PRINCIPLES
OF
NANO-OPTICS

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TO OUR FAMILIES
(JESSICA, LEONORE, JAKOB, DAVID, NADJA, JAN)

.. it was almost worth the climb *(B.B. Goldberg)*

PREFACE

Why should we care about nano-optics? For the same reason as we care for optics! The foundations of many fields of the contemporary sciences have been established using optical experiments. To give an example, think of quantum mechanics. Blackbody radiation, hydrogen lines, or the photoelectric effect were key experiments that nurtured the quantum idea. Today, optical spectroscopy is a powerful means to identify the atomic and chemical structure of different materials. The power of optics is based on the simple fact that the energy of light quanta lies in the energy range of electronic and vibrational transitions in matter. This fact is at the core of our abilities for visual perception and is the reason why experiments with light are very close to our intuition. Optics, and in particular optical imaging, helps us to consciously and logically connect complicated concepts. Therefore, pushing optical interactions to the nanometer scale opens up new perspectives, properties and phenomena in the emerging century of the nano-world.

Nano-Optics aims at the understanding of optical phenomena on the nanometer scale, i.e. near or beyond the diffraction limit of light. It is an emerging new field of study, motivated by the rapid advance of nanoscience and nanotechnology and by its need for adequate tools and strategies for fabrication, manipulation and characterization at the nanometer scale. Interestingly, nano-optics even predates the trend of nanotechnology by more than a decade. An optical counterpart to the scanning tunneling microscope (STM) was demonstrated in 1984 and optical resolutions had been achieved that were significantly beyond the diffraction limit of light. These early experiments sparked a field initially called *near-field optics* since it was realized quickly that the inclusion of near fields in the problem of optical imaging and associated spectroscopies holds promise for achieving arbitrary spatial resolutions thus providing access for optical experiments on the nanometer scale.

The first conference on near-field optics was held in 1992. About seventy participants discussed theoretical aspects and experimental challenges associated with near-field optics and near-field optical microscopy. The following years, are characterized by a constant refinement of experimental techniques, as well as the introduction of new concepts and applications. Applications of near-field optics soon covered a large span ranging from fundamental physics, and materials science to biology and medicine. Following a logical development the strong interest in near-field optics gave birth to the fields of *single-molecule spectroscopy* and *plasmonics*, and inspired new theoretical work associated with the nature of optical near-fields. In parallel, relying on the momentum of the flowering nanosciences, researchers started to tailor nanomaterials with novel optical properties. Photonic crystals, single-photon sources, and optical microcavities are products of this effort. Today, elements of nano-optics are scattered across the disciplines. Various review articles and books capture the state-of-the-art in the different subfields but there appears to be no dedicated textbook that introduces the reader to the general theme of nano-optics.

This textbook is intended to teach students at the graduate level or advanced undergraduate level about the elements of nano-optics encountered in different subfields. The book evolved from lecture notes that have been the basis for courses on *Nano-Optics* taught at the Institute of Optics of the University of Rochester, and at the University of Basel. We were happy to see that students from many different departments found interest in this course which shows that nano-optics is important to many fields of study. Not all students were interested in the same topics and depending on their field of study, some students needed additional help with mathematical concepts. The courses were supplemented with laboratory projects that were carried out in groups of two to three students. Each team picked the project that had most affinity with their interest. Among the projects were: surface enhanced Raman scattering, photon scanning tunneling microscopy, nanosphere lithography, spectroscopy of single quantum dots, optical tweezers, and others. Towards the end of the course, students gave a presentation on their projects and handed in a written report. Most of the problems at the end of individual chapters have been solved by students as homework problems or take-home exams. We wish to acknowledge the very helpful input and inspiration that we received from many students. Their interest and engagement in this course is a significant contribution to this textbook.

Nano-optics is an active and evolving field. Every time the course was taught new topics have been added. Also, nano-optics is a field that easily overlaps with other fields such as physical optics or quantum optics, and thus the boundaries cannot be clearly defined. This first edition is an initial attempt to put a frame around the field of nano-optics. We would be grateful to receive input from our readers related to corrections and extensions of existing chapters and for suggestions of new topics.

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Chapter 1

Introduction

In the history of science, the first applications of optical microscopes and telescopes to investigate nature mark the beginning of new eras. Galileo Galilei used a telescope to see for the first time craters and mountains on a celestial body, the moon, and also discovered the four largest satellites of Jupiter. With this he opened the field of Astronomy. Robert Hooke and Antony van Leeuwenhoek used early optical microscopes to observe certain features of plant tissue that were called "cells", and to observe microscopic organisms, like bacteria and protozoans, thus marking the beginning of biology. The newly developed instrumentation enabled the observation of fascinating phenomena not directly accessible to human senses. Naturally, the question was raised whether the observed structures not detectable within the range of normal vision should be accepted as reality at all. Today, we have accepted that in modern physics, scientific proofs are verified by indirect measurements, and that the underlying laws have often been established on the basis of indirect observations. It seems that as modern science progresses it withholds more and more findings from our natural senses. In this context, the use of optical instrumentation excels among other ways to study nature. This is due to the fact that because of our ability to perceive electromagnetic waves at optical frequencies our brain is used to the interpretation of phenomena associated with light, even if the structures that are observed are magnified thousandfold. This intuitive understanding is among the most important features that make light and optical processes so attractive as a means to reveal physical laws and relationships. The fact that the energy of light lies in the energy range of electronic and vibrational transitions in matter allows us to use light for gaining unique information about the structural and dynamical properties of matter and also to perform subtle manipulations of the quantum state of matter. These unique spectroscopic capabilities associated with optical techniques are of great importance for the study of biological and solid-state nanostructures.

Today we encounter a strong trend towards nanoscience and nanotechnology. This trend originally was driven by the benefits of miniaturization and integration of electronic circuits for the computer industry. More recently a shift of paradigms is observed that manifests itself in the notion that nanoscience and technology are more and more driven by the fact that as we move to smaller and smaller scales, new physical effects become prominent that may be exploited in future technological applications. The recent rapid advances in nanoscience and technology are due in large part to our newly acquired ability to measure, fabricate and manipulate individual structures on the nanometerscale using scanning probe techniques, optical tweezers, high-resolution electron microscopes and lithography tools, focused-ion beam milling systems and others.

The increasing trend towards nanoscience and nanotechnology makes it inevitable to address the question of the possibility of optics on the nanometer scale. Since the diffraction limit does not allow us to focus light to dimensions smaller than roughly one half of the wavelength (200 nm), traditionally it was not possible to optically interact selectively with nanoscale features. However, in more recent years, several new approaches have been put forth to 'shrink' the diffraction limit (confocal microscopy) or to even overcome it (near-field microscopy). A central goal of "nano-optics" is to extend the use of optical techniques to length scales beyond the diffraction limit. The most obvious potential technological applications that arise form breaking the diffraction barrier are super-resolution microscopy and ultra-high-density data storage. But the field of nano-optics is by no means limited to technological applications and instrument design. Nano-optics also opens new doors to basic research on nanometer sized structures.

Nature has developed various nanoscale structures to bring out unique optical effects. A prominent example are photosynthetic membranes which use light-harvesting proteins to absorb sunlight and then channel the excitation energy to other neighbor-

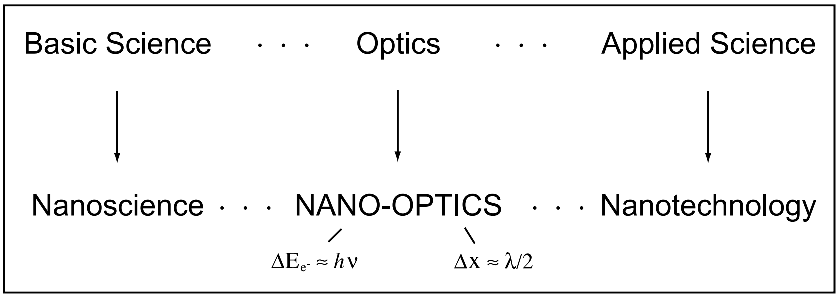

Figure 1.1: Nano-optics as part of nanoscience and nanotechnology. While the major advantage of nano-optics are the spectroscopic capabilities ($\Delta E = h\nu$), the challenge is to find ways to surpass the diffraction limit ($\Delta x = \lambda/2$).

ing proteins. The energy is guided to a so-called reaction center where it initiates a charge transfer across the cell membrane. Other examples are sophisticated diffractive structures used by insects (butterflies) and other animals (peacock) to produce attractive colors and effects. Also, nanoscale structures are used as anti-reflection coatings in the retina of various insects and naturally occurring photonic bandgaps are encountered in gemstones (opals). In recent years, we succeeded in creating different artificial nanophotonic structures. A few examples are depicted in Fig. 1.2. Single molecules are being used as local probes for electromagnetic fields and for biophysical processes, resonant metal nanostructures are being exploited as sensor devices, localized photon sources are being developed for high-resolution optical microscopy, extremely high Q-factors are being generated with optical microdisk resonators, nanocomposite materials are being explored for generating increased nonlinearities and collective responses, microcavities are being built for single-photon sources, surface plasmon waveguides are being implemented for planar optical networks, and photonic bandgap materials are being developed to suppress light propagation in specific frequency windows. All of these nanophotonic structures are being created to create unique optical properties and phenomena and it is the scope of this book to establish a basis for their understanding.

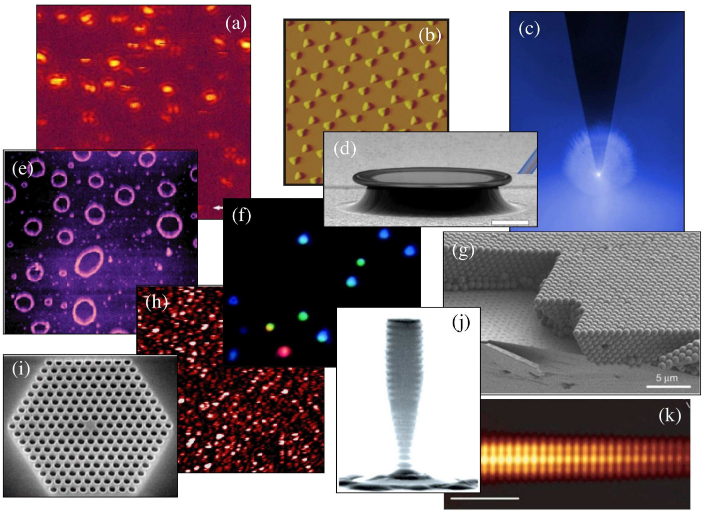

Figure 1.2: Potpourri of man-made nanophotonic structures. (a) Strongly fluorescent molecules, (b) metal nanostructures fabricated by nanosphere lithography (from [1]), (c) localized photon sources, (d) microdisk resonators (from [2]), (e) semiconductor nanostructures, (f) particle plasmons (from [3]), (g) photonic bandgap crystals (from [4]), (h) nanocomposite materials, (i) laser microcavities(from [5]), (j) single photon sources (from [6]), (k) surface plasmon waveguides (from [7]).

1.1 Nano-optics in a nut shell

Let us try to get a quick glimpse at the very basics of nano-optics just to show that optics at the scale of a few nanometers makes perfect sense and is not forbidden by any fundamental law. In free space, the propagation of light is determined by the dispersion relation $\hbar k = c \cdot \hbar\omega$ which connects the wave vector k of a photon with its angular frequency ω via the speed of propagation c. Heisenberg's uncertainty relation states that the product of the uncertainty in the spatial position of a microscopic particle in a certain direction and the uncertainty in the component of its momentum in the same direction cannot become smaller than $\hbar$. For photons this leads to the relation

$$\Delta\hbar k_{\mathrm{x}} \cdot \Delta x \geq \hbar \tag{1.1}$$

which can be rewritten as

$$\Delta x \geq \frac{1}{\Delta k_{\mathrm{x}}} \ . \tag{1.2}$$

The interpretation of this result is as follows: The spatial confinement that can be achieved for photons is inversely proportional to the spread in the magnitude of wave vector components in the respective spatial direction. Such a spread in wave vector components occurs e.g. in a light field that converges towards a focus, e.g. behind a lens. The maximum possible spread in the wave vector component is the total length of the free-space wave vector k=$2\pi/\lambda$.* This leads to

$$\Delta x \geq \frac{\lambda}{2\pi} \tag{1.3}$$

which is very similar to the well-known expression for the Rayleigh diffraction limit. Note that the spatial confinement that can be achieved is only limited by the spread of wave vector components in a given direction. In order to increase the spread of wave vector components we can play a mathematical trick: If we choose two arbitrary perpendicular directions in space, e.g. x and z, we can increase one wave vector component to values beyond the total wave vector while at the same time requiring the wave vector in the perpendicular direction to become purely imaginary. If this is the case, then we can still fulfil the requirement for the total length of the wave vector is still $2\pi/\lambda$. If we chose to increase the wave vector in the x-direction then also the possible range of wave vectors in this direction is increased and the confinement of light is no longer limited by Eq. (1.3). However, the possibility of increased confinement has to be payed for and the currency is a confinement also in the z-direction resulting from the purely imaginary wave vector component in this direction that is necessary to compensate for the large wave vector component in the x-direction.

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[*] For a real lens this must be corrected by the numerical aperture.

When introducing the purely imaginary wave vector component into the expression for a plane wave we obtain $\exp(ik_z z)$=$\exp(-|k_z|z)$. In one direction this leads to an exponentially decaying field, an evanescent wave, while in the opposite direction the field is exponentially increasing. Since exponentially increasing fields have no physical meaning we may safely discard the just outlined strategy to obtain a solution and state that in free space Eq. (1.3) is always valid. However, this argument only holds for the infinite free space! If we divide our infinite free space into at least two half spaces with different refractive indices, then the exponentially decaying field in one half space can exist without needing the exponentially increasing counterpart in the other half space. In the other halfspace a different solution may be valid that fulfils the boundary conditions for the fields at the interface.

These simple arguments show, that in presence of an inhomogeneity in space the Rayleigh limit for the confinement of light is no longer strictly valid, but in principle infinite confinement of light becomes, at least theoretically, possible. This insight is the basis of nano-optics. One of the key questions in nano-optics is how material structures have to be shaped to actually realize the theoretically possible field confinement. Another key issue is the question of what are the physical consequences of the presence of exponentially decaying and strongly confined fields which we will discuss in some detail in the following chapters.

1.2 Historical survey

In order to put this text on nano-optics into the right perspective and context we deem it appropriate to start out with a very short introduction to the historical development of Optics in general and the advent of Nano-Optics in particular.

Nano-optics builds on achievements of classical optics, the origin of which goes back to antiquity. At that time, burning glasses and the reflection law were already known and Greek philosophers (Empedocles, Euclid) speculated about the nature of light. They were the first to do systematical studies on optics. In the 13th century the first magnifying glasses were used. There are documents reporting the existence of eye glasses in China several centuries before. However, the first optical instrumentation for scientific purposes was not built before the beginning of the 17th century, when modern human curiosity started to awake. It is often stated that the earliest telescope was the one constructed by Galileo Galilei in 1609, as there is definite knowledge of its existence. Likewise, the first prototype of an optical microscope (1610) is also attributed to Galilei [8]. However, it is known that Galilei knew from a telescope built in Holland (probably by Zacharias Janssen) and that his instrument was built according to existing plans. The same uncertainty holds for the first microscope. Already in the 16th century craftsmen used glass spheres filled with water for the magnification of small details. As in the case of the telescope, the development of the microscope extends over a considerable period and cannot be attributed to one single inventor. A pioneer who advanced the development of the microscope as already mentioned was Antony van Leeuwenhoek. It is remarkable that the resolution of his microscope , built in 1671, was not exceeded for more than a century. At the time being, his observation of red blood cells and bacteria was revolutionary and discussed controversially. In the 18th and 19th century the development of the theory of light (polarization, diffraction, dispersion) helped to significantly advance the optical technology and instrumentation. It was soon realized that optical resolution cannot be improved arbitrarily and that a lower bound was set by the diffraction limit. The theory of resolution was formulated by Abbe in 1873 [9] and Rayleigh in 1879 [10]. It is interesting to note, as we saw above, that there is a close relation to Heisenberg's uncertainty principle. Different techniques such as confocal microscopy [11] were invented over the years in order to stretch the diffraction limit beyond Abbe's limit. Today, confocal fluorescence microscopy is a key technology in biomedical research [12]. Highly fluorescent molecules have been synthesized that can be specifically attached to biological entities such as lipids, muscle fibers, and various cell organelles. This chemically specific labelling and the associated discrimination of different dyes based on their fluorescence emission allows scientists to visualize the interior of cells and study biochemical reactions in live environments. The invention of pulsed laser radiation propelled the field of nonlinear optics and enabled the invention of multiphoton microscopy which is slowly replacing linear confocal fluorescence microscopy [13]. However, multiphoton excitation is not the only nonlinear interaction that is exploited in optical microscopy. Second harmonic, third harmonic, and coherent anti-Stokes Raman scattering (CARS) microscopy[14] are other examples of extremely important inventions for visualizing processes with high spatial resolution. Besides nonlinear interactions, it has also been demonstrated that saturation effects can, in principle, be applied to achieve arbitrary spatial resolutions provided that one knows what molecules are being imaged [15].

A different approach for boosting spatial resolution in optical imaging is provided by near-field optical microscopy. In principle, this technique does not rely on prior information. While it is restricted to imaging of features near the surface of a sample it provides complimentary information about the surface topology similar to atomic force microscopy. A challenge in near-field optical microscopy is posed by the coupling of source (or detector) and the sample to be imaged. This challenge is absent in standard light microscopy where the light source (e.g. the laser) is not affected by the properties of the sample. Near-field optical microscopy was originally proposed in 1928 by Synge. In a prophetic article he proposed an apparatus which comes very close to present implementations in scanning near-field optical microscopy [16]. A minute aperture in an opaque plate illuminated from one side is placed in close proximity to a sample surface thereby creating an illumination spot not limited by diffraction. The transmitted light is then collected with a microscope, and its intensity is measured with a photo-electric cell. In order to establish an image of the sample, the aperture is moved in small increments over the surface. The resolution of such an image should be limited by the size of the aperture and not by the wavelength of the illuminating light as Synge stated correctly. It is known that Synge was in contact with Einstein about his ideas and Einstein encouraged Synge to publish his ideas. It is also known that Synge later in his life was no longer convinced about his idea and proposed alternative, however as we know today, incorrect ideas. Anyway, due to the obvious experimental limitations at that time, Synge's idea was not realized and forgotten soon. Later, in 1956, O'Keefe proposed a similar set-up without knowing of Synge's visionary idea [17]. The first experimental realization in the microwave region was performed in 1972 by Ash and Nichols, again without knowledge of Synge's paper [18]. Using a $1.5\,mm$ aperture, illuminated with $10\,cm$ waves, Ash and Nichols demonstrated subwavelength imaging with a resolution of $\lambda/60$.

The invention of scanning probe microscopy [19] at the beginning of the 1980's enabled distance regulation between a probe and a sample with high precision, and hence set the ground for a realization of Synge's idea at optical frequencies. In 1984 Massey proposed the use of piezoelectric position control for the accurate positioning of a minute aperture illuminated at optical frequencies [20]. Shortly later, Pohl, Denk and Lanz at the IBM Rüschlikon Research Laboratory managed to solve the remaining experimental difficulties of producing a subwavelength sized aperture: a metal coated pointed quartz tip was 'pounded' against the sample surface until some light leakage through the foremost end could be detected. In 1984 the IBM group presented the first subwavelength images at optical frequencies [21] and almost simultaneously, an independent development was realized by Lewis et al. [22]. Subsequently, the technique was systematically advanced and extended to various applications mainly by Betzig et al. who showed subwavelength magnetic data storage and detection of single fluorescent molecules [24, 25]. Over the years, various related techniques were proposed, such as the photon scanning tunneling microscope, the near-field reflection microscope, microscopes using luminescent centers as light emitting sources, microscopes based on local plasmon interaction, microscopes based on local light scattering, and microscopes relying on the field enhancement effect near sharply pointed metal tips. All these techniques have in common that they provide a confined photon flux between probe and sample. However, the confined light flux is not the only limiting factor for the achievable resolution. In order to be detectable, the photon flux needs to have a minimum intensity. These two requirements are in some extent contradictory and a compromise between light confinement and light throughput has to be found.

1.3 Scope of the book

Traditionally, the field of Optics is part of both, the basic sciences (e.g. quantum optics) and applied sciences (e.g. microscopy and optical communication and computing). Therefore, as illustrated in Fig. 1.1 nano-optics can be defined as the broad spectrum of optics on the nanometer ranging from nanotechnology applications to fundamental nanoscience.

On the nanotechnology side, we find topics like nanolithography, high-resolution optical microscopy, and high-density optical data storage. On the basic science end, we might mention atom-photon interactions in the optical near-field and their potential applications for atom trapping and manipulation experiments. Compared with free propagating light the optical near-field is enriched by so-called virtual photons that correspond to the exponentially decaying fields introduced before. The virtual-photon picture can be used to describe local, non-propagating fields in general. Theses virtual photons are the same sort of particles that are also responsible for molecular binding (van der Waals and Casimir forces) and therefore promise to have a potential for selective probing of molecular scale structures. The consideration of virtual photons in the field of quantum optics will enlarge the range of fundamental experiments and will result in new applications. The present book provides an introduction to nano-optics that reflects the full breadth of the field between applied and basic science that is summarized in Fig. 1.3.

We start out by providing an overview of the theoretical foundations of nano-optics. Maxwell's equations being scale invariant provide a secure basis also for

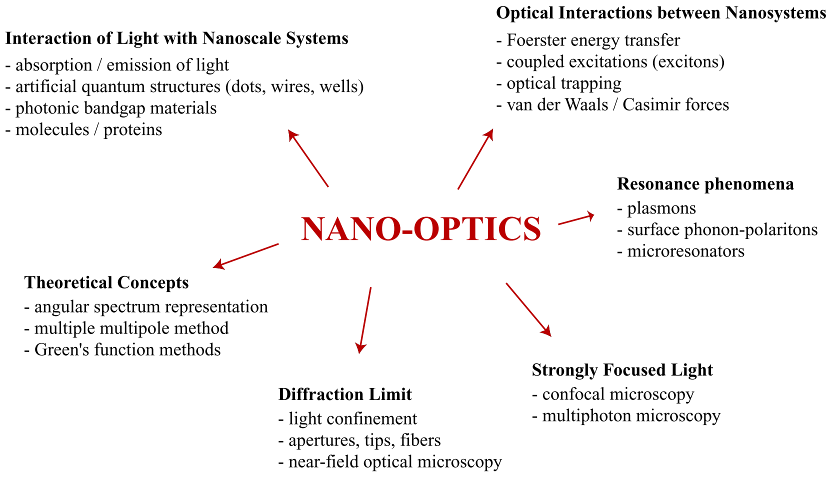

Figure 1.3: Constituents of the field of Nano-Optics.

nano-optics. Since optical near-fields are always associated with matter, we review constitutive relations and complex dielectric constants. The systems that are investigated in the context of nano-optics, as we saw, must separate into several regions that are divided by boundaries. Representations of Maxwell's equations valid in piecewise homogeneous media and the related boundary conditions for the fields are therefore derived. We then proceed with the discussion of fundamental theoretical concepts, such as the Green's function and the angular spectrum representation, that are particularly useful for the discussion of nano-optical phenomena. The treatment of the angular spectrum representation leads us to thorough discussion of evanescent waves which correspond to the new virtual photon modes just mentioned.

Light confinement is a key issue in nano-optics. To set the ground for further discussions, in chapter3, we analyze what is the smallest possible confinement of light that can be achieved by classical means, i.e microscope objectives and other high numerical aperture focusing optics. Starting out with the treatment of focused fields in the paraxial approximation which yields the well-known Gaussian beams, we proceed by discussing focused fields beyond the paraxial approximation as they occur for example in modern confocal microscopes.

Speaking of microscopy, spatial resolution is a key issue. Several definitions of the spatial resolution of an optical microscope exist that are related to the diffraction limit. An analysis of their physical foundations in chapter 4 leads us to the discussion of methods that can be used to enhance the spatial resolution of optical microscopy. Saturation effects and the difference between spatial position accuracy and resolution are discussed.

The following three chapters then deal with more practical aspects of nano-optics related to applications in the context of near-field optical microscopy. In chapter 5 we discuss the basic technical realizations of nano-optical microscopes starting with confocal microscopy, and proceeding with various near-field techniques that have been developed over the time. Chapter 6 then deals with the central technical question of how light can be squeezed into subwavelength regions. This is the domain of the so-called optical probes, material structures that typically have the shape of pointed tips and exhibit a confined and enhanced optical field at their apex. Finally, to complete the technical section, we show how such delicate optical probes can be approached and scanned in close proximity to a sample surface of interest. A method relying on the measurement of interaction (shear) forces between probe and sample is introduced and discussed. Taken together, the three chapters provide the technical basics for the understanding of the current methods used in scanning near-field optical microscopy.

We then proceed with the discussion of more fundamental aspects of nano-optics, i.e. light emission and optical interactions in nanoscale environments. As a starting point, we show that the light emission of a small particle with an electronic transition can be treated in the dipole approximation. We discuss the resulting fields of a radiating dipole and its interactions with the electromagnetic field in some detail. We proceed with the discussion of spontaneous decay in complex nano environments which in the ultimate limit leads us to the discussion of dipole-dipole interactions, energy transfer and excitonic coupling.

Having discussed dipolar emitters without mentioning a real-world realization we discuss in chapter 9 some experimental aspects of the detection of single quantum emitters such as e.g. single fluorescent molecules and semiconductor quantum dots. Saturation count rates and the solutions of rate equation systems are discussed as well as fascinating issues such as the nonclassical photon statistics of fields emitted by quantum emitters and coherent control of wave functions. Finally we discuss how single emitters can be used to map nano-optical fields in great detail.

In chapter 10 we pick up again the issue of dipole emission in a nanoscale environment. Here, we treat in some detail the very important and illustrative case of dipole emission near a planar interface. We calculate radiation patterns and decay rates of dipolar emitters and also discuss the image-dipole approximation that can be used to obtain approximate results.

If we consider multiple interfaces instead of only one, that are arranged in a regular pattern, we obtain a so called photonic crystal. The properties of such structures can be described in analogy to solid-state physics by introducing an optical band structure that may contain band gaps in certain directions where propagating light can not exist. Defects in photonic crystals lead to localized states, much similar as for their solid-state counterparts, which are of particular interest in nano-optics since they can be considered as microscopic cavities with very high quality factors.

Chapter 12 then takes up the topic of surface plasmons. Resonant collective oscillations of the free surface charge density in metal structures of various geometries can couple efficiently to optical fields and, due to the occurrence of resonances, are associated with strongly enhanced and confined optical near fields. We give a basic introduction to the topic covering the optical properties of noble metals, thin film plasmons, and particle plasmons.

In the following chapter we discuss optical forces occurring in confined fields. We formulate a theory based on Maxwell's stress tensor that allows to calculate forces of particles of arbitrary shape once the field distribution is known. We then specialize the discussion and introduce the dipole approximation valid for small particles. Practical applications discussed include the optical tweezer principle. Finally, the transfer of angular momentum using optical fields is discussed, as well as forces exerted by optical near-fields.

Another type of forces is discussed in the subsequent chapter, i.e. forces that are related to fluctuating electromagnetic fields which include the Casimir-Polder force and electromagnetic friction. On the way we also discuss the emission of radiation by fluctuating sources.

The current textbook is concluded by a summary of theoretical methods used in the field of nano-optics. Hardly any prediction can be made in the field of nano-optics without using adequate numerical methods. A selection of the most powerful theoretical tools is presented and their advantages and drawbacks are discussed.

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