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Measurement of the decay lengths of the near-field signal in tapping mode

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1. Introduction

The characterization and the imaging of nanostructures and of fields strength above them, are becoming a domain of interest in physics, chemistry, biology, and the data storage [\[1–4\]](#page--1-0). The apertureless scanning near-field optical microscopes (ASNOM) allow the detection of the optical field at a few nanometers above the nanostructures. The ASNOM techniques are based on the detection of the variations of the electromagnetic signal scattered during the scan of a subwavelength-sized probe above the investigated sample. Due to the nanometric size of the probe end, the nanostructure and the confinement of the light, the signal-to-noise ratio obtained from such a configuration is usually low. The ASNOM spectroscopic studies are almost always required in order to produce a valuable optical signal. However, neither Raman nor fluorescence signals are large enough to perform lock-in techniques (instead, photon counting is essential [\[5,6\]\)](#page--1-0). Nevertheless, there are applications where homodyne or heterodyne techniques are used: with polarized light, or with multiple wavelengths (in sequential imaging) [\[7,8\]](#page--1-0) or if the near-field strength has to be investigated. In this last case, performing the reconstruction of the data is of interest in order to get knowledge of the vertical variations of the near-field.

The homodyne technique consists in recording the first Fourier harmonics of the signal, by locking the detection on the vertical

ABSTRACT

In near-field optics, the use of vertical vibration of the probe is of great interest in order to prevent the tip crash, in tapping mode. The optical signal is often obtained through a lock-in amplifier using a feedback on this vertical vibration. Therefore, harmonics of the optical signal are available. The reconstruction of the near-field signal, from these harmonics, enables the knowledge of the vertical variations of the near-field signal, without the use of the slower technique of approach curves or the use of photocounter. In this work, we use this reconstruction to measure the vertical decay lengths of the near-field data with the Prony's and the simplex methods. We compare both methods and we discuss the results in terms of filtering by the lock-in.

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vibration of the probe (for example). The investigation of a single harmonic is not sufficient to get physical information such as contrast [\[9,10\]](#page--1-0). Obviously, considering only one Fourier's harmonic of a signal is not significant when the whole signal must be studied. Neglecting the lock-in (as well as the probe) in the image formation process may be hazardous especially because the recorded data depend on the amplitude of vibration of the probe and the corresponding filtering may lead to a contrast reversal by varying this last parameter [\[11\].](#page--1-0) Fortunately, a reconstruction of the ''real" optical signal, from all the available harmonics is possible and helpful to discuss the contrast of the data [\[12,13\]](#page--1-0). Consequently, any signal processing should be applied after the reconstruction of the near-field optical signal [\[14,15\].](#page--1-0) Moreover, the reconstruction gives the variations of the near-field signal along the vertical vibration of the probe and therefore, a measurement of the decay length of the near-field can be deduced. An experimental measurement of the decay length has been recently driven with the Prony's fitting method [\[16\].](#page--1-0) In this reference, it has been shown that it was impossible to connect the decay lengths to the properties of the object, consecutively to the strong interaction of the light, with the probe end and with the sample.

In this work, we introduce the filtering of the lock-in, we detail the method of reconstruction and we discuss on the ability of measuring the decay lengths of the near-field signal from a theoretical point of view. The decays lengths are computed from an appropriate fit of the reconstruction. For this purpose, we compare two numerical methods to recover the exponential decay of a complex signal. The first one is the exponential non linear fit (the Prony's

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method) and the other one is an exponential fit using the simplex optimization method. We illustrate the filtering properties of the lock-in with the concept of transfer function.

2. The lock-in filtering properties

In this study, we consider a vertical vibration of the probe, working in tapping mode, with amplitude A, at frequency f, and therefore, the distance z between the probe and the sample can be written as: $z(t) = A[1 - \cos(2\pi ft)]$, assuming a harmonic vibration. $z(t)$ varies between 0 and 2A. In the computations, we consider a period of vibration, for $t\in [-1/2f; 1/2f]$ or z varying from 2A to 0 and then to 2A. The recorded signals through the lock-in detection are the amplitudes and phases of the first Fourier harmonics H_0, H_1, \ldots, H_N of the detected field $\mathcal{S}(x, y, z)$ during the scan of the probe in the xy-plane [\[12–14\].](#page--1-0) H_0 is the constant term delivered by the lock-in, is usually noisy and not recorded, but must be conserved for the reconstruction.

To illustrate the ASNOM ability to measure decay lengths, we first suppose a signal composed of pure exponential decreases, with an increasing decay length D_p along $x:\mathscr{S}(\mathsf{x},z)=\mathsf{exp}[-z/D_p(\mathsf{x})].$ D_p is the decay as the tip-sample distance z increases. Fig. 1a shows

Fig. 1. (a) Theoretical signal: series of exponential decreases with increasing decay lengths. (b) Computed lock-in data: the harmonics are normalized for clarity and logscale are used in abscissa.

the variations of $\mathcal{S}(x, z)$ as a function of z/A and the normalized decay D_n/A .

Fig. 1b shows the theoretical simulation of the lock-in data. H_0 corresponds to the mean value of the signal along z and therefore, is increasing with D_p (high-pass behaviour). The other harmonics are band-pass for D_p/A . The shape of H_i exhibits a maximum which depends on the order i of the harmonic. Indeed, the signal observed in harmonics is only a limited part of the whole near-field signal. The maximum of harmonics $H_1, \ldots, 7$ occurs for $D_p/A = 0.66$, 0.21, 0.10, 0.058, 0.040, 0.028, 0.019, respectively. This effect can be interpreted as a maximum of sensitivity of each harmonic for a given D_p/A or as a filtering effect of the lock-in. The bands of each harmonic are overlapping each other and therefore, the near-field information appears in all harmonics.

The large ratio D_p/A are blocked by the lock-in and will not be measured. This is commonly considered as an advantage of the lock-in: the useless background, which varies slowly, is rejected. Actually, also near-field is rejected. For example, the exponential decay with $D_p = 2A$ will be attenuated in H_1 by a factor 0.6 and by a factor lower than 0.2 in higher harmonics. Typically, the AS-NOM using $A = 18$ nm as in Ref. [\[13\]](#page--1-0) is not able to measure $D_p > 36$ nm. Consequently, decreasing the amplitude of vibration induces a filtering of the near-field as well as of the far field. On the other hand, increasing the harmonics N enables the detection of short decay lengths. Nevertheless, the band-pass behaviour of each harmonic induces a ''cut-off frequency" for the detection of small D_p . More generally, the finite number of recorded harmonics and the finite amplitude of vibration of the probe, limit the resolution power of the ASNOM for both the small and the large D_p . Moreover, the harmonics considered independently, cannot enable the measurement of decay length as their sensitivity is a nonmonotonic function of D_p/A . Hopefully, the reconstruction can help.

3. The reconstruction of the detected signal

The detected signal as well as its reconstruction from N harmonics H_i , are even functions of time. The reconstructed ASNOM signal R_N can be expressed as a function of the position of the probe z or as a function of time t [\[13\]](#page--1-0):

$$
R_N(x, y, t) = \sum_{n=-N}^{N} H_n(x, y) \cos(n2\pi ft)
$$
 (1)

$$
R_N(x, y, z) = \sum_{n=-N}^{N} H_n(x, y) \cos \left[n \arccos \left(1 - \frac{z}{A}\right)\right]
$$
 (2)

$$
=\sum_{n=-N}^{N}H_{n}(x,y)T_{|n|}\left(1-\frac{z}{A}\right),\tag{3}
$$

where T_n are the Chebyshev's polynomials. R_N is therefore, an approximation of the approach curve obtained directly from all the data obtained from the lock-in [\[14,17\]](#page--1-0). No a priori information is required and no restrictive hypothesis is made on the image formation. The reconstruction may be considered as an alternative to decrease the influence of the lock-in. If a low pass filtering is needed to decrease the far-field contribution, it must be applied after reconstruction [\[15\]](#page--1-0) to keep all the near-field information (Fig. 1b). Theoretically, an infinite number of harmonics is necessary to perfectly reconstruct the near-field and the accuracy of the reconstruction increases with the number of recorded harmonics. The [Fig. 2](#page--1-0) illustrates the reconstruction of the signal showed in Fig. 1, with only one harmonic (a), two harmonics (b), three (c) and four (d), respectively.

The short exponential decays cannot be recovered, but the reconstruction R_4 seems to give the best results, as expected. A first visual investigation of this figure reveals that R_1 is enough to

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