' and 'apertureless' scanning near-field optical microscopy. Synge was also the first to propose the principle of scanning which forms today a key ingredient in a wide range of technologies ranging from television to scanning electron microscopy (SEM). Before Synge's time the idea of manipulating the position of a source relative to a target in an imaging apparatus and thereby improving its imaging capabilities was not known (McMullan, 1990). In a follow-up paper in 1932, Synge suggested the use of piezo-electric quartz crystals for rapidly and accurately scanning the specimen (Synge, 1932). He estimated that a translation of $5\mu m$ can be accomplished with a voltage of 250 V. This estimate matches perfectly the sensitivity of present-day piezo-electric transducers used in scanning probe microscopy. The idea of using piezo-electric transducers to control the gap between the probe (e.g. aperture) and the sample surface did not occur to him. This concept had to wait fifty years and was a key ingredient in the development of scanning tunneling microscopy (STM) (Binnig, 1982). In the same 1932 paper, Synge also suggested for the first time to use image processing to highlight certain features of an image before displaying it. According to McMullen (McMullan, 1990), Synge had many other visionary ideas, some of which became of importance in other scientific fields. In 1936 Synge had a mental breakdown and had to spend the rest of his life in a Dublin nursing home.

Although Einstein didn't believe in the practical realization of Synge's original idea the concept of using the scattered light from a tiny particle as a light source is today wellestablished and experimentally verified (Fischer and Pohl, 1989; Malmqvist and Hertz, 1994; Kawata, 1994; Anger, 2006; Kühn, 2006). In 1985 John Wessel proposed a method very similar to Synge's original idea (Wessel, 1985). His proposed arrangement is depicted in Fig. 3b. Wessel suggests to excite the particle resonantly thereby creating a locally enhanced field which serves as a light source. He is the first to mention the analogy to classical antenna theory. He writes: "The particle serves as an antenna that receives an incoming electromagnetic field." Wessel did not know of Synge's ideas and his proposal was likely inspired by the invention of STM (Binnig, 1982) and the discovery of surface enhanced Raman scattering (SERS) (Fleischmann, 1974; Jeanmaire and Van Duyne, 1977; Albrecht and Creighton, 1977). The quest for an understanding of SERS gave rise to many theoretical studies aimed at predicting the electromagnetic field enhancement near laser-irradiated metal particles and clusters thereof (Gersten and Nitzan, 1980; Wokaun, 1981; Boardman, 1982; Metiu, 1984; Meier, 1985). This era can be considered as the first phase of what is called today *nanoplasmonics* (Xia and Halas, 2005). In 1988 Ulrich Ch. Fischer and Dieter W. Pohl have carried out an experiment that is very similar to Synge's and Wessel's proposal (Fischer and Pohl, 1989). Instead of using a laser-irradiated solid metal particle as a local light source, they used a gold covered polystyrene particle (c.f.

Fig. 3c), a structure that was later extensively developed and named *gold nanoshell* (Jackson, 2003). Fischer and Pohl imaged a thin metal film with 320nm holes and demonstrated a spatial resolution of ≈ 50 nm. Their results provide the first experimental evidence that near-field scanning optical microscopy as originally proposed by Synge is feasible.

As an illustration of the experimental feasibility of Synge's original idea (and Wessel's proposal) Fig. 4 shows a fluorescence image of single molecules dispersed on a quartz substrate and imaged with a single laser-irradiated gold particle (Anger, 2006; Kühn, 2006). It turns out that the polarization conditions of the exciting laser radiation are important, - an ingredient that is not discussed in neither Synge's or Wessel's proposal. In many experiments Synge's particle is replaced by a bare metal tip and these methods became to be known as 'apertureless' or 'tip-enhanced' near-field optical microscopy (Zenhausern, 1995; Novotny, 1998). Ironically, the very idea of Synge, communicated in his first letter of 22 April 1928 to Einstein, has been patented in 2001 by Fuiji Xerox Co. (Ueyanagi, 2001). The inventor, Kiichi Ueyanagi describes different applications making use of an antenna-structure (such as a particle) fabricated into the end face of a dielectric medium to localize optical radiation. One of the figures is depicted in Fig. 3d.

4 First developments

Before the first experimental developments have been undertaken, Einstein's and Synge's idea of using an irradiated aperture in a flat metal screen (Synge, 1928) resurfaced in several other proposals. Not knowing of Synge's prophetic work, John A. O'Keefe publishes



Figure 4: Single molecule fluorescence imaging using the scattered light from a laserirradiated gold particle as light source (Anger, 2006). (a) Experimental arrangement. An 80nm gold particle is attached to the end of a etched glass tip and irradiated by a radially polarized laser beam. A sample with dispersed fluorescent molecules is raster-scanned underneath the gold particle. (b) Corresponding near-field fluorescence image.

in 1956 a short proposal starting with "The following is presented as a concept illustrating a method by which it might conceivably be possible to go beyond the resolving power of visible light" (O'Keefe, 1956). Similar to Synge, he proposes to employ an irradiated small hole in an aluminum coating as a light source. He writes "By scanning, an image can be built up whose detail will correspond to the size of the hole, even if it is smaller than the wavelength of the light used." O'Keefe indicates that he believes the realization of his proposal is remote because of the difficulty of providing for relative motion between the pinhole and the object. Only a few months later, Albert V. Baez publishes an article in which he discusses the results of a near-field acoustical experiment (Baez, 1956). For his demonstration, Baez used acoustic waves with a wavelength of 14cm (2.4 kHz) and confirmed a resolution not limited by diffraction. Baez used his own fingers as test objects and his results are mentioned only very qualitatively in the article.

More than ten years later Charles W. McCutchen discusses the possibility of overcoming the diffraction limit of optical imaging by convolving the spatial frequencies of the sample with the spatial frequencies of a probe object (McCutchen, 1967). His consideration is based on the assumption that the plane containing the probe object constitutes a spatial filter function for the field emanating from the sample plane. The probe object shifts the spatial spectrum of the sample field thereby making high spatial frequencies propagate. This principle is analogous to a radio receiver where a local oscillator (probe particle) shifts the signal frequencies (sample field) into the pass band (propagating radiation). According to McCutchen, it doesn't matter whether a particle or an aperture is scanned over the surface of a sample. What matters is that the probe object is very small so it provides the necessary high spatial frequencies. Of course, the application of simple linear filter theory is an oversimplification and it cannot account for strong interactions between sample and probe, e.g. for field enhancement effects and resonant interactions. Nevertheless, it provides an intuitive picture which is important for the understanding of image formation in near-field optical microsopy. McCutchen believes that it isn't practical to place the probe object right next to the sample and he considers the effect of moving the probe object behind the condenser lens. He argues that the bandwidth of spatial frequencies can be doubled, - a principle employed today in structured illumination microscopy (Gustafsson, 2005). McCutchen writes "In an extreme form, the technique would consist of scanning the specimen past a minute aperture, much smaller than the Abbe limit.

... Obviously, it would be hard to use this method on any but the flattest of surfaces, and I wonder if there are many jobs it could do that reflection electron microscopy would not do better." McCutchen's scepticism is justified, but it is the scientific curiosity and the potential benefit of combining the chemical specificity of optical spectroscopy with high-resolution optical microscopy which drives the progress in near-field optics forward. In 1984, Gail A. Massey uses Fourier optics to formulate the image forming process described by McCutchen (Massey, 1984), however without knowing of McCutchen's work. Massey

concludes that for an aperture of size d a lateral resolution of $\approx d$ can be achieved with a depth of focus of $\approx d/3$ (Massey, 1984).

In 1970, H. Nassenstein proposes to illuminate a sample with evanescent waves thereby generating scattered waves which contain information on spatial frequencies of the sample spectrum beyond the classical resolution limit (Nassenstein, 1970). Similar experiments were reported before in the context of holography Stetson (1967). Since evanescent waves are bound to their source (primary or secondary), Nassenstein's proposal implies that a secondary object (the probe) be brought close to the sample. Today, total internal reflection fluorescence (TIRF) microscopy is a powerful tool in biological research because it effectively reduces background fluorescence. There are commercially available TIRF microscope objectives with NA's of 1.5 - 1.65.

The first experimental validation of near-field microscopy using electromagnetic radiation has been undertaken by Eric A. Ash and G. Nicholls at University College, London. In a paper published in 1972 they use 10 GHz microwaves ($\lambda=3$ cm) and an aperture of 1.5 mm to image an aluminum test pattern deposited on a glass slide (Ash and Nicholls, 1972). At a separation between aperture and sample plane of 0.5 mm they are able to achieve a resolution of better than $\lambda/60$, clearly beyond the diffraction limit of standard microscopy. Ash and Nicholls state "... one might hope to build a super-resolution optical microscope," and they add "Although the optical microscope may not be beyond reach, in our view a more immediately hopeful application is the construction of an infrared microscope with a resolution comparable with that normally attainable in the visible optics spectrum." However, the history of near-field optics proceeded in the other sequence. In 1982 the first optical scan images were recorded by Dieter W. Pohl and co-workers at the IBM Research Laboratories (c.f. Fig. 2) and the infrared analogue has been demonstrated later in 1986 by Gail A. Massey using 100 μ m radiation (Massey, 1985).

5 Surface Plasmons and Surface Enhanced Raman Scattering

The field of near-field optics has been greatly established by the quest for diffractionunlimited microscopy and spectroscopy. But there are at least two other important developments which had a significant influence on the shaping of the field: 1) research in the field of metal optics and 2) the discovery of surface enhanced Raman scattering (SERS). In 1968 two different experiments, one by Andreas Otto and the other by Erwin Kretschmann and Heinz Räther, have demonstrated that plasma oscillations on thin metal films can be excited with an optical beam undergoing total internal reflection (Otto, 1968; Kretschmann and Räther, 1968). It was shown that these plasma oscillations are polaritons, i.e. their existence is coupled to an electromagnetic field, and today these excitations are referred to as surface *plasmon* polaritons. It is interesting to note that already in 1959 T. Turbadar used the Kretschmann configuration for reflectance measurements on aluminum films and he noticed a characteristic reflection dip beyond the critical angle of total internal reflection. The reflection dip was shown to be consistent with thin film theory and it was noted that it must be associated with an evanescent wave because there is no transmitted light (Turbadar, 1959). The existence of optical surface waves on metal surfaces was also noticed in 1941 by Ugo Fano in his theoretical analysis of anomalous diffraction gratings (Fano, 1941). The later experiments of Otto, Kretschmann and Raether gave rise to various dedicated experiments aimed at understanding the confinement of optical fields near the surface of metals. On planar metal surfaces, plasmons are nonradiative. They propagate along the surface and their wavelength at a given frequency ω is shorter than the one in free space. At the surface plasmon frequency, and in the ideal case of no damping, the wavelength of the plasmon polariton goes to zero and the associated field becomes localized to the very surface of the metal, i.e. the decay length of the evanescent wave goes to zero. Besides the interest in surface plasmons propagating along extended interfaces, experimental and theoretical work had also been undertaken to understand the optical response of finite sized metal particles. The work of Uwe Kreibig provided an understanding for the transition of macroscopic electromagnetic theories applicable to particles larger than the mean-free path of electrons in the metal to quantum theories applicable to clusters of a few metal atoms (Kreibig, 1974). Surface plasmons associated with finite metal particles and clusters thereof gained a lot of importance for the understanding of the SERS effect which was discovered in 1974 (Fleischmann, 1974). Shortly after, it was realized that the phenomenon is due to the increased surface of roughened metal surfaces and the associated electromagnetic field enhancement (Jeanmaire and Van Duyne, 1977; Albrecht and Creighton, 1977). In the early 1980s much theoretical work was produced with the aim of understanding the SERS effect (Chang and Furtak, 1981). The near-fields of various metal nanoparticle arrangements were calculated and it was concluded that the effect is mediated by so-called 'hot spots', i.e. localized regions between metal particle aggregates that exhibit particularly strong electromagnetic field enhancement. A recent retrospective has been written by Martin Moskovits (Moskovits, 2005). The enhanced near fields around metal nanoparticles has also been shown to lead to several interesting photochemical phenomena, including near-field driven plasmon-resonant particle growth (Chen and Osgood, 1983). The quest for an understanding of the SERS effect marked the beginning of the first wave of *nanoplasmonics* and gave rise to new schemes of near-field optical microscopy, such as Wessel's proposal (Wessel, 1985). The electromagnetic theories and calculations developed in the early phase of SERS proved very beneficial for the understanding of light localization and near-field interactions. On the other hand, the development of near field optical microscopy provided for the first

time direct access to the fields associated with surface plasmon polaritons (Specht, 1992; Marti, 1993; Dawson, 1994) and gave rise to the recent revival of nanoplasmonics.

6 Studies and Applications of Energy Transfer

Independent of the activities in SERS, plasmonics, or near-field microscopy, short-range optical interactions were also investigated in the context of energy transfer between molecules (Förster, 1946) and fluorescence quenching near metal surfaces (Kuhn, 1970; Drexhage, 1970; Zingsheim, 1976). Already in 1970 Hans Kuhn suggested to make use of optical near-fields for contact imaging (Kuhn, 1970). He envisioned to use short-range energy transfer from electronically excited dye molecules as a method for the duplication of nanostructures. The scheme was later implemented by Hans P. Zingsheim and Ulrich Ch. Fischer (Fischer and Zingsheim, 1982). In this experiment a monomolecular film of



Figure 5: Near-field optical contact printing originally proposed by Hans Kuhn. A nanoscale pattern is transfered to a dye layer by placing a laser-irradiated nanostructured metal film on top of the dye layer. Short-range bleaching the fluorescence of the dye layer gives rise to spatial patterning of the fluorescence intensity. (a) Fluorescence image of the dye layer during contact with a metal pattern consisting of platinum disks (diameter 8μ m). Close to the disks the fluorescence is diminished because of fluorescence quenching. (b) After release of the metal mask regions of the dye layer which were close to individual metal disks exhibit stronger fluorescence (quenching lowers the bleaching rate). (c,d) show the corresponding arrangements of layer and mask. From (Fischer and Zingsheim, 1982). dye molecules serves as a light sensitive film, and a very thin, only partially absorbing, planar metal pattern which is embedded into the surface of a pliable polymer film, serves as a conformal mask (c.f Fig. 5). After the film is brought into contact with the mask and after it has been irradiated with light, the structure is transferred from the mask to the monomolecular film as a pattern of areas where the dye is bleached and where it is not bleached. The resolution of this pattern transfer is not limited by the wavelength of light but by the range of the energy transfer mechanism and the distance between the mask and the dye layer (Fischer, 1998). In principle, this process of pattern transfer makes use of the short range of the near-field of single electronically exited molecules in order to obtain diffraction-unlimited resolution down to the molecular scale. Studies of energy-transfer processes established that many types of near-field interactions can be understood on a classical phenomenological basis (Chance, 1978; Kerker, 1980). The phenomenological theory treats the quantum mechanical transition probability between two states as a classical dipole oscillating at the transition frequency. The theory turns out to be equivalent to a rigorous quantum-electromagnetic theory in the weak coupling regime (Novotny and Hecht, 2006) but it is much easier to deal with. An extensive review of surface enhanced spectroscopy and energy transfer processes between molecules and surfaces of various shapes has been provided in 1984 by Horia Metiu (Metiu, 1984) and also by George W. Ford and Willes H. Weber (Ford, 1984). The latter review also discusses the nonlocal dielectric response near material boundaries (spatial dispersion). In later work, Förster energy transfer has been proposed as an interaction mechanism for near-field optical imaging (Kopelman and Tan, 1994; Fujihira, 1996; Sekatskii, 1996; Vickery and Dunn, 1999). In 1999 the group of Robert C. Dunn provided the first experimental demonstration of near-field energy transfer microscopy with sub-wavelength resolution (Vickery and Dunn, 1999) and the following year the emission from a single molecule was used for the first time as a local light source (Michaelis, 2000).

7 First developments of Near-field optical Microscopy

First dedicated efforts of demonstrating near-field microscopy at *optical* frequencies started in the early 1980s. The first developments proceeded without the knowledge of previous proposals and the prophetic papers by Synge. On 27 December 1982 Dieter W. Pohl, then working at the IBM Research Laboratory in Switzerland, filed a patent titled *optical* near-field scanning microscope (Pohl, 1984b) which describes an aperture-based method very similar as conceived by Synge more than fifty years earlier. The patent states "The term 'near-field' is intended to express the fact that the aperture is located near the object at a distance smaller than the wavelength. The term 'aperture' is used here to describe the pointed end of a light waveguide which forms an entrance pupil with a diameter of less than $1\mu m$." In the same year, Pohl and co-workers managed to overcome the remaining experimental hurdles and recorded the first optical scan trace shown in Fig. 2. Their first scientific publication appeared with some delay in 1984 (Pohl, 1984). In this publication near-field optical microscopy is referred to as *optical stethoscopy* in analogy to the acoustic stethoscope used in medical diagnosis. Pohl, Denk, and Lanz write "*The familiar medical doctor's stethoscope, for instance, allows localization of the position of the heart to within less than 10 cm by moving the stethoscope over the patient's chest, and listening to the sound of the heart beat. Assuming a sound frequency of 30-100 Hz, corresponding to a wavelength of almost 100m, the stethoscope provides a resolving power of roughly \lambda/1000 ! "Their aperture light-source was formed by pressing an aluminum coated, electrochemically etched quartz crystal towards a transparent sample. Laser light is coupled into the crystal and as soon as light is transmitted through the sample a tiny aperture has been formed. This procedure for forming apertures has been later named <i>pounding* or *punching*.

During the same time other groups have started similar efforts. Results by Ulrich Ch. Fischer were presented in a talk by Hans Kuhn at the second Meeting of Molecular Electronic Devices in 1983. The proceedings of this meeting were published much later, in 1987, because the editor passed away in the mean time (Kuhn, 1987). One of the scan traces was also reproduced in the 1984 yearbook of the Max-Planck-Society (Kuhn, 1984) (summary of research activities of 1983) and is shown in Fig. 6 along with the outline of the experimental setup. In their experiments, Fischer and Kuhn used a glass hemisphere coated with a Tantalum/Tungsten layer in which a 100nm hole was fabricated. A similar metal layer (20nm thickness) with ten times larger holes $(1\mu m)$ was fabricated onto a glass slide and served as a test sample. The hemisphere was brought close to the sample and then scanned (without feedback control) along the surface. In Fig. 6b two different



Figure 6: Experiment by Ulrich Ch. Fischer performed in 1983 while working with Hans Kuhn. a) Schematic of the experiment. A metal-coated glass hemisphere with a 100nm hole is used to irradiate a test sample made of a metal layer with 1μ m holes deposited onto a glass slide. From (Kuhn, 1987). b) Optical scan traces recorded along the solid line (top trace) and along the dashed line (bottom trace). From (Kuhn, 1984).

scan traces are shown, one through the center of the sample hole (top) and one closer to its edge (bottom). The slope of the upper trace indicates a resolution better than the diffraction limit. During the same time, Fischer and Zingsheim pioneered what is called today nanosphere lithography (Fischer and Zingsheim, 1982). In this technique, a suspension of colloidal particles with a diameter of > 100 nm is deposited onto a plane surface. Subsequent evaporation of the solvent arranges the particles in a hexagonal twodimensional lattice. Vacuum deposition of metals and other materials through the voids between neighboring spheres leads to arrays of triangular structures on the surface. The size and the periodicity of these patterns can be varied by the size of the original particles. Double exposure from different angles of incidence allows more complex pattern to be created (Haynes and Van Duyne, 2001). The resulting *Fischer patterns* are used as test samples for microscopy or for Raman active substrates for the detection of target agents.

At the time of the experiments of Fischer and the experiments performed at IBM by Pohl and co-workers another development was underway at Cornell University. Aaron Lewis and co-workers studied light transmission through arrays of holes in planar metal films and were working towards a "scanning nanometer optical spectral microscope." The first document mentioning their effort is an abstract from the 1983 Biophysics Meeting (Lewis, 1983). One year later they published an article presenting light transmission through 30 nm apertures in metal films (Lewis, 1984). In the same article they discuss "the possibility of constructing a scanning optical microscope based on near field imaging which could potentially have spatial resolutions as small as one-tenth (of) the wavelength of the incident light' (Lewis, 1984). Their first experimental results made use of thermally pulled glass capillaries, as used in the patch-clamp technique, and were published in 1986 (Harootunian, 1986). These experiments used near-field excited fluorescence as contrast mechanism and demonstrated a resolution on the order of the aperture diameter of 100 nm. In the same publication, Lewis and co-workers introduced the acronym NSOM. standing for "near-field scanning optical microscopy". Earlier, in the same year, Urs Dürig, Dieter W. Pohl and Flavio Rohner publish transmission near-field images recorded with aperture probes and by using active distance control via electron tunneling between probe and sample (Dürig, 1986). They refer to the technique as NFOS microscopy. The acronym SNOM was created later in 1988 to emphasize the analogy to SEM, STM, and other scanning microscopies. The optical near-fields near irradiated apertures were not only considered for microscopy but also for sensing applications. In 1986 Ulrich Ch. Fischer publishes a study on the transmission of tiny apertures in metal films (Fischer, 1985). The following year he demonstrates that the aperture acts as probe of its microenvironment through enhanced light scattering and fluorescence (Fischer, 1986). This approach has been later revisited and applied to studies of single-molecule dynamics (Levene, 2003).

A significant breakthrough in the application of near-field optical microscopy came in 1991 when Eric Betzig and co-workers introduced aperture probes formed at the end of metal coated, thermally pulled quartz fibers (Betzig, 1991). This method became widely used and was also adopted by the first companies that pursued a commercialization of near-field optical microscopes. Another important technical improvement came in the following year, in 1992, when two independent groups introduced the shear-force technique to control the distance between probe and sample (Toledo-Crow, 1992; Betzig, 1992). In 1995, the shear-force method was combined with the sensing capability of a tuning fork crystal (Karrai and Grober, 1995) which, today, is the most widely used method for probe-sample distance control. The group of Betzig at Bell Laboratories pioneered many applications of near-field optical microscopy, published many highlights, and contributed to the general acceptance of the method (Betzig and Trautman, 1992). In 1994, near-field microscopy was applied for the very first imaging of single fluorescent molecules (Trautman, 1994). While single molecules have been detected before by use of spectral identification at cryogenic temperatures (Moerner and Kador, 1989; Orrit and Bernard, 1990), it was the *spatial* mapping which triggered the birth of *single molecule* spectroscopy (Xie and Trautman, 1998). As an illustration, Fig. 7 shows a fluorescence image of a sample with single dye molecules recorded with an aperture probe that was created by focused ion beam milling (Veerman, 1998).

Over the years, many variations of near-field optical microscopy have been explored and developed. Probably the most notable one is photon scanning tunneling microscopy (PSTM), also called scanning tunneling optical microscopy (STOM), developed in 1989 by three different groups (Courjon, 1989; Reddik, 1989; deFornel, 1989). This method



Figure 7: (a) SEM image of smooth aperture probe fabricated by slicing away the end of a metal coated, tapered optical fiber with a focused ion beam (FIB). From (Veerman, 1998). (b) Single molecule fluorescence image acquired with a FIB milled aperture probe. From (Moerland, 2005).

is the optical analog of STM as it probes the tunneling photons between a surface and a local probe. Typically, an evanescent field is created by total internal reflection at a dielectric-air interface and the tip of a pointed optical fiber is used to locally convert the evanescent field into a propagating waveguide mode, similar to frustrated total internal reflection (Novotny and Hecht, 2006). PSTM is a very attractive method because of its simple physical picture, but the recorded images were not easy to interpret, mainly because of multiple scattering between separated objects on the sample. The reconstruction of the sample features based on the measured optical information, the inverse scattering problem, can be facilitated by tuning the angle of incidence of the total internally reflected wave (Garcia and Nieto-Vesperinas, 1995). Several groups have theoretically established that PSTM measurements are inherently holographic and that they provide enough information to determine the two dimensional structure of a thin sample (Greffet, 1995; Bozhevolnyi and Vohnsen, 1996; Carney, 2004). A series of related studies on near-field phase conjugation have been performed by Sergey I. Bozhevolnyi and coworkers (Bozhevolnyi, 1994, 1995).

In modified form, the PSTM found interesting applications in optoelectronics and optical waveguides. Here, the evanescent tail of a waveguide mode is directly measured with a near-field probe rendering spatial maps of the electromagnetic field distribution. In a landmark experiment, Niek van Hulst's group combined this method with heterodyne detection and generated phase and amplitude maps of femtosecond pulses propagating along ridge waveguides (Balistreri, 2001). These experiments, shown in Fig. 8, directly visualized the electromagnetic field associated with an optical pulse and demonstrated



Figure 8: Measurement of the electromagnetic field of a light pulse propagating along a ridge waveguide. Left: Schematic of a heterodyne PSTM with variable time delay in the reference arm. The instrument probes the evanescent tail of a waveguide mode and renders a temporally and spatially resolved map of the phase and amplitude distribution of the electromagnetic field associated with the pulse. Right: Map of the measured amplitude multiplied with the cosine of the measured phase for a single tracked pulse. The area depicted is an enlargement of a small part of the actual scan. From (Balistreri, 2001).

that optical processes and phenomena can be probed by sampling their evanescent fields. Recently it was demonstrated that waveguide modes can also be probed by light scattering from a metal tip (Stefanon, 2005). PSTM was also applied to various other phenomena, such as light localization in random media (Gresillon, 1999; Bozhevolnyi, 2002) or propagation of surface plasmon polaritons (Marti, 1993; Dawson, 1994; Krenn and Weeber, 2004). Fig. 9 shows an example of a recent measurement by the group of Joachim R. Krenn. The figure depicts a map of the measured light intensity of a surface plasmon propagating along a silver nanowire.

In the 1990s, near-field optical microscopy has been applied to various problems and the progress is best summarized by referring to more detailed review papers (Girard and Dereux, 1996; Fischer, 1998; Dunn, 1999; Pohl, 2004). Very high spatial resolutions were demonstrated, but the nature of the optical contrast in the recorded images was often not understood. In fact, some optical images exhibited suspiciously close resemblance with the simultaneously recorded shear-force images. It was soon realized that two distinct properties contribute to the recorded optical signal: 1) the local material-specific response due to the probe-sample interaction, and 2) the vertical motion of the probe due to probe-sample distance control. While the former is the true optical contrast, the latter is an artifact and is due to the fact that the strength of the near-field interaction depends on the proximity of the probe. A variation of the optical signal is generated even if the probe is located at a fixed lateral position and its vertical position over the sample surface is varied. In 1997, Bert Hecht and collaborators have published an article titled Facts and Artifacts in Near-field Optical Microscopy in which they concluded that many published images represent the path of the probe rather than the true optical properties of the sample (Hecht, 1997). Following this paper, previous results had to be re-examined and new results had to be critically tested before being published.



Figure 9: Cylindrical surface plasmon propagating towards the end of a silver nanowire. The figure shows a map of the light intensity measured with a PSTM. From (Ditlbacher, 2005).

8 Theoretical Near-field Optics

Initial theoretical work in the field of near-field optics aimed at developing an understanding of image formation in near-field optical imaging. Approximate methods such as scalar diffraction theory break down in the near-field making it necessary to solve the full vectorial wave equation for a given problem. In a 1931 publication, Synge writes "This theory (diffraction theory) is, in most cases, a very good approximation, and forms the basis of the theory of resolution of optical instruments as usually presented, but it is by no means an absolute theory, such a theory requiring the solution of the electromagnetic equations. subject to boundary conditions which have been a bar to their solution except in one very simple case. In general we may say that when we come down to magnitudes of the order of a wavelength the approximate theory ceases to be a good approximation" (Synge, 1931). The theory for understanding the field distributions near tiny apertures in metal screens has been developed long before the interest in near-field optics started. It was Hans Bethe in 1944 who provided the first rigorous description (Bethe, 1944). His theory was later corrected and generalized by C. J. Bouwkamp in 1950 (Bouwkamp, 1950,b). For real metals and for screens of finite thickness the BetheBouwkamp theory still agrees qualitatively with the true field distribution (Novotny and Hecht, 2006). First theoretical studies of near-field optical microscopy made use of the formulas of Bethe and Bouwkamp and used Fourier optics to propagate the fields (Massey, 1984; Betzig, 1986; Roberts, 1987; Leviatan, 1986). Also, intuitive models were developed in which the near-field probe was treated as an elementary dipole (Van Labeke and Barchiesi, 1993; Labani, 1990). In the 1990s different methods were introduced to solve the full vectorial wave equation for a given near-field



Figure 10: Calculations of the electromagnetic local density of states (LDOS). (a) LDOS above a circular 'optical corral' built from dielectric particles deposited on a quartz substrate ($\lambda = 440$ nm). From Ref. (Colas des Francs, 2001). (b) LDOS over a semi-infinite sample of aluminum as a function of frequency and evaluated at different heights. From Ref. (Joulain, 2003).

configuration. Among the most widely used methods were plane wave expansion techniques (Van Labeke and Barchiesi, 1992), Green's function techniques (Dereux and Pohl, 1993; Girard and Dereux, 1994; Martin, 1994), and the multiple multipole (MMP) technique (Novotny, 1994,b, 1995). These methods have an analytical foundation and are summarized in Ref. (Novotny and Hecht, 2006). A review of early theoretical activities in near-field optics has been written by Christian Girard and Alain Dereux (Girard and Dereux, 1996). The goal of these early studies was to understand how interactions in the near-field are mapped to the farfield. Most commonly, the inverse scattering problem cannot be solved in a unique way and calculations of field-distributions are needed to provide prior knowledge about source and scattering objects and to restrict the set of possible solutions. For example, experimental near-field images revealed contrast reversal if the collection angle of the near-field scattered light was modified (Hecht, 1995). It was shown that this contrast reversal originates from evanescent wave scattering giving rise to supercritical light propagation (forbidden light) in a dielectric sample (Novotny, 1997). In 1995 it was shown that in some cases it is possible to assume that the interaction between probe and object can be neglected (weak coupling) which results in a simple linear detection process (Carminati and Greffet, 1995). In this regime, the reciprocity theorem requires that near-field optical images recorded in the PSTM mode and images recorded with aperture-based microscopy are equivalent (Mendez, 1997). The theory underlying image formation in near-field optics has been reviewed in 1997 by Jean-Jacques Greffet and Remi Carminati (Greffet and Carminati, 1997). Theoretical studies gave input to improved instrument design and predicted new detection strategies for optimizing the signal-to-noise ratio (SNR) in a given measurement (Girard, 1994; Greffet and Carminati, 1997; Novotny, 1997b; Hecht, 1998). Ultimately, it is the SNR which determines the best achievable resolution because, according to the principle of analytical continuation, a signal with finite support can be exactly reconstructed from a noise-free measurement in an arbitrarily small spatial domain (Devaney and Wolf, 1973; Wolf and Nieto-Vesperinas, 1985).

More recent theoretical studies aimed at understanding the physical properties of optical near-fields. Among the topics studied were coherence properties (Carminati and Greffet, 1999; Roychowdhury and Wolf, 2003), the polarization state of optical nearfields (Setälä, 2002; Ellis, 2005), spontaneous emission (Girard, 1995; Novotny, 1996) and local density of states near nanoscale structures c.f. Fig. 10) (Colas des Francs, 2001; Joulain, 2003; Novotny and Hecht, 2006), reciprocity relations in the optical nearfield (Carminati, 1998), and fluctuation-induced friction (Zurita-Sanchez, 2004). The fluctuational properties of optical near-fields have been discussed in two recent review papers (Henkel, 2005; Joulain, 2005). Studies of near-field inverse scattering have been pursued by different groups, mainly using the PSTM geometry (Garcia and Nieto-Vesperinas, 1995; Greffet, 1995; Bozhevolnyi and Vohnsen, 1996; Greffet and Carminati, 1997; Carney and Schotland, 2003). It was shown that samples can be uniquely reconstructed from near-field optical measurements by use of near-field tomography (Carney, 2004).

While first studies on the quantum nature of evanescent fields have been performed already in the 1970s by Girish S. Agarwal (Agarwal, 1975) and Chuck Carniglia and Leonard Mandel (Carniglia and Mandel, 1971) a quantized theory of spatially confined light has been put forth by Ole Keller in 1998 (Keller, 1998, 2000b, 2005). Keller also described the birth process of the photon wavefunction and pointed out that in a nearfield interaction the photon is destroyed before it is fully born (Keller, 2002). Another problem is the fact that it is not strictly possible to separate the source of radiation from the sink of radiation. Instead, source and sink appear as a coupled object or, more formally, it is not possible to independently define the state of source and detector (Power and Thirunamachandran, 1997). A remarkable result of Keller's work is the finding that only after an infinite time after its birth, the energy of a photon is $\hbar\omega_o$, ω_o being the transition frequency. At shorter times, the photon energy is larger than $\hbar\omega_{\alpha}$. Part of the problem of defining a near-field photon is associated with the fact that the near-field is not purely transverse, which can be easily verified for an evanescent wave and its excitation. Standard quantum electrodynamics (QED) proceeds by invoking the Coulomb gauge and quantizing the retarded transverse field. It gives little attention to the 'attached' field. However, from single molecule experiments it is known that a molecule close to an interface interacts with the *total* field and not only with its transverse part (Drexhage, 1974). Future theories and experiments will shed more light on the existence of *near-field photons*.

Because of their localized nature, optical near-fields can vary substantially over the length scale defined by a quantum system, such as a quantum dot or a molecule. Hence, the light-matter interaction can no longer be restricted to the dipole selection rules and higher order multipolar transitions need to be considered (Zurita-Sanchez and Novotny, 2002). In the extreme case, the multipolar expansion does not converge and the optical near-field becomes a probe for the local orbital overlap between the system's ground state and excited state. It is also interesting to note that in the light-matter interaction the momentum of a photon $(p = 2\pi\hbar/\lambda)$ is typically neglected because it is much smaller than the electron momentum in matter $(p = \sqrt{2m_{eff}E})$. Consequently, photoinduced band-to-band transitions in an electronic dispersion diagram happen vertically. However, the momentum of a photon associated with the optical near-field is no longer defined by the wavelength but by a characteristic length d associated with the optical confinement $(p = 2\pi\hbar/d)$. This makes the near-field momentum comparable with the electron momentum and hence intraband transitions become possible (Beversluis, 2003). The high momenta associated with the optical near-field as well as the possibility of accessing dipole-forbidden transitions will enrich optical spectroscopy and open up new and exciting frontiers.

9 Near-field Scattering and Field Enhancement

Let us now go back in time to revisit Synge's original proposal of using the light scattered by a small particle as a light source. When brought close to a sample surface, the particle not only scatters the incident field but also the field that is scattered from the surface. In fact, there is an infinite number of scattering iterations between particle and sample. Depending on the properties of particle and sample and on the excitation conditions only one or a few terms in this series are relevant. For example, in Wessel's proposal (Wessel, 1985), the particle's response is resonant with the incident field and hence the enhanced field generated by the particle can be regarded as an independent light source exciting the sample at short distance. On the other hand, one could consider a situation in which the interaction between the exciting field and the sample is more dominant than the interaction of the external field and the particle. In this case, the particle acts as a passive probe that scatters away the near-field of the irradiated sample. Both approaches have been implemented in near-field optics using pointed probes such as dielectric or metal tips as local scatterers. In *weak scattering* the probe acts as a local perturbation (Zenhausern, 1994; Bachelot, 1994; Inouye and Kawata, 1994) whereas in strong scattering the interaction between the probe and the exciting field dominates and the probe acts as an *optical*



Figure 11: 'Apertureless' near-field optical microscope using heterodyne detection. Original drawing from the patent of H. Kumar Wickramasinghe and Clayton C. Williams (Wickramasinghe and Williams, 1990). (12) end of a tip, (14) tip, (40) optical source, (42) acousto-optic modulator, (44,46) lens, (48) beam splitter, (50) pin photodiode.

antenna (c.f. section 10), a device that efficiently converts the energy of free propagating radiation into localized energy, and vice versa (Keilmann, 1995; Novotny, 1998). Whether a probe acts as a local perturbation or as an optical antenna depends on the particular experimental implementation. A recent review of tip-based near-field optical microscopy can be found in Ref. (Novotny and Stranick, 2006).

As mentioned before, the first experimental demonstration of Synge's particle-based idea has been presented by the pioneers of near-field optical microscopy, Ulrich Ch. Fischer and Dieter W. Pohl (Fischer and Pohl, 1989). In 1992, scattering from a metal tip was applied for the detection and imaging of surface plasmon polaritons (Specht, 1992; Hollander, 1995) and in 1994 experiments by the groups of Satoshi Kawata (Inouye and Kawata, 1994), A. Claude Boccara (Bachelot, 1994), and H. Kumar Wickramasinghe (Zenhausern, 1994) established that scattering-based near-field microcopy is a viable alternative to standard aperture based approaches. While the original role of the aperture was to confine an optical field beyond the limits of diffraction, the role of the tip was to establish a local interaction and to scatter away the local field. To express this different viewpoint, scattering-based approaches are also referred to as *apertureless* near-field optical microscopy. Wickramasinghe's experiments were already proposed in a patent filed in 1989 in which he named the method "apertureless near-field optical microscopy" (Wickramasinghe and Williams, 1990). Fig. 11 depicts the first figure from this patent. They write: "For example, an ideal conical tip having a single atom or group of atoms at the very end which is illuminated by a focused light source, will result in optical evanescent fields diverging from the tip. The divergent fields will interact with the sample surface on a local scale. These fields will be scattered by the surface and a portion will



Figure 12: Scattering-based near-field optical microscopy of a gold Fischer pattern with polystyrene residues deposited on a silicon surface. The method detects the backscattered optical signal at a higher harmonic of the modulation frequency Ω . (a) Schematic of the experiment, (b) topographical map, (c) optical map. Based on the different optical contrast, gold and polystyrene can be distinguished with a spatial resolution of $\approx 10 \text{ nm}$. From (Hillenbrand and Keilmann, 2002).

propagate into the far field, where the fields may be detected, providing a useful signal for measuring the local optical and topographical properties of the surface with high resolution." This sentence assumes that the interaction between the tip and the excitation field is stronger than the interaction between the sample and the excitation field, similar to Wessel's proposal. In their patent, Wickramasinghe and Williams propose to use a double modulation technique combined with heterodyne detection in order to extract the near-field signal from the detected scattered light. In their later experiments, Wickramasinghe and co-workers implemented a slightly different interferometric detection scheme and demonstrated extremely high spatial resolutions (Zenhausern, 1995). However, as discussed before, the origin of the contrast in the recorded images has been debated. A Japanese patent with very similar ideas was filed in 1992 by Satoshi Kawata and coworkers (Kawata, 1992). The patent describes a near-field optical microscope using a combination of total-internal reflection illumination and light scattering from a vertically modulated tip. A parallel effort in scattering-type near-field microscopy was also pursued by Fritz Keilmann with experiments performed at radio and microwave frequencies and later in the infrared (Keilmann, 1996; Knoll, 1997; Knoll and Keilmann, 1998). In 1989, a few months after Wickramasinghe's patent submission, Keilmann filed a patent titled scanning tip for optical radiation in which he proposes the fabrication of a coaxial tip. i.e. an aperture with a center conductor (Keilmann, 1991), an endeavor first undertaken by Fee, Chu, and Hänsch at microwave frequencies (Fee, 1989) and later by Fischer and Zapletal in the optical frequency regime (Fischer and Zapletal, 1992). Coaxial waveguides have no cut-off and therefore provide near-unity power transmission. Later, Fritz Keilmann and Bernhard Knoll implemented a method to extract the near-field signal from the scattered light (Wurtz, 1998; Knoll and Keilmann, 2000). The method makes use of vertical probe modulation with frequency Ω and demodulation of the scattered light at higher harmonics $n\Omega$. A combination with heterodyne detection allowed Fritz Keilmann and Rainer Hillenbrand to separately measure amplitude and phase of the scattered signal and to extract material specific optical parameters (Hillenbrand and Keilmann, 2000, 2002; Keilmann and Hillenbrand, 2004). As an example, Fig. 12 shows an image of a gold Fischer pattern with polystyrene residues deposited on a silicon surface. Polystyrene and gold yield clearly different optical contrast.

In 1997 it was proposed to use the enhanced field at a laser-irradiated metal tip as an excitation source, similar to Wessel's idea (Novotny, 1998). The interaction of the locally enhanced field with the sample surface generates an optical response which is then coupled out by the same tip and detected in the farfield. This strong-scattering scheme makes it possible to detect a spectroscopic response at frequencies *different* from the excitation frequency thereby making it possible to explore the full range of linear and nonlinear optical spectroscopy. The first experimental demonstration employed two-photon excited fluorescence and was published in 1999 (Sanchez, 1999). Similar experiments have also been

performed by other groups (Hamann, 2000) and today, the method is most widely referred to as *tip-enhanced near-field optical microscopy* (Novotny and Stranick, 2006; Bouhelier, 2006). Following these experiments, the method was extended to other spectroscopic interactions such as Raman scattering (Stöckle, 2000; Anderson, 2002; Hayazawa, 2002; Hartschuh, 2003) and coherent anti-Stokes Raman scattering (Ichimura, 2005). As an illustration, Fig. 13 shows a near-field Raman scattering image of a single-walled carbon nanotube sample along with a Raman scattering spectrum recorded when the tip is placed on top of the nanotube.

The field enhancement at a laser-irradiated metal tip has been the subject of many theoretical studies. Winfried Denk and Dieter W. Pohl demonstrated that the quasi-static fields in the gap between a tip and a substrate can be extremely strong and that this effect can be instrumental for inelastic tunneling and light emission during scanning tunneling microscopy (STM) (Denk and Pohl, 1991). Later, it was theoretically established that the field enhancement effect must be driven by an external field polarized along the axis of the pointed probe (Novotny, 1997; Martin and Girard, 1997; Furukawa and Kawata, 1998). Interestingly, the field enhancement was also studied in the context of STM as it was believed to mediate the transfer of atoms from the tip to the sample (Jersch and Dickman, 1996; Gorbunov and Pompe, 1994; Bragas, 1998).



Figure 13: Near-field Raman imaging of a single-walled carbon nanotube sample. (a) Topography showing a network of carbon nanotubes overgrown with water droplets. (b) Raman scattering 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 $\nu = 2615 \, cm^2$ (indicated by the yellow stripe in c). (c) Raman scattering spectrum recorded on top of the nanotube. Adapted from (Hartschuh, 2003).

10 Near-field Optics and Antenna Theory

In this last section I intend to outline similarities between near-field optics and antenna theory (Pohl, 2000). These similarities project the roots of near-field optics more than 100 years back to the time of Guglielmo Marconi's experiments on radiowave transmission. The quality of an antenna is characterized by quantities such as radiation efficiency and antenna gain, and it is plausible that similar quantities are applicable to optical near-field probes. Therefore, established antenna-concepts can provide inspiration for novel nearfield probes.

The primary function of a near-field probe is the concentration of electromagnetic energy on a sample surface, similar to a standard electromagnetic antenna that concentrates propagating radiation into a confined zone called the feedgap. In the feedgap, electric circuitry either releases or receives the power associated with the electromagnetic field. The challenge in the design of an antenna is to efficiently couple the power flow between the near-zone and the far-zone of the source (or receiver). This criterion holds also for a quantum source, such as a single molecule, which emits a single photon at a time. The most efficient antenna designs that have been implemented at optical frequencies are the half-wave antenna (Mühlschlegel, 2005) and the bow-tie antenna (Grober, 1997; Crozier, 2003; Schuck, 2005; Farahani, 2005). However, by making use of electromagnetic resonances associated with surface plasmons and phonons any nanostructure can be viewed as an optical antenna. Of course, the efficiency depends on the material properties and the geometry of the nanostructure.

As mentioned earlier, the association of near-field optical microscopy with antenna theory has already been made by Wessel (Wessel, 1985). However, the notion of 'optical antennas' can be traced back even earlier. For example, an antenna attached to a metalto-metal point contact was used in 1968 by Ali Javan and co-workers for frequency mixing of infrared radiation (Hocker, 1968). The rectification efficiency of these whisker diodes could be increased by suitably kinking or bending the wire antenna (Matarrese and Evenson, 1970). The length L (tip to kink) had to be adjusted in relation to the wavelength and angle of incidence, and the strongest response was obtained for the fundamental resonance of $L \approx \lambda/2.7$ (Rothammel, 2001). These experiments were performed at infrared wavelengths of $\lambda = 10...337 \mu m$ where metals are good conductors. Whisker wires are routinely employed for delivering electromagnetic energy to miniature semiconductor circuits such as diodes or field effect transistors. Similar frequency mixing experiments have later been performed by Wolfgang Krieger *et al.* at the tunnel junction of an STM (Krieger, 1990).

In essence, an antenna efficiently converts the energy of free-propagating radiation to localized energy, and vice versa. For example, the antenna of a cell phone is used to concentrate the energy of incoming radiation onto a receiver chip with dimensions much smaller than the wavelength of the incoming radiation. In the context of microscopy, an optical antenna basically replaces a conventional focusing lens (objective), thereby concentrating external laser radiation to dimensions much smaller than the diffraction limit. The controlled and reproducible development of efficient optical antenna designs will provide new applications and opportunities not only in near-field optical microscopy but also in sensing applications and in optical device architectures.

Due to reciprocity a good transmitting antenna is also a good receiving antenna. However, most radiowave or microwave antennas are employed only in one mode or the other. In near-field optical microscopy, on the other hand, the near-field probe can act as both a receiving antenna for localizing optical energy and a transmitting antenna for emitting



Figure 14: Adaptation of the $\lambda/2$ antenna to the optical regime. (a) Resonant field distribution near a 220nm gold rod ($\lambda = 1250$ nm, thickness 20nm). The curve under the figure depicts the charge distribution at an instant of time. (b) Spectral dependence of the intensity enhancement at the extremities of a 220nm gold rod. (c) Resonant field distribution near a 220nm rod made of an ideal conductor ($\lambda = 520$ nm, thickness 20nm). (d) Spectral dependence of the intensity enhancement at the extremities of a 220nm ideally conducting rod. The resonance deviates from $\lambda/2$ because of the finite thickness of the rod. (a,c) The contourlines are displayed with a logarithmic scaling (factor of 2 between successive lines).

the optical response. Classical antenna design assumes that there is no time lag between the conduction electrons and the exterior field. Consequently, electromagnetic penetration into the metal (skin effect) is negligible. At optical frequencies, on the other hand, the 'inertia' of conduction electrons causes electromagnetic fields to penetrate into the metals and antenna design is no longer a pure function of the the 'exterior' wavelength of the fields. Therefore, antenna designs cannot be directly downscaled from the microwave to the optical regime (cf. Fig. 14) and a scaled (effective) wavelength has to be invoked instead (Novotny, 2007). Existing antenna concepts can provide the necessary inspiration for their optical counterparts. For example, the self-similar antenna proposed by Mark Stockman and co-workers (Li, 2003) has similarities with the well-established Yagi-Uda antenna developed in the 1920s in Japan and today widely used as receiving antenna in the UHF/VHF region. As an illustration, Fig. 14 shows a calculation of the resonance of a $\lambda/2$ antenna. Because of surface plasmon resonances, the response at optical frequencies is very strong but the resonance is no longer at $\lambda/2$. Instead, the resonance wavelength is determined both by the external irradiation and the dielectric properties of the material.

Pointed metal structures were used for the localization of electromagnetic energy already in the 18th century, long before the era of antenna design began. The most prominent invention making use of this property is Benjamin Franklin's lightning rod (Krider, 2006). It can be viewed as a sharply pointed electrode with a strongly enhanced static field at its apex. This field originates from the potential difference between the tip's support (ground) and a counter electrode (atmosphere). For strong enough fields the tip initiates a plasma channel (discharge) and conducts electrons into ground thereby low-



Figure 15: Sketch from Benjamin Franklin's letter sent to Peter Collinson on 29 July 1750. The contour defined by A, B, C, D, E represents an electrified body and the dashed lines around it indicate an "Atmosphere of Electrical Particles". See text for details. From (Labaree, 1961b).

ering the initial potential difference. In a letter sent to Peter Collinson, a fellow of the Royal Society in London, on 25 May 1747, Benjamin Franklin writes ".. In pursuing our Electrical Enquiries, we had observ'd some particular Phenomena, ..." and then adds "The first is the wonderful Effect of Points both in drawing off and throwing off the Electrical Fire" (Labaree, 1961). If the 'Fire' to which Franklin is referring to were associated with 'Radiation' we could accept his phrase as the definition of an antenna, namely a device that attracts or sends off electrical radiation. The sketch shown in Fig. 15 has been depicted by Franklin in a letter to Collinson, dated 29 July 1750 (Labaree, 1961b). The contour defined by A, B, C, D, E represents an electrified body and the dashed lines around it indicate an "Atmosphere of Electrical Particles", i.e. the electric field. Franklin argues that the Atmosphere to the right of L,C,M has the least surface to rest on and hence the attraction at the point C is weakest. As a consequence, the particles expand easiest at point C. He concludes "On these Accounts, we suppose Electrified Bodies discharge their Atmospheres upon unelectrified Bodies more easily, and at greater Distance, from their Angles and Points, than from their smoothe Sides." Franklin adds "These Points will also discharge into the Air, when the Body has too great an electrical Atmosphere ... " In the same letter, Franklin speculates that the Fires of Electricity and Lightning are the same and he proposes the construction of grounded lightning rods to be fixed on buildings for the protection against lightning. He also writes "To determine the Question, Whether the Clouds that contain Lightning are electrified or not, I would propose an Experiment to be try'd where it may be done conveniently." His following proposal inspired the famous 'sentry-box' experiment performed at Mary-la-Ville, France, in 1752 by Dalibard and Delors (Krider, 2006).

It might appear to be a far stretch from Franklin's lightning rod to near-field optical microscopy but, similar to an antenna or a near-field probe, the efficiency of a lightning rod is characterized by its ability to concentrate and localize electric fields (Denk and Pohl, 1991). However, despite this apparent similarity a lightning rod is not an antenna. The lightning rod is a static device whose properties are dictated by the Laplace equation. Consequently, the surface of the lightning rod is an equipotential surface and the geometrical singularity at the tip apex gives rise to a singular field, irrespective of the shape of the shaft or wire on which the tip is mounted. On the other hand, an antenna interacts with electromagnetic radiation and hence its behavior is dictated by the wave equation. Because of the finite penetration of optical radiation into metals the quasi-static approximation is only valid for structures smaller than the skin depth but such small structures are not the most efficient antennas. Therefore, inspiration for efficient optical near-field probes has to be drawn from antenna theory despite the fact that a direct downscaling of traditional antenna designs to optical wavelengths is not possible.

11 Concluding Remarks

This article provides an overview of discoveries and achievements which were influential to the shaping of near-field optics. Important achievements cannot always be accredited to a single person or a single group because different developments progressed in parallel. I would like to emphasize that the history presented in this article cannot be complete because all resources are finite. I hope no important details have been left out. Any omissions are solely due to my personal ignorance.

As is evident from this article the roots of near-field optics are not well defined and the shaping of the field depended on many developments. The modern interest has its origins in the 1982 experiments by Pohl and co-workers and in parallel developments by other groups. Near-field optics received important inspiration from the early work in nanoplasmonics which was mainly aimed at answering open questions in SERS, and from studies of energy transfer. The prophetic papers and letters of Synge emerged at a later stage. From todays perspective, near-field optics has a lot in common with classical antenna theory extended into the optical regime and this is where it connects to the current second wave of nanoplasmonics. Interestingly, the first experimental developments in near-field optical microscopy were concentrated on the idea of using an aperture as a means to further confine a diffraction-limited focus and this mindset delayed experimental work on antenna-inspired concepts. Today, Synge's original particle (tip) idea finds applications in various studies and, despite Einstein's initial skepticism, it is likely to be adopted in future commercial instruments.

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