III Scattering and Extinction of Evanescent Waves by Small Particles

The theory laid out in chapter II.2 showed, that, due to the strong transverse intensity gradient of evanescent waves, high-order multipoles are strongly enhanced in scattering and extinction of evanescent waves, compared to plane waves. In many cases this leads to strongly enhanced resonances in the corresponding spectra. Specific examples for the case of homogeneous spheres and for coated spheres are discussed in this chapter. In addition, the range of refractive indices, particle sizes, and thicknesses of the coating layer is investigated, for which well-defined multipolar resonances are obtained in the visible spectral range. A plane evanescent wave is generally produced by total internal reflection from an interface between two media. We initially want to assume, that the distance of the particles from the interface is large enough to justify the neglect of multiple scattering effects. The latter will be discussed later in this chapter.

III.1 Isolated Particles in Free Space

Homogeneous dielectric spheres

For dielectric spheres, the only possible resonances are the so-called morphology-dependent resonances (MDR’s), which are due to constructive interference and are observed for size parameters of order unity or larger. This critical size parameter depends on the real part $n$ of the index of refraction. It decreases, if $n$ increases. In order to obtain strong MDR’s, it is necessary to have a low absorption, i.e. a low imaginary part of the index of refraction. The MDR’s of large dielectric spheres are also called whispering gallery modes (WGM’s).

If the radius of the particles increases, the number of MDR’s and their amplitudes increase, whereas their half widths and the distances between them decrease in a given spectral range. In quantum optical applications very sharp and strong resonances can be important, e.g. in order to allow Rabi-splittings, other nonlinear effects or altered spontaneous emission characteristics to be observed at low pump power. E.g. for a silica sphere with radius $65 \, \mu m$ and refractive index $n = 1.4518$ at a wavelength of 900 nm the half-width amounts to 270 kHz, corresponding to a quality factor $Q = 1.4 \times 10^9$. This value is still limited by residual absorption and does not represent the intrinsic $Q$ value.

An example for the significant changes in the scattering spectra due to evanescent-wave excitation is shown in Fig. III.1, which displays the scattering cross sections of a spherical diamond particle of diameter $2a = 600 \, nm$. Similar results would be obtained for particles of lower refractive
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Obviously, evanescent-wave excitations do not modify the peak positions and the half widths, but merely the amplitudes of the resonances. Due to the oscillatory nature of the Ricatti-Bessel functions in the denominator of the wavelength-dependent Mie-coefficients multipole resonances of the same orders $n$ exist in the UV regime but are not considered here. For this reason, the modes are characterized by a second quantum number marking the radial dependence. This radial quantum number is equal to 1 for the resonances visible in Fig. III.1 and is not explicitly given there. The quantum numbers shown in Fig. III.1 correspond to the multipolar order of the resonance and, hence, describe the angular dependence of the scattered field. From the structure of the evanescent waves it follows, that TM modes can be excited more efficiently by p-polarized (TM) evanescent waves and TE modes by s-polarized (TE) evanescent waves.

In total, Fig. III.1 demonstrates the extremely efficient excitation of MDR’s by evanescent waves compared to the case of plane-wave excitation$^2$. For comparison, the different scales of the ordinate axes should be taken into account. The difference is easily understood by looking at Fig. III.2, which shows the instantaneous intensity distribution of the total field for the TM$_{14}$ resonance at $\lambda = 386$ nm of a polystyrene sphere, $r = 700$ nm, excited by a p-polarised evanescent wave. The figure suggests a mechanical analogue for evanescent-wave excitation of WGM’s consisting of a chain (representing the evanescent wave) driving a gear (representing the WGM). Fig. III.3 displays the time-averaged intensity of the scattered field alone in the same case on linear and logarithmic intensity scales. It may be easily recognised, that the intensity is confined to a narrow ring in the plane of incidence close to the inner surface of the sphere. If, contrary to the procedure laid

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig3.png}
\caption{Scattering spectra of a spherical diamond particle of diameter $2a=600$ nm for an incident plane wave (a) and for p- and s-polarized evanescent waves (b, c), respectively. For the evanescent waves an angle of incidence of 60° of the totally reflected wave and a constant index of refraction of the prism $n_p = 1.5$ is assumed. Note the different scales in b, c.}
\end{figure}
out in chapter II, the axis of quantization is chosen perpendicular to this ring, then, for evanescent-wave excitation, the WGM contains predominantly the m = n component of the vectorial multipole. For comparison, Fig. III.4 shows the same quantity for the case of plane-wave excitation. Here, the plane wave propagates from left to right. The intensity distribution is now very different from the one discussed above, and, in particular, is not confined to a narrow ring. Whereas in Fig. III.3 the WGM strongly dominates the intensity, the sphere acts mainly as a ball lens, when excited by a plane wave, and, correspondingly, the WGM is hardly recognisable in Fig. III.4. This is in agreement with the inefficiency of plane-wave excitation of WGM’s demonstrated in Fig. III.1. Correspondingly, the field distribution shown in Fig. III.4 contains significant contributions from other multipoles.

Another remark should be made here concerning the angular momentum of the electromagnetic field: In the case of evanescent-wave excitation of the MDR of a homogeneous particle (Fig. III.2, Fig. III.3) the ring-shaped pattern of intensity packets is rotating in the plane of incidence.
corresponding to a nonvanishing angular momentum of the electromagnetic field. This is contrary to the case of plane-wave excitation, where two counterpropagating waves are generated, which travel close to the inner surface of the sphere and produce a nonrotating intensity pattern with vanishing angular momentum and much smaller intensity.

Homogeneous semiconductor spheres
As a further example, Fig. III.5 depicts the optical scattering and extinction cross sections of a compact crystalline silicon sphere with \( 2a = 200 \) nm excited by a plane wave (Fig. III.5a) and by p- and s-polarized evanescent waves (Fig. III.5b,c), respectively.

Compared to diamond (\( n=2.4 \)), silicon has a much larger refractive index (\( n=3.5-6 \)) in the visible range. In addition, because the excitation energy is above the bandgap for Si, there is considerable absorption, whereas diamond is transparent in this range. The scattering spectra are qualitatively similar to those of the diamond particle. Contributions from higher-order multipoles to the extinction cross section are, however, stronger than for the scattering cross section, as the squared absolute values of the Mie coefficients, which appear in (II.60) of chapter II.2.2.2 decrease faster with order \( n \) than the real parts (II.59). E.g. Fig. III.5b shows, that for p-polarized evanescent-wave excitation, the absorption (extinction minus scattering) is marginal in the case of the TM2-mode, whereas the absorption cross section case is of the same order of
magnitude as the scattering cross section for the TM$_3$-mode. Comparison of the TE$_2$ and TE$_3$ modes for the case of excitation by a s-polarised evanescent wave yields qualitatively the same results (Fig. III.5c). It should be added, that the optical absorption cross section generally exhibits a stronger contrast between off-resonant and on-resonant conditions than the scattering cross section.

Homogeneous metallic spheres
Very small particles exhibit the well-known dipolar surface plasmon resonance at around 365 nm due to collective electron oscillation. As a first example, we consider the extinction cross sections of a very small silver particle (Fig. III.6). Because in this case, the Mie coefficients $a_n$ of the electric multipoles are much larger than the Mie coefficients $b_n$ of the magnetic multipoles, the TE modes do not appear in the spectra. Furthermore, because the particle is so small, only the dipolar coefficient $a_1$ contributes to the spectra to any significant extent. The shape of the extinction spectrum for evanescent waves is therefore unchanged in this case compared to plane-wave excitation, and the polarisation-dependent cross sections for extinction as well as for scattering are for all wavelengths very nearly in the ratio $N_s^{-1}T_1 / 1 / N_n^{-1}\Pi_1$ ($\Pi_1 = 1$) for p-polarised evanescent-wave excitation, respectively (compare eqs. II.59, II.60 and Fig. II.3 of chapter II.2.2.2). The comparison between the cross sections for plane-wave and evanescent-wave excitation depends on the definition of the normalisation factor of the cross sections, but the comparison between both polarizations in the case of the evanescent wave excitation does not.

For larger silver particles, like those of Fig. III.7 (2a=200nm), the extinction cross section is strongly enhanced for shorter wavelengths in the case of evanescent-wave excitation. This effect is particularly distinct for the case of excitation by a p-polarized evanescent wave. The ratio of the excitation-dependent cross sections in Fig. III.7 now depends on the wavelength, in
contrast to the spectra in Fig. III.6. In order to understand the spectra shown in Fig. III.7, Fig. III.8 displays the decomposition of the extinction spectra into the different multipolar contributions for p-polarized evanescent waves (Fig. III.8a) and for plane waves (Fig. III.8b), the numbers indicating the multipolar order n of the TM$_n$-resonances.

Fig. III.8 clearly reveals, that the maxima in the extinction spectra are caused by a strong enhancement of higher multipoles in the case of evanescent-wave excitation. The electric quadrupole and particularly the electric octupole give rise to increased extinction in the range of shorter wavelengths and a double-peak structure, if the excitation is a p-polarized evanescent wave. The enhancement of the higher multipoles is not caused by an excitation-dependent susceptibility of the particles, but merely by the fact, that the excitation itself contains stronger contributions from higher multipoles due to the large transverse field gradient of the evanescent wave. The difference in the cross sections for s-polarized and p-polarized evanescent-wave excitation, respectively, for a spherical particle is caused by the different internal structure of the exciting field in both cases. In the case of s-polarization, the electric field of the wave oscillates perpendicular to the plane of incidence, whereas in the p-polarized case, the electric field rotates in the plane of incidence due to the complex phase of the totally reflected wave. For this reason,

![Fig. III.7](image1.png)

**Fig. III.7:** Wavelength dependence of the cross sections for extinction of plane waves and evanescent waves by a spherical particle with 2a=200nm. As in Fig. III.6a the solid lines are the analytical results from (II.59 and II.60) and the squares indicate the numerical results obtained by means of the MMP method.

![Fig. III.8](image2.png)

**Fig. III.8:** Decomposition of the extinction cross section into individual multipole contributions for a silver particle with 2a=200 nm. Excitation p-polarized evanescent wave (a), plane wave (b). The numbers indicate the order of the multipole, whose contribution to the total extinction cross section is shown. Results for s-polarized evanescent waves are not shown here, but are similar to those for p-polarization.
for a spherical particle in front of a metallic mirror a polarization-dependent cross section is also expected even for the case of plane-wave excitation. In order to demonstrate, that the numerical calculations yield the same results as the analytical formulae, the symbols representing the numerical results have been added in Fig. III.6 and Fig. III.7. The numerical method will be used below to take multiple scattering at the interface into account.

**Coated spheres**

Whereas for homogeneous dielectric spheres either a large sphere radius or a sufficiently large refractive index is required to obtain morphology-dependent resonances, comparatively small particles can exhibit resonances in the visible spectral region, if they are coated by a metal. The resonance position for the coated spheres can be calculated by formula (II.84 and II.85). The imaginary part of the index of refraction can be neglected for silver-coated particles, because it is sufficiently small for silver and does not vary drastically with the wavelength. The index of refraction for silver, as well as the resonance positions for a compact silver sphere with diameter 2a=22 nm and for a coated sphere with a silica core of diameter 2a=20 nm and a silver shell of thickness d=1 nm are shown in Fig. III.9 for a broad spectral range.

Fig. III.10 displays the dependence of the extinction spectra of silica particles coated with a silver shell of thickness d = 10 nm for the case of p-polarised evanescent-wave excitation and core radii...
a = 20 to 150 nm in 10 nm steps. For clarity the spectra are shifted vertically by a constant amount. The individual multipolar contributions are identified. The weak line marked with a star arises from all multipoles appearing in the spectrum and corresponds to the resonances close to $\varepsilon_1(\text{Ag}) = 0$, compare Fig. III.9. For the smallest core radius of $a = 20$ nm only the dipole contribution is observed. With increasing core size higher multipoles appear and the splitting between the resonance positions increases. At the same time resonances due to lower multipole orders broaden and shift to the red. The resonance at $\lambda \approx 340$ nm corresponding to $\varepsilon_1(\text{Ag}) = 0$ remains at approximately the same position for all core sizes. It should be mentioned, that the calculated spectra shown in Fig. III.10 do not take into account size-dependent corrections of the dielectric function, which are known to be necessary for small metallic structures. These corrections essentially lead to a broadening of the calculated resonances.

Fig. III.11 shows the dependence of the observable resonance positions on core size for thicknesses $d = 1, 2, 5, 10$ nm. The

\begin{figure}
\centering
\includegraphics[width=\textwidth]{fig11.png}
\caption{Resonance positions of the electric multipoles $\text{TM}_n$ for silver-coated silica particles as a function of core radius and thickness of the shell. The straight lines are guidelines for the eye.}
\end{figure}
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lines through the calculated positions are only guidelines to the eye, but to a good approximation a linear relationship is observed.

In an attempt to verify the calculations for metal-coated spherical particles shown above homogeneously silver-coated latex spheres have been prepared in our group in the meantime\(^5\). Smooth silver coatings of thickness \(d=11.5\ \text{nm}\) were obtained. Confocal wavelength-resolved scattering microscopy of individual particles of this kind did not reproduce, however, the resonances discussed above, and a considerable variation of the scattering spectra was obtained from particle to particle. This is ascribed to the strong influence of the unavoidable grain structure of the silver shell on the scattering spectra (compare here also chapter V).

III.2 Effects of Multiple Scattering at the Surface of a Substrate

In evanescent-wave scattering experiments the particle is almost always in contact with the interface, where the evanescent-wave is generated by total internal reflection. Modifications of the scattering spectra due to backscattering from the interface must then be taken into account. These effects will now be discussed for a number of examples. Applications to total internal reflection microscopy\(^6\) are discussed. The following discussion is also relevant for apertureless near-field optical microscopy (chapter VI).

**Homogeneous silver spheres:**

As an example, the cross sections for a spherical silver particle in contact with a glass prism were calculated via the MMP technique and are shown in Fig. III.12: a redshift of the spectra due to the presence of the interface is universally recognised.

![Figure III.12](image-url)  
**Fig. III.12.** Comparison of the cross sections of a silver sphere with diameter \(2a=200\ \text{nm}\) in free space (solid lines) and on the glass prism (squares) for excitation with a plane (lower two curves) and a p-polarized evanescent wave (upper two curves).
Moreover, the scattering cross section is enhanced in the wavelength range $\lambda > 0.4 \mu m$ in both polarisations, whereas the absorption is strongly reduced in $p$-polarisation for $\lambda < 0.4 \mu m$.

Homogeneous dielectric spheres

In this case, again considerable modifications of the scattering spectra occur. As an example, in Fig. III.13 the cross sections for a spherical diamond particle in contact with a glass prism are shown: Compared to the isolated particle, the resonance wavelengths are again slightly shifted to longer wavelengths. The resonances are broadened considerably, and, correspondingly, their amplitudes are reduced substantially.

An equivalent model consisting of a spherical scattering object, which is embedded in a medium with a higher index of refraction approximately describes the spectra. If the index of refraction of the environment increases, the resonances shift to longer wavelengths and in this way this simple model can describe the observed red shift. The red shift is larger for TM modes than for TE modes. This may be explained in a heuristic fashion by the fact, that for TE modes the electric field is parallel to the surface, which corresponds to a serial arrangement of the impedances of both halfspaces, whereas for TM modes there is a field component normal to the surface, which means that the impedances are parallel. Simplifying the surface as an equivalent electric circuit consisting of two serial or parallel capacitances, respectively, a larger effective dielectric constant for the TM modes is obtained. Comparison of Fig. III.1 and Fig. III.13 makes it clear, that large resonance enhancements are possible, when the particle is held at some distance from the prism surface. On the other hand, even when the sphere is in contact with the surface, the spectra for evanescent-wave excitation differ strongly from those for plane-wave excitation, and resonances with large peak amplitudes are observed for $s$-
as well as p-polarised evanescent waves. The broadening and the reduction of the peak heights may be explained by the disturbance of the total internal reflection due to multiple scattering processes. The latter result in a smaller transverse field gradient, equivalent to reduction of the multipolar enhancement factors, and lead to a strong energy flux into the prism, equivalent to a reduction of the quality factor. This energy flux is described by the Poynting vector. Fig. III.14 shows the time-averaged Poynting vector for the TM₆ resonance of the particles of Fig. III.1 and Fig. III.13 and demonstrates the large changes of the scattered field due to multiple scattering processes, if the particle is in contact with the interface.

Multiple scattering effects like those shown in Fig. III.14 are obviously highly relevant for Total Internal Reflection Microscopy (TIRM). In this experimental technique light scattering by particles, which move in an evanescent wave, is used to study the motion of the particle perpendicular to the interface with high sensitivity to small changes in the distance to the interface, and to characterise in this way particle-interface potentials.

It is thereby generally assumed, that the

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Fig. III.14: Time-averaged Poynting vector of the scattered field for a diamond particle of radius $a=300\text{nm}$ and for p-polarized evanescent-wave excitation at the TM₆ resonance at 456 nm in free space (a), and at 458 nm, when the particle is in contact with the prism surface (b). The latter is indicated by the straight line, and the prism is to the right of the line. The evanescent wave propagates from the top of the figure to the bottom and decays from the right to the left.
scattered power is proportional to that of the exciting evanescent wave at the center of the sphere, and, hence, decreases exponentially with the distance \( d \) to the interface. Under this assumption, the particle-wall interaction potential can be calculated from the probability distribution of \( z \) and the use of the Boltzmann probability distribution: \( w(d) \sim \exp(\phi(d)/kT) \). This was first applied by Prieve et al.\(^6\) in order to study colloidal forces in the proximity of a surface. In recent experiments\(^7\) the Coulomb interaction between negatively charged particles (assumed to be infinitely hard in first approximation) and a negatively charged surface was determined in this way. Gravity appears as an additional potential in this experiment.

The method is very sensitive, because the forces acting on the particles can be measured in the range of 10 fN. The calculated potential, however, will be affected by any deviation from the exponential dependence due to multiple scattering effects in the vicinity of the surface. The method can be used for the examination of surfaces, e.g. to distinguish between surfaces of different polarity.

We have calculated the power scattered from a diamond sphere (\( r=300 \) nm, same as above) as a function of the distance \( d \) from the interface, where the evanescent wave is generated. Similar results are expected for polymeric particles of somewhat larger size. Scattering cross sections for a \( p \)-polarized evanescent wave at two different wavelengths, \( \lambda = 458 \) nm, corresponding to the resonance (\( \text{TM}_6^1 \)) of the isolated sphere, and \( \lambda = 488 \) nm, corresponding to off-resonant excitation, were determined as a function of \( d \). The differential scattering cross section was integrated I) over a solid angle of \( 4\pi \) in order to obtain the total power scattered by the particle and II) over a finite solid angle (aperture angle \( 53^\circ \)) in the vacuum half space above the particle in order to calculate the signal obtained experimentally with a \( N = 0.8 \) microscope lens.

Fig. III.15 displays the results for the case of resonant excitation of the whispering-gallery resonance \( \text{TM}_6^1 \) at \( \lambda = 458 \) nm, as well as for off-resonant excitation \( \lambda = 488 \) nm. As in Fig. III.14b strong scattering into the prism is easily recognised, when the particle is in contact with the substrate (Fig. III.15a,b). Moving the particle away from the surface by a few hundred nanometers results in a drastic reduction of this directional scattering into the prism (Fig. III.15c,d). Furthermore, in the case of resonant excitation of a WGM, the differential cross section for scattering scattering into the vacuum half space exhibits much stronger modulations as a function of the scattering angle than in the case of off-resonant excitation. This holds independent of the particle-interface distance. The
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Fig. III.15: Absolute value of time-averaged Poynting vector for a diamond particle of radius $a=300\text{nm}$, which is excited by a $p$-polarized evanescent-wave of wavelength of $\lambda=458\text{ nm}$ (TM$^6$ resonance: part (a,c)) and 488 nm (off-resonant excitation, b,d), respectively. The images a,b show the results for the particle in contact with the interface, where the evanescent wave is generated, and b,d those for the particle 550 nm above it. The boundary is indicated by the straight line, and the optically denser medium is to the right of this line. The evanescent wave propagates from the top of the figure to the bottom and decays from the right to the left. The scattered fields are shown (left and middle part) in the plane of incidence. The modification of the corresponding potentials (explanation in the text) due to multiple scattering processes are shown in the right part for the resonant (e) and the non-resonant (f) case. The inserts in (e,f) show the integrated powers on a logarithmic scale. The main graphs show the integrated powers on a linear scale after normalisation to the expected exponential dependence. The black line represents the corrections necessary in TIRM.

inserts in the graphs (Fig. III.15e,f) show the integrated powers on a logarithmic scale. In the main graphs these results are shown on a linear scale after normalisation to the expected exponential dependence.

Due to multiple scattering effects involving the substrate and particle boundaries, an increase of the total scattered power (red lines) over the simple exponential is clearly observed. This is related to the strong scattering into the prism, when the particle is close to contact with the interface. The power scattered into the lens (black lines), on the other hand decreases by about 25% in the case of resonant excitation and by about 10% for off- resonant excitation, relative to the expected exponential dependence. This can
be explained by the strong directional emission into the medium with the higher index of refraction discussed above, which leads to reduced scattering into the vacuum half space. The inserts in both graphs show, that the intensities have to be determined with high accuracy over several orders of magnitude, which is experimentally quite difficult.

Our calculations show, that it is important at which wavelength the TIRM experiment described above, is carried out and that sizable deviations from the exponential distance dependence indeed exist, especially at wavelengths, where whispering gallery modes (WGM’s) of the sphere are resonantly excited. These corrections lead, of course, to altered particle-wall potentials.

We have investigated not only diamond particles in air, but also polystyrene spheres in water on a substrate surface, because real experiments often employ this set-up. The corresponding results at a wavelength of $\lambda = 500$ nm are presented in Fig. III.16. If the particle is in contact with the surface, a highly directional emission into the optically denser medium is again observed. Because of the relatively small difference of the index of refraction of water and polystyrene and the relatively small particle diameter, the light is scattered mainly in the direction of propagation of the evanescent wave, almost tangential to the interface. This applies to the scattered intensity in both

![Fig. III.16: Absolute value of the time-averaged Poynting vector for a polystyrene particle of radius $a = 300$ nm in water, which is excited by a $p$-polarized evanescent wave at a wavelength of $\lambda = 500$ nm. The images (a,c) show the results for the particle in contact with the interface, where the evanescent wave is generated, and the images (b,d) those for the particle 550 nm above it. The boundary is indicated by the straight line, and the optically denser medium is to the right of this line. The evanescent wave propagates from the top of the figure to the bottom and decays from the right to the left. The scattered fields are shown in the plane of incidence (a,b) and perpendicular to it (c,d). The main graphs in part (e) show the integrated powers on a logarithmic scale. In the insert, these results are shown on a linear scale after normalisation to the expected exponential dependence.](image)
media (glass, H₂O). It is easy to recognize, that most of the scattered power does not enter the lens in this case.

The total scattered power depends on the surface-to-surface distance in a way quite different from the case considered above. Here, not only the power scattered into the solid angle in water, corresponding to the numerical aperture of the lens, but also the total scattered power decreases with decreasing surface-to-surface distance. Besides, the power scattered into the lens decreases already for a distance of about 500 nm. This must be ascribed to the change in the field distribution with increasing distance, which is different in the present case, compared to the case considered in Fig. III.15.

### III.3 Interacting Dielectric Spheres in free Space

In many cases scattering and extinction of more than one particle is of interest. If the particles are close to each other the most important change in the spectra is a splitting of the resonances due to the interaction of the resonant modes. For metallic particles the situation becomes actually quite involved the delocalised nature of the electronic wavefunction. This is discussed in detail in chapter V. In the present paragraph only dielectric particles are considered. In this case the whispering-gallery modes of the individual spheres interact due to the evanescent fields of these modes outside of the particles. The coupling is, however, relatively weak, and the condition for constructive interference of the WGM is approximately unchanged. Therefore, the resonant modes of two interacting spheres (‘photonic molecule’⁸) can still be classified according to the TE and TM modes of the individual spheres. The main effect of the interaction is then a splitting of the whispering-gallery resonances. This is demonstrated in Fig. III.17, which displays the scattering spectra of two interacting polystyrene spheres, r = 700 nm, in a narrow spectral range. The spectra have been calculated using the MMP technique, and multiple scattering processes due to the presence of the substrate surface have been neglected here.

Fig. III.17: Scattering cross sections for two contacting polystyrene spheres (r=700nm) excited by an evanescent wave. The spectra were calculated using the MMP technique. Multiple scattering processes due to the presence of the substrate surface have been neglected here.
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and 388 nm (c,d). The two resonances differ, because the phase difference of the two WGM’s of both spheres is different at the two wavelengths. This phase difference is determined by the wavelength of the evanescent wave and the distance of the two particles. It controls the coupling of the two WGM’s at the contact point. Fig. III.19 shows, that the almost isotropic intensity distribution for a single particle in the plane of incidence (compare the left column of images in Fig. III.3 becomes highly anisotropic for a dimer of such particles. Light is emitted predominantly in two directions corresponding to the upper right and lower left corners of the images in Fig. III.19.

III.4 Interacting Dielectric Spheres on the Surface of a Substrate

If the particle dimer is now placed on a substrate surface, multiple scattering processes lead again to a substantial modification of the intensity distribution (Fig. III.18). As in the case of an isolated particle on a substrate (Fig. III.14, Fig. III.15) strong emission into the substrate is observed. The emission pattern, however, is much more complicated than in the case of isolated spheres. This applies also to the emission pattern into the vacuum half space.

It is immediately evident, that the field distribution will become even more complicated, if particle arrays containing more than two particles are considered.
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Such arrays may be interesting, e.g. for optical computing purposes, or in order to create photonic crystals with band structures in two or three dimensions.

Using the MMP method, we have modeled the scattering of evanescent waves by a chain of six latex spheres (r=0.7µm) on the surface of a substrate (n=1.5) for different wavelengths of the exciting light. Multiple scattering effects at the boundary of the surface are taken into account in this calculation. We have chosen a range of wavelengths, where resonances of the single particle occur. The scattering spectrum of such a particle was shown in Fig. III.17. The images in Fig. III.20 show the absolute value of the time-averaged Poynting-vector in the plane of incidence. Similar models were recently examined by other groups. Within the spheres and in the immediate neighborhood a complicated intensity distribution is obtained. Noticeable is the high intensity contrast in the images, and the sensitive dependence of the local intensity on the excitation wavelength. In order to understand the basic features of these results, it is useful to reconsider the spectra for two such spheres in contact with each other (Fig. III.17). These spectra show a splitting of the whispering-gallery resonances into two modes. It is expected, that in the case of a chain of n spheres, the resonances split into n components. The rapid intensity variations as a function of excitation wavelength can hence be related to multiple splitting of the single-particle resonances. For large chains or two-dimensional structures, which may be considered as ‘photonic crystals’, these splittings correspond, of course, to the band structure of the crystal. The k vector of the mode excited by the evanescent wave is then determined by the angle of incidence of the wave inside the substrate.

A scan of the latex chain with a SNOM tip at a constant height above the chain would scan images depending strongly on the scan height. The images displayed in Fig. III.20 show highly directed radiation into the optically denser medium and, less intense, into the vacuum. It is possible to address different spheres by choosing the appropriate wavelength. For example, the first image in the top row of Fig. III.20 (λ=408nm) shows a high intensity at the top sphere, and the first image in the bottom row (λ=414nm) a high intensity at the second latex sphere from the bottom. This ‘addressing’ of individual spheres concerns both, the radiation into the substrate and into the vacuum domain. This may be of interest for the construction of optical switches in the field of optical computing. In order to address individual spheres in a 2-dimensional lattice of latex spheres, it is necessary to generate two exciting evanescent waves, which pro-
pagate in two different directions on the substrate surface. If the latex array becomes very large, however, the addressing of the inner spheres will become more difficult, because the inner spheres in the array become more and more equivalent. As may be recognized from Fig. III.20, the radiation is emitted mainly from the spheres at the end of the chain and only weakly from the spheres in the middle part.

Whispering-gallery modes can be efficiently excited not only by evanescent waves, but also by light-emitting dipoles within the spheres. Experimentally, such active spheres can be realized by doping the spheres with fluorescent molecules, semiconductor quantum dots or other fluorescent particles\(^{10}\). The latter may be excited by a plane wave, whose frequency fits to an absorption of the dopant. Lasing in micro-cavities is currently studied in many groups, and in this context, stimulated emission in photonic crystals made of latex spheres is of considerable interest.
Fig. III.20: Linear chain of six latex spheres (radius \( r = 700 \text{ nm} \)) in vacuum on a glass substrate \( (n=1.5) \), where an evanescent wave is generated by total internal reflection of a plane wave (white arrows) incident under an angle of \( 60^\circ \) on the glass-vacuum interface. In the glass medium a standing wave is formed. The evanescent wave propagating in the vacuum domain is scattered by the latex spheres. The intensity (absolute value of the time-averaged Poynting vector) was calculated by the MMP-method for 8 different wavelengths (values given on top of the individual images) and plotted using a logarithmic color-coded scale.
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References

2. Any sizable absorption, for example due to impurities in the material, would of course broaden the resonances and strongly reduce the resonant cross sections. In fact the line widths found experimentally in ref. 1 were limited by residual absorption.