Exploring the detection limits of infrared near-field microscopy regarding small buried structures and pushing them by exploiting superlens-related effects

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Abstract: We present a study on subsurface imaging with an infrared scattering-type scanning near-field optical microscope (s-SNOM). The depth-limitation for the visibility of gold nanoparticles with a diameter of 50 nm under Si3N4 is determined to about 50 nm. We first investigate spot size and signal strength concerning their particle-size dependence for a dielectric cover layer with positive permittivity. The experimental results are confirmed by model calculations and a comparison to TEM images. In the next step, we investigate spectroscopically also the regime of negative permittivity of the capping layer and its influence on lateral resolution and signal strength in experiment and simulations. The explanation of this observation combines subsurface imaging and superlensing, and shows up limitations of the latter regarding small structure sizes.

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1. Introduction

Within the last decades fabrication and characterization techniques for electronic devices have reached the nanoscale. For the characterization of structural properties a variety of electron and scanning probe microscopy techniques exists with a spatial resolution down to the nanoscale or even smaller [1, 2]. However, for the dielectric properties conventional optical microscopy and spectroscopy still faces the problem of being diffraction limited to about one half of the used wavelength $\lambda$. The use of mid-infrared (mid-IR) light enables sensitivity to molecular vibrational modes as well as phonon excitations and free charges [3–11], but the resolution is usually limited to the micrometer scale. A possibility to circumvent this limitation is the use of optical near-fields [12]. In scattering-type scanning near-field optical microscopy (s-SNOM) a standard atomic force microscope (AFM) is extended by an optical setup illuminating the tip with a focussed laser beam and collecting the backscattered light. At the apex of the tip, evanescent fields are generated [12, 13]. Those near-fields couple to the sample region directly underneath the tip. Therefore, an optical image can—simultaneously to the topography scan—be acquired, providing information about the local dielectric properties of the sample [6,14,15]. The resolution is limited by the radius of curvature of the tip at its apex, which is typically in the range of about 20 nm [13]. By using the AFM in tapping mode and evaluating the amplitude $s_n$ for higher demodulation orders $n$ of the tapping frequency, the background is suppressed, however at the cost of lower overall signal strength [13]. Since the near-fields penetrate into a dielectric sample, non-destructive imaging of subsurface structures is possible up to a depth of about 100 nm, although with a loss in resolution and signal strength [16–22]. Even larger probing depths can be reached by the use of superlenses [23–26]. The capability of non-destructive imaging through thin layers of a dielectric material provides a useful tool for the investigation of nanocomposite materials: nanoparticles are embedded into a dielectric medium in order to tailor specific properties as e.g. absorption in solar cells [27,28] or antimicrobial resistance and water-repellance of textile fibres [29]. Another important application are nanoelectronic devices that are often covered by dielectric capping layers such as SiO$_2$ or Si$_3$N$_4$ in order to e.g. prevent them from oxidation [30,31].

The change of contrast [18] and visibility [19] of subsurface structures with different tapping amplitudes as well as the influence of the demodulation order [18] have already been investigated on structures with a size of about 100 nm, which is much larger than the resolution of s-SNOM at the surface. All these measurements have been performed at wavenumbers where the permittivity of the cover layer was positive ($\varepsilon > 0$).

The goal of this work is to study the limitations of subsurface imaging regarding smaller structure sizes as well as the dependence on the dielectric function of the capping layer. We image small metallic nanoparticles below a Si$_3$N$_4$-membrane and vary membrane thickness and particle size. In order to be comparable with the previous reported results [18, 19], we first choose a wavenumber for imaging and modelling where the permittivity of the cover layer is positive. TEM imaging enables a correct attribution between structure size and the corresponding optical signals, which in turn are compared with a simple model calculation yielding quite good agreement. Finally, we perform spectroscopic s-SNOM measurements and simulations in the spectral range where the permittivity of Si$_3$N$_4$ also becomes negative for optimizing the imaging conditions: The resolution considerably changes and the contrast even inverses as the optical properties of the capping layer are strongly wavelength-dependent.

2. Setup and sample system

A commercial s-SNOM system (neaSNOM by neaspec) with a pseudo-heterodyne detection scheme [32] has been used in this work. The SNOM tips are Pt-coated silicon tips (Nanoworld Arrow NCPT). Our sample system is depicted in the upper part of Fig. 1(a). It consists of commercially available Si$_3$N$_4$ TEM membranes of various thicknesses ($d_{Si_3N_4} = 30 \text{nm}/50 \text{nm}$: Ted \#251220. 

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which have been chosen as $\text{Si}_3\text{N}_4$ is a commonly used capping material for nanoelectronic devices. $\text{Si}_3\text{N}_4$ has a Reststrahlenband within the mid-IR. This means that for wavenumbers $\nu = \frac{\lambda}{\lambda - 1}$ between 870 cm$^{-1}$ and 1090 cm$^{-1}$ the real part of the dielectric function becomes negative. Within the Reststrahlenband surface phonon polaritons (SPhPs) can be excited at any scattering source and propagate along the dielectric/air interface, resulting in high near-field amplitudes [33]. The SPhPs exhibit a resonance for $\text{Re}[\varepsilon] = -1$ (superlens condition [23]), which is only observable as long as $\text{Im}[\varepsilon]$ is small. Imaging within both regimes of positive and negative permittivity shall be investigated in this work.

Au nanospheres (bbi solutions) of different sizes have been drop-casted onto the backside of the membrane as a model system: they can be produced in a very accurate size distribution and due to the highly negative dielectric function of Au in the mid-IR [34] they do not show any resonance. This sample system allows for topography-free imaging, excluding topographical influence of the optical signals.

3. SNOM imaging of nanoparticles buried below a dielectric

The samples have been investigated by evaluating the optical amplitude $s_2$ images of the second demodulation order (unless otherwise stated), in order to have the background suppressed while on the other hand having sufficient overall signal strength. The SNOM images were acquired with a tapping amplitude of 60 nm as a compromise between mechanical stability, high contrast and low noise [19]. A pixel size of 20 nm and a sampling time of 30 ms/pixel have been chosen. The illuminating laser has been set to a wavenumber of $\nu = 1240\text{cm}^{-1}$, where $\varepsilon_{\text{Si}_3\text{N}_4} > 0$, and a laser power of 4 mW. In Fig. 1 SNOM images of Au spheres ($\varnothing = 50\text{nm}$) under 30 nm,

Fig. 1(b), and 50 nm, Fig. 1(c), of $\text{Si}_3\text{N}_4$ are shown. In Fig. 1(b) bright spots with a diameter of 130 nm can be revealed (black circle). These spots have no equivalent in the corresponding topography images (not shown), therefore they can be expected to stem from the gold spheres underneath the membrane. In Fig. 1(c) however there are some black dots that can be attributed
to dirt on top of the sample that appears in the topography images as well. The black circle and rectangle mark two regions of higher SNOM signal. In the square there is a clearly visible bright spot that appears larger than the spots in Fig. 1(b). In the circle there is a slightly brighter region only hardly visible that has no counterpart in the topography image. Therefore, this seems to stem from underneath the membrane as well. This observation in combination with the spot size being much larger than the particle size raises the question if the larger spot might stem from a cluster of Au spheres instead of a single sphere. In order to ensure that only spots of single spheres are evaluated, brightfield mode TEM images of the same sample regions have been acquired, Figs. 1(d) and 1(e). They clarify the real distribution of the spheres: The large spot in Fig. 1(c) corresponds to a cluster of four spheres and the hardly visible brighter region corresponds to a single sphere. From this we can conclude that in a depth of 50 nm the coupling is getting nearly vanishingly weak and we are very close to the limit of depth, in which the Au spheres can be detected.

4. Simulation

For the evaluation of the imaging properties two quantities are important: the spot size and the signal strength. In order to describe the sample system theoretically, in a first step we performed numerical simulations based on the finite element method with the commercial software COMSOL. In order to keep the computation times short, we restricted the simulation to two dimensions. The simulations have been performed for a 30 nm thick membrane and an elliptical SNOM tip with an apex radius of 30 nm and 300 nm in length. The incidence angle of the illumination has been set to 60° from the normal to the membrane. Floquet conditions have been used as boundary conditions. In Fig. 2 the field distribution is shown for a wavenumber \( \nu = 1240 \text{ cm}^{-1} \) for two different cases. In the first case, Fig. 2(a), the SNOM tip is set on top of the membrane right above a Au particle of 50 nm in diameter, in the second case, Fig. 2(b), the SNOM tip is above the membrane without a particle underneath. The second case represents the situation of the tip being far away from the particle, which could be used for referencing the signal strength to. In order to determine the spot size, many simulations for varying tip position above the particle would have been needed, which was not possible due to limited computing resources. In order to have theoretical predictions for our experiments we therefore use an analytical model, which is very simplifying, but a lot faster to calculate and still it gives a quite good qualitative estimate for the experimental results, as long as we are in the regime of positive permittivity for Si₃N₄.
5. Model calculation

The analytical model calculations have been done in the electrostatic approximation [12]. This is reasonable, since all important length scales such as the radii of tip \( r_t = 30\) nm and Au sphere \( r_s \) and their distance are much smaller than the wavelength of the mid-IR light. The simplifying assumptions of our model are depicted in Fig. 1(a): Tip and Au particle are in a first approximation described by spheres with a point dipole in the center. The strength of their polarizabilities ˆ\( \alpha_t \) (tip) and ˆ\( \alpha_s \) (sphere) depends on the radii \( r_t \) and \( r_s \), respectively. In order to take account for the higher polarizability ˆ\( \alpha_t \) along the shaft of a SNOM tip, its z-component has been enhanced by a factor of 10 [35]:

\[
\alpha_t = \begin{pmatrix}
\alpha_x & 0 & 0 \\
0 & \alpha_x & 0 \\
0 & 0 & 10\alpha_x
\end{pmatrix} \quad \text{with} \quad \alpha_x = 4\pi \varepsilon_0 \frac{\varepsilon_t - \varepsilon_{Si_3N_4}}{\varepsilon_t + 2\varepsilon_{Si_3N_4}}
\]

(1)

The two dipoles have been embedded into a surrounding with the dielectric function \( \varepsilon_{Si_3N_4} \) of \( Si_3N_4 \), neglecting the \( Si_3N_4/\text{air} \)-boundaries. In order to take account for the experimental angle of incidence, the orientation of the incident electric field \( E_{\text{inc}} \) has, as in the simulation, been set to 60°. The application of this very simplified model calculation allows for good predictions for the signal strength and resolution in the experiments as shown in Figs. 3 and 4 and further discussed in the corresponding experimental sections. Signal strength and spot size are determined in experiment and model calculation from a gaussian fit to line plots through acquired or calculated 2D images. The spot size is in both cases given as the FWHM of the gaussian fit.

For the signal strength slightly different definitions have been chosen:

\[
\text{experiment:} \quad s_{2,\text{rel}} = \frac{s_{2,\text{max}} - s_{2,\text{bg}}}{s_{2,\text{bg}}}
\]

(2)

\[
\text{calculation:} \quad E_{\text{rel}} = \frac{E_{\text{max}} - E_{\text{bg}}}{E_{\text{tip}}}
\]

(3)

\( s_{2,\text{max}} \) and \( E_{\text{max}} \) are the maxima and \( s_{2,\text{bg}} \) and \( E_{\text{bg}} \) the background level of the gaussian fits. \( E_{\text{tip}} \) is the pure scattered field of the tip. We chose these definitions in order to have comparable quantities by eliminating differences between experiment and model calculation as good as possible. First of all the neglected membrane/air interface could lead to a different overall background level compared to the experiment. This can be subtracted out by taking the difference between the maxima and the background level. Another difference is the influence of the particle for the case of the tip being far away from the particle. In the experiment the influence of the particle can be neglected for the tip being far away. However, in the calculation the background level of the gaussian fit is given by the scattering of tip and particle, even if they do not couple (\( E_{\text{bg}} = (\hat{\alpha}_t + \hat{\alpha}_s)E_{\text{inc}} \)). Therefore, in this case the pure scattered field of the tip \( E_{\text{tip}} = \hat{\alpha}_t E_{\text{inc}} \) has been used for normalization.

6. Particle size dependence

We investigated the dependence of spot size and signal strength on the size of the Au particles. Different sizes of particles have been drop-casted onto one 30 nm thick membrane and imaged with TEM and SNOM in the same way as the images in Fig. 1. The experimental results in Fig. 3(a) (blue data points) reveal an increase in spot size for larger particles. However, the spots of smaller particles appear enlarged. In a comparison of the spot size with the actual size of the particle as shown in Fig. 3(b), it can be seen that the ratio is much larger for small particles, but approaches 1 for the largest particles investigated. The smallest particles with a diameter of 30 nm have a size close to the resolution limit of the SNOM at the surface. Our measurements show that if they are buried under a thin dielectric layer the resolution in terms of spot size gets...
worse, but nevertheless, the detection is still possible with sub-wavelength resolution of about $\lambda/60$. The model calculation shows the right trend for spot size, Figs. 3(a) and 3(b), and signal strength, Fig. 3(c). The former is underestimated and the latter fits the data quite well except for larger particle diameters, since the point dipole approximation is more accurate for smaller particles. The discrepancies between model calculation and experimental data can be explained by the simplifications of the model calculation. Besides the point dipole approximation and the neglected membrane/air interface, the model calculation does not include the demodulation of the signals with the higher harmonic of the tapping frequency. Including this would presumably lead to higher spot sizes, as the distance between tip and particle dipole is on average larger than only the thickness of the membrane. Another source of error could be a larger tip radius than the one used for the model calculation ($r_{\text{tip}} = 30\text{nm}$). Even if care was taken, degradation of the SNOM tip during the measurements cannot be excluded, too. For future investigations, an extension of the model including the demodulation and an observation of the tip radius e.g. by SEM measurements could lead to better agreement between experiment and calculations. However, despite the simplicity of the model calculation, it already yields a good qualitative estimate.

7. Spectroscopic investigation

In the second part of our work, we focussed on the investigation of the wavelength dependence in order to find the best imaging conditions. As already mentioned, the dielectric function of Si$_3$N$_4$ changes sign within the mid-IR, influencing the imaging. For this study a single Au sphere ($\varnothing = 50\text{nm}$) has been scanned under a membrane thickness of 30 nm with various wavenumbers $\nu$ between 925 cm$^{-1}$ and 1240 cm$^{-1}$. Since the model calculation does not include the membrane/air interface, which plays an important role in the resonant wavelength regime due to the presence of SPPhPs, here we compare our results with simulations. The signal strength in the simulation $E_{\text{rel}}$ is defined as the difference between the electric fields at the tip apex for the cases with (cf. Fig. 2(a)) and without (cf. Fig. 2(b)) a Au sphere underneath the membrane normalized to the field without the sphere. The results are plotted in Fig. 4(a).
Note that for the experimental data due to a high background signal, in contrast to all other measurements described here, for this investigation the $s_3$-signal has been evaluated. The contrast strongly varies with $\nu$ and even inverses below $960 \text{ cm}^{-1}$: The Au sphere does not appear as a bright spot, but as a dark spot compared to the background of the membrane, as can be seen in the SNOM images in Fig. 4(b). Such a contrast inversion has also been observed for silicon carbide superlensing [36], and it nicely fits to the results from the simulation. Only in the range for larger wavenumbers there is a discrepancy between simulation and experiment, which might be due to a mismatch of the used dielectric function [33], stemming from a different method of deposition or another film thickness of the Si$_3$N$_4$ of the sample from [33].

The spectral behavior of the spot size is plotted in Fig. 5(a). The expectation is related to the resonance of the surface phonon polaritons (SPhPs) for $\text{Re}[\varepsilon] = -1$: At the resonance $\lambda_{\text{SPhP}}$ becomes small. The propagation of the SPhPs is sharply dampened, leading to a strong confinement. A small wavelength corresponds to high spatial frequencies $k_x = \frac{2\pi}{\lambda_{\text{SPhP}}}$, which are expected to result in a high spatial resolution [37]. In Fig. 5, the spot size for a single particle, Fig. 5(a), is compared to a $\nu$-dependent measurement of an edge of a membrane field from the same commercially available membranes as for the previous experiments, which means an edge between Si and air underneath Si$_3$N$_4$, Fig. 5(b). The resolution of the edge has been determined by fitting a logistic function and determining the width of the 10\% to 90\% - regime of the signal levels. The overall resolution is much better for the small particle than for the edge. This can be explained by the similar size of tip and particle: they both couple best to the same spatial frequencies corresponding to the inverse of their size. This higher coupling leads to a better resolution. In Fig. 5(c) $\varepsilon_{\text{Si3N4}}$, taken from [33], is plotted. Red arrows indicate the positions, where the superlensing condition would be fulfilled for this dielectric data. In the case of the Si/air edge two distinct minima in resolution can be seen (red arrows). These positions are shifted in regard to the expected positions from the dielectric function. This discrepancy might again be due to a mismatch of the dielectric function. Since the real part of $\varepsilon_{\text{Si3N4}}$ has
Fig. 5. Spectroscopic investigation of the resolution for a single Au sphere (Ø = 50 nm, (a)) and an edge between Si and air (b) underneath 30 nm Si$_3$N$_4$. Red arrows mark the wavenumbers revealing the best resolution for the case in (b). The real (black solid line) and imaginary part (red dashed line) of the dielectric function of Si$_3$N$_4$ are plotted in (c). Red arrows mark the wavenumbers for which the superlensing condition is—corresponding to this data—met [23] and therefore the best resolution is expected. Note that (a) has been evaluated from the s$_3$-signal, (b) from the s$_2$-signal. [Dielectric data taken from [33].]

A quite weak dispersion in the region of interest, even a small difference in Re[$\varepsilon_{\text{Si}_3\text{N}_4}$] might lead to the big shift observed for the Si/air edge. The spot size of the single Au sphere differs for varying wavenumber, too, but such distinct minima cannot be seen in this case. This effect can be explained the following way: The structure underneath the membrane couples the best to spatial frequencies proportional to the inverse of its structure size. The imaging mechanism therefore works by far more efficiently if these corresponding spatial frequencies are still transferred through the membrane. Smaller spatial frequencies penetrate deeper into a sample. A cutoff for the transferred spatial frequencies of a superlens is usually defined by calculating the drop of the transfer function $T$ corresponding to the transfer matrix method as explained e.g. in [37] below $|T| = 0.5$. The cutoff depends on the membrane thickness $d$ as well as Im[$\varepsilon$]:

$$k^\text{cutoff}_x = \frac{1}{d} \ln \left( \frac{2}{\text{Im}[\varepsilon]} \right)$$

(4)

This usual definition of the cutoff at $|T| = 0.5$ for a superlens cannot be seen as a strict limit for s-SNOM imaging through a superlens. It could be shown already in literature that also higher spatial frequencies can be caught up by the SNOM tip [24]. Therefore, this cutoff has to be seen as an estimate for a minimum structure size for which the effect of improved resolution at the superlensing-wavenumber is observable. It shows up the influence of the imaginary part of the dielectric function and the thickness of the membrane on this structure size. Calculating this for our case ($d = 30\text{ nm}$) and Im[$\varepsilon$] = 1.7 for the superlensing condition with the smallest imaginary part (at $\nu = 1049\text{ cm}^{-1}$) results in a structure size of

$$R^\text{min}_x \sim \frac{2\pi}{k^\text{cutoff}_x} = \frac{\lambda}{8} = 1160\text{ nm},$$

(5)

which is much larger than the size of the single Au particle investigated spectroscopically. This means, that for such small particles no superlensing effect in terms of improved resolution for
the wavenumber corresponding to the superlens condition can be observed, due to the high imaginary part of the dielectric function. In Fig. 6 this influence of \( \text{Im}[\varepsilon] \) is visualized. We performed simulations without including the tip in order to get an estimate for the dependence of the spot size on the dielectric function within a reasonable computation time. In Fig. 6(a) the dielectric case for \( \nu = 1240\,\text{cm}^{-1} \) (\( \text{Re}[\varepsilon] > 0 \)) is compared to the resonant case for \( \nu = 1049\,\text{cm}^{-1} \) (\( \text{Re}[\varepsilon] = -1 \)) in Fig. 6(b). The simulation does not show big differences between both cases. However, if the imaginary part for the superlensing condition is artificially set to a much smaller value of 0.1, as simulated in Fig. 6(c), a coupling to the upper membrane/air interface can be seen and the fields are much more confined to a smaller spot.

In contrast to the small particle, the step-like Si/air edge transfers into a broad range of spatial frequencies. Therefore, it is reasonable that the dips in resolution can be observed for the latter case, but not for the particle. All in all these observations show that for a capping material revealing SPhP resonances within the mid-IR, even if it is not a classical superlensing material with very low \( \text{Im}[\varepsilon] \) at the resonance as e.g. silicon carbide, it might be beneficial for a good resolution to measure at wavenumbers corresponding to the superlensing condition. However, the improved resolution can only be obtained, if the structure contains low spatial frequencies, i.e. is a step edge or is large enough, whereas the minimum size is dependent on the imaginary part of the dielectric function limiting the superlensing capabilities of the material.

8. Summary

In this study the limits of subsurface imaging for small particles have been investigated. For a 50 nm particle the limit in depth for the detection underneath a Si\(_3\)N\(_4\)-layer has been determined to be around 50 nm. For the regime of a dielectric cover layer with positive permittivity, a simplified model calculation has been introduced, giving a very good qualitative agreement and at least for the investigation of different particle sizes also a good quantitative trend for the experimental results. We could show that for small particles close to the resolution limit of the SNOM the observed spot size underneath 30 nm of Si\(_3\)N\(_4\) appears enlarged, whereas for larger particles of about 200 nm the spot size corresponds to the particle diameter. In a spectroscopic investigation we then addressed also the regime of negative permittivity of the cover layer. We observed in experiment and simulations a strong dependence of resolution and signal strength on the actual value of the dielectric function. A contrast inversion for wavenumbers below 960 cm\(^{-1}\), as well as minima in resolution close to the wavenumber positions for the superlensing effect, have been revealed. Those minima are not observable for single 50 nm Au spheres. This gives a hint to a limitation of superlensing regarding small structure sizes given by the cutoff for high spatial frequencies, which can be calculated by the transfer function. The investigation of this limitation is subject to further research.

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