Full simulations of the apertureless scanning near field optical microscopy signal: achievable resolution and contrast

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Abstract: We simulate apertureless near-field optical imaging and obtain phase and amplitude scans of structured substrates for elastic scattering. The solution of the three-dimensional Maxwell equations does not involve approximations and we include large tips and substrates, strong interaction, interferometric detection and demodulation at higher harmonics. Such modeling represents a significant step towards quantitative simulations and offers the attractive possibility to study the individual influence of each relevant experimental parameter. We typically obtain highly localized signatures of the interaction of the tip with gold inclusions, superposed on a slowly varying background signal. The relative importance of both contributions and the achievable lateral resolution are strongly dependent on the geometry and scanning conditions. The simulations show sensitivity mostly to the first nanometers of the sample and underline the importance of scanning near the sample and being careful with mechanical anharmonicities on the tip oscillation. They also help to determine the influence of oscillation amplitude and demodulation harmonic.

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1. Introduction
Among the techniques developed in recent years to obtain optical lateral resolution beyond the diffraction limit [1, 2, 3, 4] apertureless scanning nearfield optical microscopy (aSNOM) is particularly promising [5, 6]. It is fundamentally surface-sensitive and has been shown to give optical information with lateral resolution even below the 10nm range [7, 8]. Usually, a sample is externally excited, and the radiation scattered by a probing tip carries local information about the volume in the proximity of its apex. Experimental work shows clear images for a wide array of sample types [9, 10, 11].

A fundamental challenge to obtain, optimize and interpret experimental images is the dependence of the measured signal on various experimental parameters, not all of which are necessarily known or well controlled. In this context, numerical simulations offer an opportunity to define and change relevant conditions at will and are useful for a better understanding of the phenomena involved. Solving the Maxwell equations under conditions which are found in many experiments is, however, challenging: the substrate can be complex, the size and shape of the tip can influence the results [12, 13, 14], strong interaction [15, 16, 17, 18, 19] between tip and sample is common and while the tip and the substrate can be large relative to the excitation wavelength, other parameters of importance such as the tip-substrate distance can be
in the nanometer range. Further, the way in which the signal is detected has been observed to be critical; very significantly for elastic information, demodulating the signal at the higher harmonic of the frequency of oscillation \([20, 21]\) can enhance the contributions described by fast spatial variation, i.e. those enabling high resolution \([22]\). Interferometric detection allows to obtain both phase and amplitude information.

Much of the theoretical understanding of aSNOM has been gained by reasoning with one of two simplifying assumptions. The first considers a passive – or weakly perturbing – tip that serves to scatter the fields present in the sample \([23, 24, 25, 26]\). We note that as far as the effect of the tip on the field distribution is not critical, relevant results can also be drawn from work not specific to aSNOM discussing the spatial behavior of near fields in general. The localized nature of the near fields, which allows to obtain high lateral resolution \([27]\), the convenience to scan near the sample \([28, 29]\), the influence of optical and topographical contrast \([30]\) and the effect of higher harmonic demodulation \([31, 22]\) have all been studied. The second alternative assumes a tip that strongly interacts with a flat substrate. Under such conditions, the signal is understood as probing the strength of the interaction, and consequently the dielectric constant of the surface \([32, 6]\).

For interpreting measurements, however, one would desire to understand in detail the origin of the optical signal in a general scenario, which is not restricted to one of the limiting cases discussed above; a tip scans a sample of complex topography and dielectric properties, and can interact more or less strongly with the sample according to position. First studies have already started to tackle some of the questions of interest \([33, 34, 35]\), but simulating the conditions of typical experiments is still highly challenging. Here, we aim to improve the understanding of aSNOM by studying in detail the process of image formation for models close to experiments under general conditions. In particular, we solve for elastic scattering the three-dimensional Maxwell equations simultaneously including patterned samples, elongated tips, strong tip-sample interaction, interferometric detection and higher harmonic demodulation. We use our simulations to discuss the need of including long tips for realistic simulations and to quantitatively study the dependence on relevant experimental parameters of key aspects of aSNOM imaging: signal strength, resolution, background suppression and contrast between areas of different physical properties.

2. Model

For the simulation, we use the MaX-1 software package, a program based on the multiple multipole method \([36, 37]\). It uses multipole and other field distributions that verify the bulk Maxwell equations and serve as expansion sets for the resulting electromagnetic field distributions. The expansion coefficients are determined by a minimization of the error in the boundary conditions. In our case, we keep the average error smaller than 0.5 percent and verify that increasing the number of free parameters in the expansions by \(\sim 25\) or \(\sim 50\) percent does not significantly affect the discussed results.

Figure 1 sketches the simulated geometry. For the environment we assume vacuum \((\varepsilon_0 = 1.0)\). Unless mentioned otherwise, we concentrate on a 500\(nm\) long conical tip of 10 degree opening angle, capped with two hemispheres. The tip apex has radius \(R = 10nm\). The tip is made of silicon \((\varepsilon_{Si} = 17.76 + 0.508i[39])\) and it oscillates at an angular frequency \(\Omega\) along the \(z\) axis, simulating the motion of a non-contact mode atomic force probe along this axis.

The substrate is a spherical gold inclusion of 10\(nm\) radius at a small distance \(d\) to the surface of a much larger glass sphere, of 2\(\mu m\) diameter \((\varepsilon_{Au} = -3.95 + 2.58i[38], \varepsilon_{glass} = 2.1)\). The line joining the centers of both spheres is parallel to the tip axis. The point of the glass-vacuum interface exactly above the center of the inclusion is the origin of the coordinate system used to facilitate the discussion. The small gold inclusion facilitates illustration of the excel-
lent lateral resolution that is one of the main justifications for aSNOM. Changing the depth of the inclusion $d^{(i)}$ also opens a natural way to study how far the tip probes into the substrate. The radius of the glass substrate is large enough to be locally almost flat at the region of interest. We used a curved substrate to avoid numerical convergence difficulties when evaluating the scattered fields at the detector. Both the size of the large substrate sphere and of the tip length, comparable to the wavelength, represents a reasonable compromise between numerical requirements and the desire to resemble typical experimental conditions. We have discussed previously the influence of changing the tip length and substrate radius [19].

A p-polarized planar wave of wavelength $\sim 514.5\text{nm}$, propagation vector $\mathbf{k}$ and electric field $\mathbf{E}$ illuminates the sample at an incident angle of $70^\circ$ with respect to the tip axis. We calculate the fields from the large sphere in isolation and from the complete tip-substrate system; we subtract the former from the latter to concentrate on the contribution from the tip and the inclusion. Simulating focused illumination is useful to avoid scattering from the top cap of long tips [19]. However, the radius of the illumination focus for typical aSNOM experiments under lateral illumination can easily be significantly bigger than the tip length in this paper, so that the gaussian illumination is similar to a planar wave in the region of interest. We thus consider the simple planar wave illumination to be sufficient for the tip lengths and purposes of the present paper.

A lens of numerical aperture $\sim 0.342$ situated at $1\text{mm}$ distance in the back scattering direction collects the scattered fields. We ignore any effects of further optical elements or propagation on

Fig. 1. Illustration of the geometry used. The illumination is modeled as a planar wave, and for collection an aplanatic lens situated at $1\text{mm}$ distance is considered. A reference beam is used for amplified, interferometric detection. The upper point of the glass substrate serves as the origin of the coordinate system.
the collected fields until they impinge normally on a photodetector, but we do include interferometric mixing with a collinear reference planar wave polarized in the \(xz\) plane (p polarized). The lens is an idealized aplanat, that is, a non-absorbing, non-reflecting converter of a spherical waves centered on \(x = z = 0\) into a beam of planar phase front propagating towards the photodetector.

Further, we consider the effect of a dual-phase lock-in amplifier operating at angular frequency \(n\Omega\), which subsequently gives the amplitude and phase of the \(n\)-th harmonic of the signal \(H_n\) generated by the photodetector. Assuming the reference beam used for interferometric detection much stronger than the fields scattered by the tip and sample, we write [40]

\[
H_n \approx \frac{e^{i\phi_i}}{2T} \int_{0}^{T} e^{i n \Omega t} \int_{S_{det}} \mathcal{E}^*(r, x, d^{(ts)}(t)) d^2 r dt
\]  

\(\mathcal{E}^*(r, x, d^{(ts)}(t))\) describes the magnitude and phase of the complex conjugate of the scattered electric field amplitude at the photodetector, for the p-polarized component. The presence or not of complex conjugates in the terms of Eq. (1) depends on the exact experimental implementation [40] but is of little importance here. \(\mathcal{E}^*\) is a function of the area element location \(r\) and tip position at a given time \((x, d^{(ts)}(t))\). If not mentioned otherwise, the tip oscillates along \(z\) as \(d^{(ts)}(t) = d_{\text{min}} + A [1 + \cos(\Omega t)]\), where \(A\) is the amplitude of the oscillation and \(d_{\text{min}}\) the minimum tip-substrate distance, constant during a given scan. \(S_{\text{det}}\) is the area of the detector where the fields are non-zero, as defined by the numerical aperture of the lens, and \(\phi_i\) allows to consider the internal phase of the lock-in amplifier or the phase of the reference beam, which only enter a constant offset to the phase of the signal. To simulate a scan along \(x\), we evaluate Eq. (1) by varying \(d^{(ts)}\) for each \(x\) position of the tip. That is, we consider the tip being displaced over a fixed substrate rather than the sample under a horizontally fixed tip, which may be advantageous in experiments using focused beam excitation [8]. For the plane wave excitation assumed here, the difference is a simple phase shift proportional to the tip position. The units of the obtained amplitude signal are arbitrary, as they depend on the reference signal power and photodetector sensitivity, but they are kept constant throughout the manuscript.

Generally, the simulated \(|H_n|\) scans exhibit a component slowly varying across the inclusion on a scale significantly larger than the inclusion and a peak-like signature confined to small tip-inclusion distances. For the purpose of this paper, we refer to the former as background, whereas the latter we associate with signal due to tip-inclusion interactions. We discuss the images in terms of the peak height (absolute and relative to the background level) and the full width half maximum \(\text{FWHM}\) of the central peak. Thus, to calculate the \(\text{FWHM}\) we first subtract from \(|H_n|\) an estimation of the background, given by the linear interpolation between the values at \(x = -39\, \text{nm}\) and \(x = 39\, \text{nm}\).

3. Results

3.1. General characteristics of the obtained signal

Figure 2(a) illustrates how the tip interacts with the substrate. Strong near fields appear between the tip and the closely situated gold inclusion, significantly more intense than between the tip and the glass. For more qualitative information, the insert shows the normalized value of the near field maximum in the proximity of the apex for separation \(d_{\text{min}} = 1\, \text{nm}\) and different tip positions along \(x\). The near fields are significantly stronger when the tip is close to the gold inclusion than to the glass substrate, as expected [41]. However, also in the insert, no corresponding far field signature appears clearly in the plotted behavior of \(|\int_{S_{\text{det}}} \mathcal{E}(r, x, d^{(ts)}) = 1\, \text{nm} |d^2 r|\) before demodulation.

A representative example of one-dimensional scans \((d_{\text{min}} = 1\, \text{nm}, d^{(i)} = 1\, \text{nm}, \text{and} A = 10\, \text{nm})\)
Fig. 2. (a) Example of the field distribution of the average scattered electric field in the proximity of the tip apex, for 500nm long tip and $d^{(i)} = d^{(j)} = 1\text{nm}$. The colors indicate values between 0 and 15, where the excitation field strength is 1. The insert indicates the normalized value of the near field maximum in the proximity of the apex (continuous blue) and the normalized amplitude of the interferometric term of the signal before demodulation (dashed red), for 1nm distance to the substrate. (b,c) Examples of the obtained amplitude (b) and phase (c) for one-dimensional scans of the samples, for the first (circles), second (dashed) and third (continuous) harmonics, the geometry described in (a), $d_{\text{min}} = 1\text{nm}$ and $A = 10\text{nm}$. For better visibility, the first harmonic amplitude is scaled by 1/5.
at the first three harmonics is plotted in Fig. 2(b,c). Not plotted, the continuous component 
\(|H_0|\) gives a signal \(\sim 25\) times larger than the first harmonic and similar in shape to the signal 
before demodulation \(\int_{S_{det}} \delta(r,x,d^{(ts)}) = 1nm)d^2r\) shown in the insert of Fig. 2(a). Looking 
at the amplitude of the detected signal, a clearly stronger signal near the inclusion appears 
at the second and third harmonics, but not in the first. The signal is stronger for the lower 
harmonics, but the contrast between the maximum and the signal for \(x \gg R\) diminishes. The 
width of the maxima is much smaller than the wavelength, which confirms the possibility of 
lateral resolution much below the diffraction limit. In Fig. 2(b), the \(FWHM\) of the second and 
third harmonic is \(\sim 21nm\) and \(\sim 17nm\), respectively.

Our model is able to simulate both amplitude and phase information of the demodulated 
complex optical field strength [42] as is often recorded in experimental work. Here we note 
that we have found the detected phase (Fig. 2(c)) at second and third harmonic demodulation 
to also reveal local extrema near \(x = 0\). As for the amplitude, the phase signature appears more 
pronounced in the third harmonic than the second. Notice that in accordance to our discussion 
of Eq. (1), the sign of the phase –and thus the character of the extrema as minima or maxima– 
depends on the particular experimental implementation. For the remainder of this manuscript 
we concentrate on amplitude signals exclusively.

3.2. Tip length and inclusion depth

Theoretical aSNOM studies often model small tips, and we briefly consider in Fig. 3 conse-
quences of such an assumption for conditions otherwise identical to Fig. 2(b). For a tip modeled 
as a sphere of 10nm radius (equal to the apex radius of the long tips), the signal is consider-
ably weaker in Fig. 3, an effect especially pronounced for the first harmonic, where the strong 
scattering from the bulk of long tips is more relevant [19]. The central maximum dominates 
the obtained signal even for the first harmonic and is not situated at \(x = 0\), i.e., the asymme-
try introduced by the illumination has a measurable influence. Notice that in the long tip case 
(Fig. 2(b)), the second and third harmonic signal were much more symmetric. The maxima 
are also narrower for the small than for the long tip, with the \(FWHM\) at the third harmonic 
\(\sim 15 – 20\) percent smaller than before.

Back to the long tip case, we consider in Fig. 4 the influence of the depth of the inclusion, 
\(d^{(i)}\) [41, 43]. When \(d^{(i)}\) is increased, the amplitude of the signal remains approximately constant 
for \(|x| \gg R\) but rapidly diminishes at the central maximum. For \(d^{(i)} = 5nm\), the maximum peak 
signal after substracting the background is approximately three times weaker than for 1nm. The
Fig. 4. Influence of the depth of the inclusion, for the third harmonic, on the amplitude of the signal for 1-dimensional scans. 500nm long tip, oscillation amplitude 10nm and $d_{min} = 1nm$ are considered.

Fig. 5. Influence of the oscillation amplitude on the obtained signal amplitude for the second (a) and third (b) harmonics using 500nm long tips. $d^{(i)} = 1nm$ and $d_{min} = 1nm$. The insert in (b) shows the influence of the oscillation amplitude on the FWHM of the central maximum.

FWHM increases for the larger values of $d^{(i)}$, $\sim 17$ and $\sim 21nm$ for 1 and 5nm, respectively.

3.3. Scanning parameters

We consider next the influence of the scanning parameters on the resulting scans, starting with the oscillation amplitude $A$. A Taylor expansion of the signal with respect to $z$ demonstrates that, in the infinitesimal amplitude case, the amplitude of the $n$-th harmonic signal is proportional to the $n$-th power of the oscillation amplitude [44]. For the second harmonic central maximum and amplitudes varying from 2nm up to values on the order of the apex radius, Fig. 5(a) shows a dependence weaker than quadratic but still important. For much larger values the main effect is a shift of the whole curve towards larger signal, diminishing the contrast between the sig-
Fig. 6. Influence of $d_{\text{min}}$ on the signal amplitude for 1-dimensional scans using 500 nm long tips at the second (a) and third (b) harmonics. $d(t) = 1 \text{ nm}$ and the oscillation amplitude is $10 \text{ nm}$. The insert in (a) shows the dependence of the signal on $d_{\text{min}}$ for $x = 0$ and $x = -25 \text{ nm}$.

nal at the maximum and at values of $|x| \gg R$. A similar trend characterizes the third harmonic (Fig. 5(b)). The main difference in comparison to the second harmonic is that values of oscillation amplitudes as large as three times the apex radius still result in a clear central maximum. The insert demonstrates that increasing the amplitude also affects the FWHM [22, 45]. In our case, it is $\sim 60$ percent larger for $A = 20 \text{ nm}$ than for $2 \text{ nm}$.

Figure 6 shows, for the second (a) and third (b) harmonic, the influence of changing the minimum distance between tip and substrate $d_{\text{min}}$. Increasing $d_{\text{min}}$ on the order of half the apex radius strongly reduces the signal level, most significantly at the maximum. To further illustrate this point, the insert in Fig. 6(a) shows the evolution of the second harmonic signal for a continuous change in $d_{\text{min}}$, for the tip over the inclusion ($x = 0$) and relatively far away ($x = -25 \text{ nm}$). Values $-39 \text{ nm} > x > -25 \text{ nm}$ result in similar traces, indicating that the influence of the inclusion in this range is small. The signal increase for short $d_{\text{min}}$ is especially significant when the tip interacts strongly with the inclusion, but it is also present for $x = -25 \text{ nm}$, where the tip mostly interacts with the bare glass substrate. Increasing $d_{\text{min}}$ also increases the FWHM (In Fig. 6(a), from $\text{FWHM} \sim 21 \text{ nm}$ at $d_{\text{min}} = 1 \text{ nm}$ to $\text{FWHM} \sim 25 \text{ nm}$ at $d_{\text{min}} = 4 \text{ nm}$).

Up to now, a perfect sinusoidal movement was assumed to describe the tip oscillation. Figure 7 considers the imaging process for a tip movement along $z$ according to $d(z) = d_{\text{min}} + A[1 + b + \cos(\Omega t) + b \cos(3\Omega t)]$, where $b$ is a parameter used to change the deviation from a perfectly harmonic motion. Already for a value of $b$ of 1 percent, the change on the third harmonic signal, in particular for $|x| \gg R$, is not negligible. For a value of $b$ close to 10 percent the change is dramatic, with a much less clear central maximum. Notice that a large signal distortion of the signal could ensue if the strength of the anharmonicity changes during the scan. Such effects are of mechanical [46, 47, 44], and not optical, origin, and are generally undesired for aSNOM measurements.

4. Discussion and conclusions

Using patterned substrates, we have been able to simulate amplitude and phase images after higher harmonic demodulation. Demonstrating that phase images can be simulated is relevant
Fig. 7. Influence on 1-dimensional scans of the presence of anharmonicities (described by $b$) in the movement of a 500nm long tip. The traces correspond to the amplitude of the third harmonic signal, for $d^{(1)} = 1\, \text{nm}$, $d_{\text{min}} = 1\, \text{nm}$ and 10nm oscillation amplitude.

for aSNOM studies in which the phase detected at the lock-in amplifier gives additional information about the sample [10, 48]. As in most aSNOM studies, however, the emphasis here is on the amplitude of the final signal.

The choice of tip material serves to illustrate the possibility of aSNOM imaging with silicon tips, which can be very sharp. Indeed, commercial tips of apex radius $< 10\, \text{nm}$ are readily available. As far as the tip-substrate interaction is strong enough for discrimination of the resulting signal to be possible, we expect the tip material to affect the signal strength but be of little qualitatively significance to the general behavior. Only for sharp resonances significant spectral shifts could appear as the tip moves close to the substrate [16], modify the appearance of the simulated images, and complicate their interpretation.

Before demodulation, no clear signatures from the inclusion appear. In contrast, the typical signal after higher harmonic demodulation can be conceptually seen as a narrow maximum appearing when the tip is close to the inclusion, riding on a smooth background signal. It seems intuitive to associate the peak with a stronger tip-substrate interaction (continuous line in the insert in Fig. 2(a)). If extended to more complex scenarios, a direct connection may be drawn between the demodulated signal and the interaction strength.

The FWHM is closely related to the point spread function of conventional resolution studies and a small value is a strong indicator of good lateral resolution. We find that a FWHM as low as $\sim 12.5\, \text{nm}$ is possible at the third harmonic for small oscillation amplitude, less than $\lambda/40$ and comparable with both the tip and sphere radius. For comparison, a topography image of a $10\, \text{nm}$ radius hemisphere scanned by an AFM tip with $10\, \text{nm}$ apex radius results in FWHM more than twice larger. Notice that both the structure size and the tip apex radius – identical here – are relevant length scales, even if for simplicity we sometimes only refer to the latter.

The images are clearly dependent on the scanning conditions. Close proximity between the tip and the inclusion is convenient for strong interaction and small FWHM. A perfect sinusoidal movement of the tip is also advantageous for experiments, at least as long as the set-up is not capable of separating optical and mechanical contributions [44]. However, both desires are not completely independent in a realistic experiment: strong tip-substrate contact as a consequence of an attempt to decrease $d_{\text{min}}$ risks introducing anharmonicities in the tip movement and also fast wear of the tip apex during scanning [47, 44].

It does not seem possible to give an optimal oscillation amplitude and demodulation harmonic valid for all experimental conditions. A small amplitude and high harmonic reduces the FWHM and results in clearer maxima, but it also considerably diminishes the signal level. Too soft a cantilever with too small an amplitude can also result in jump-to-contact, that is, un-
desired mechanical anharmonicities [49, 50]. Knowing the noise level and other experimental parameters is thus important for a reliable optimization. Notice that if we select the amplitude to obtain a given signal strength, going towards ever higher harmonics is not necessarily favorable. For example, if $d_{\text{min}} = 1\,\text{nm}$ and $d^{(i)} = 1\,\text{nm}$, imaging at the second harmonic with $5\,\text{nm}$ oscillation amplitude results in a similar image as $20\,\text{nm}$ oscillation amplitude at the third harmonic, except that the FWHM is a $\sim 10$ percent smaller. As a coarse rule, second and third harmonic demodulation with amplitudes on the order of the apex radius seem promising.

For the geometry discussed here, the signal is mostly sensitive to the region close to the interface. aSNOM measurements sensitive to larger depths have been observed [51]. The structures imaged in that work were significantly bigger than in the present study, which adds further phenomena such as a modified substrate reflectivity [52, 53]. Larger apex radii could equally increase the sensitivity to larger depths.

We have also illustrated limitations of small tip models for simulating typical experimental conditions. The obtained signal under such simplifications is considerably weaker and predicts an easier discrimination of the signature from the tip-inclusion interaction than appears possible in full tip models [14, 19]. Further, the central maximum for the small tips presents a much clearer asymmetry with respect to $x = 0$ than for the $500\,\text{nm}$ long tips. The asymmetry is introduced by the oblique illumination and the reduction for long tips could be due to the additional symmetry introduced by their axis [40]. Last, a better FWHM is observed for the short tips, which points to a more spatially confined interaction [54] and reminds of the larger field confinement for isolated tips [40].

In summary, we have been able to simulate the phase and amplitude imaging process for aSNOM, including long tips and high harmonic demodulation, and shown the limitations of small tip models. The signal demonstrates subsurface imaging of small metallic particles if they are situated at nanometer distance to the interface. The signature of such inclusions is clear and strongly localized on the order of the structure size. The scanning conditions affect the obtained images, and higher harmonic demodulation, amplitudes on the order of the tip radius, scanning close to the sample and perfectly harmonic oscillation all seem favorable. Extending the present work to different substrates and tips will be helpful to guide experimental work and image interpretation by direct comparison between theory and measurements.