Introduction

Electrodynamic fields in the optical frequency range in the vicinity of nanometer-sized material structures exhibit many unusual properties, especially what the near-field of such structures concerns. Experimental and theoretical efforts to understand these properties have multiplied in recent years. The discipline has, however, still not reached a state, where merely relatively unimportant details are understood, as will, hopefully, become evident also from this thesis. In this work model systems have been investigated, the analysis of which is basic for more complex systems.

The origins of an optics of material structures in the nanometer size regime are connected with the theoretical work of Mie[1] on the scattering of light by small particles as well as with the work of Bethe[2] and Bouwkamp[3] on the transmission of small apertures in ideally conducting screens. The idea of a near-field optical instrument is based on the early ideas of Synge.[4,5] In this publications he has described in an astonishingly accurate manner a microscope achieving a resolution beyond Abbe's limit. His basic idea was the design of a subwavelength aperture scanning in the near-field of a sample. The proposed device anticipated the scanning near-field optical microscope (SNOM), which was realized several decades later. Many proposed features, such as piezoelectric control, raster-scanned images, and sharpened glass needle probes with an opaque vacuum-evaporated coating to confine the optical field are used in present-day near-field optical microscopes. Ash and Nicholls[6] were the first to achieve subwavelength imaging, in the microwave range, employing the near field of a subwavelength aperture. Important advances began with the development of the scanning tunneling microscope[7], which opened the way for many other types of raster scanning microscpes, such as the atomic force[8] and near-field optical microscopes (SNOM). Subwavelength resolution in the optical frequency range was demonstrated for the first time by Pohl[9] and independently from him by Betzig, who introduced sharpened glass fiber tips as near-field optical probes[10] and the shear-force distance regulation mechanism[11]. These advances in instrumentation gave rise to an intense discussion of all aspects of nano-optics, which has continued to the present day[12,13]. Reviews of near-field optical microcopy can be found, e.g., in references [14,15]. The proceedings of the annual conferences on near-field optics provide an overview of the present state of the art of the field[16,17].

Topics still in debate today and partly covered in this thesis include the peculiarities of the scattering of evanescent waves by small particles and multiple scattering processes at the material surface (compare chapter III), the contribution of nonlocal effects in nano-optics (compare chapter V), the absorption and the scattering of light by aggregates of metallic nano-particles (compare again chapter V), the physical basis of an apertureless version of scanning near-field optical microscopy (compare chapter VI), the participation of surface plasmon polaritons in the optical properties of metallic nano-structures (compare chapter III, V and VII), the optical properties of defects in photonic bandgap materials (compare chapter VIII), the field distribution in and the tunneling through the metal-coated tapered glass fiber of a scanning near-field optical microscope, as well as the intensity distribution in the vicinity of the aperture (compare chapter VII). An example for an engineering application of nano-optics is laser cleaning[18] and laser nanostructuring[19] of surfaces (compare chapter IV).

A major distinction of nanooptics from the conventional optics as described in textbooks like the one of Born and Wolf[20] lies in the consideration of evanescent fields in the vicinity of material structures in the nanometer size regime. These fields carry subwavelength information. Furthermore, in many cases, they can be resonantly enhanced, e.g. in the case of propagating and localized surface excitations of metallic structures close to the plasma frequency.

Based on their dispersion relations, the resonant properties of surface plasmon polaritons (SPP's) can significantly alter the near-field and far-field properties of metallic structures. Furthermore, it is possible to construct photonic band gaps for plasmonic surface modes in the visible by a modulated refractive index or a textured interface[21]. On the other hand, not only metals but also dielectric structures can exhibit interesting properties[22]. For example, it is possible to excite in spherical dielectric particles so-called whispering-gallery modes due to constructive interference of circumpropagating waves, which results in very strong enhancement factors due to the high Q values of these modes. The resonant coupling of whispering-gallery modes in an aggregate of two such particles, a so-called photonic molecule has recently been examined experimentally[23] (compare chapter III). Regular arrays of spherical dielectric particles (compare again chapter III) exhibit photonic band gaps, which allow the construction of lasers with ultralow thresholds, if active defects are incorporated within such photonic crystals (compare chapter VIII). A two-dimensional defect mode laser based on a two-dimensional band gap has been recently presented[24,25,26,27]. A number of groups have investigated the cavity quantum electrodynamics of quantum dots in 3D-microcavities[28,29]. An interesting technical application is the use of 3D-microcavities doped with quantum dots in concepts for quantum computing[30]. The theoretical investigated model system is based on a set of several atoms representing the quantum bits communicating via their interaction with a single quantized mode of an optical cavity. The atoms, fixed in the cavity are assumed to interact individually with laser beams.

Cavity-based enhancements in dielectric structures and plasmon-derived field enhancements may, of course, be combined in appropriate designed composites of dielectrics and metals. An example is the preparation of spherical metal-coated particles, which exhibit strong resonances and which can be spectrally shifted by a variation of core size and shell thickness (compare chapter III).

Within the present working, principles of different types of near-field optical microscopes have been investigated. Generalizations of Mie's theory are applicable to the examination of apertureless near-field optical microscopes (compare chapter VI). A report about the successful realization of an apertureless near-field optical microscope with ultrahigh resolution was given by Zenhausern et al. in 1995[31]. This instrument was based on the modulation of the electric field scattered by a vibrating probe tip in the vicinity of a sample. For this device a resolution of 3 nm was reported, comparable to what can be achieved with typical attractive mode atomic force microscopy. The resolution of the microscope in the vertical (z) direction was enhanced by interferometric means up to 10 Ångstrom[32]. Later on optical spectroscopy was performed and images of viruses and molecules were presented[33]. The results could recently be confirmed by another group[34], employing a similar device as described in ref. The corresponding experiments have led to a further reduction of the unwanted scattering background. Moreover, a drastic artifact[35] reduction was claimed.

In the present work, the imaging properties of apertureless microscopes are investigated theoretically (see chapter VI). The tip of the microscope is simplified as a scanning spherical silicon particle, which is interacting with an aggregate of metallic and dielectric spherical particles. The image properties of the metallic particles are studied via aggregate Mie theory as a function of frequency, especially in the range of the SPP resonance of the metallic particles[36]. The investigation includes the consideration of nonlocal effects (compare chapter V).

A very simple example of an apertureless near-field optical imaging technique is the total internal reflection microscope (TIRM)[37]. If colloidal particles are deposited on the hypotenuse of a glass prism, where an evanescent wave is generated, the exponential decay of the latter is disturbed due to multiple scattering from particles and the surface. The strong field gradient of the evanescent wave and the Brownian motion of the particles allow to determine the particle-surface interaction potential by a measurement of the light scattered from individual particles. Coating of the substrate surface permits variation of this potential and examination of the properties of different surfaces, e.g. the surface charges.

Even the conventional scanning near-field optical microscope (SNOM), based on aperture probes, is still not sufficiently understood, in spite of a large amount of theoretical work. In order to interpret the images of such a microscope in the right way, it is necessary to determine the field distribution and radiation properties of a given probe/sample structure. Two- as well as three-dimensional simplified models of such a microscope were investigated for the first time by Novotny[38,39], using the multiple multipole method (MMP). In contrast to earlier works, the metal coating was not assumed to be ideally conducting, but instead described realistically by the complex dielectric constant of aluminum. With the MMP technique the exponential decay of the field amplitude in the near-field of the aperture, as well as the polarisation-dependent properties of the near-field have been reproduced. A restriction of the simulation was the fact, that not the whole fiber tip, but only the outermost region beyond the cut-off region was considered. This is not a problem in principle, but the simulation of the whole fiber tip would extend the calculation dramatically, making such calculations impractical. Besides, only time-harmonic fields can be simulated with the program package. This allows to study only stationary field distributions and excludes the examination of time-dependent pulse propagation in the fiber.

In this dissertation, a commercial program package, based on the finite difference time domain (FDTD) method, was employed, which does allow the consideration of time-dependent fields. In recent SNOM experiments[40,41,42] short optical pulses in the femtosecond range were employed, in order to combine the high spatial resolution of these microscopes with high time resolution (compare chapter VII). This motivates the study of pulse propagation, which, moreover, allows to examine in detail the tunneling process. Femtosecond spectroscopy is also a powerful tool for the investigation of single fluorescent emitters, such as organic molecules, or semiconductor quantum dots[43,44,45]. Corresponding simulations are difficult, because it is necessary to combine the mathematical methods developed in the field of near-field optics with quantum mechanical considerations. This problem will require further study in the future. The outcome of the field calculations performed in the present thesis may serve as an input for the required quantum mechanical calculations. In contrast to a number of simulations[46] recently published, the effects of the real metal used as a coating, especially the excitation of surface waves, are included in this calculation. The latter preclude, for example, a description of the tip-sample interaction as a dipole-dipole energy transfer process[47]. An example of FDTD-modeling of continuos waves through aperture probes was given earlier.[48] The cladding was modeled as an ideal conductor in this case and the phase properties were not investigated.

The optics of nanostructures can be described by analytical as well as numerical means. An important analytical tool is the theory of dyadic Green's functions (not employed in the present work). A standard method for the description of light scattering by non-spherical particles is the extended boundary condition (or null-field) method (EBCM, not used in this thesis)[49,50]. An overview is presented in ref. 51. The generalization of Mie's theory to interacting particles has been used in this thesis in chapter II. Semi-analytical and numerical techniques for general electrodynamic field calculations include the finite difference time domain (FDTD) and the finite integration technique (FIT) (compare chapter VII), the coupled dipole approximation and the multiple multipole technique. (compare chapter II).

The present thesis deals with electrodynamic field calculations for frequencies in the visible spectral range in context with subwavelength structures, whereby the near-field of these structures is of particular interest. Obviously this is beyond the validity of geometrical optics and conventional diffraction theories. A number of analytical and numerical approaches as well as experiments are employed for this purpose. In this work, only linear optics of nano-structures and, as far as light scattering is concerned, only elastic scattering processes are considered. It should be kept in mind, however, that linear optics forms the basis also for nonlinear applications, and, moreover, the well-known enhancement of inelastic light scattering processes like surface enhanced

Raman scattering (SERS)[52,53], is also partly based on linear near-field enhancements. Quantum mechanical effects are also not considered here[54], although they are, of course, relevant for very small structures and for the realistic description of solid surfaces. For example, many interesting applications within the optics of semiconductor nanostructures are connected with quantum mechanical effects. A good overview about this theme is given in ref. 55.

The discussion presented above should illustrate some of the frontiers of nano-optics and demonstrate some of the fascinating aspects of this rapidly progressing discipline.

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