Reduction of tip–sample interaction forces for scanning near-field optical microscopy in a liquid environment

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Abstract

Tip-to-sample shear force distance control of a scanning near-field optical microscope (SNOM) is more critical to implement in a liquid environment than in air. The higher viscosity of the medium increases the interaction forces between the tip and the sample and may even damage the sample in the case of soft materials. In this work we measure the decrease of the quality factor of a vibrating fiber when it is immersed in water. The main loss of quality is already observed for situations where the part of the fiber dipped in the water is only a few tens of microns long. We propose a new experimental set-up which maintains a small (< 50 µm) and stable depth of the fiber in water, independent from the tip to sample distance. The quality factor of the fiber tip is then $Q = 52$, only five times smaller than in air. Topographical and near-field optical images of a soft hydrogel are obtained without inducing any damage on the observed material. Such images are impossible to obtain with a SNOM tip totally immersed in water, due to the very soft surface of the hydrogel which contains 70% of water. © 1998 Elsevier Science B.V. All rights reserved.

1. Introduction

Optical microscopy is essential for observing specimens in a liquid environment. For a biological sample, the aqueous environment is necessary in order to study, e.g., the structural basis function of a cell or supra molecular complexes. However, the highest-resolution achievable with conventional (far-field) optical microscopy is typically several hundreds of nanometres. Electron microscopes offer much finer resolution, but do not allow observation in aqueous media. Scanning near-field optical microscopy (SNOM) has the unique ability of a resolution below 100 nm while using the contrast methods (fluorescence, polarization, phase, etc.) of standard optics. Recently, SNOM has been used to observe samples in a liquid environment. The control of the distance between the tip and the sample, which is very critical in order to get a good spatial resolution, is achieved either by simultaneous atomic force microscopy (SNOM-AFM) [1] or by the shear force method [2,3]. Both methods are based on the attenuation of the vibration of a cantilever or a fiber when the tip is in close vicinity of a sample. The main point addressed in this paper concerns the higher interaction forces produced by
the larger viscosity of the liquid medium and how to minimize these forces in order to avoid damage to soft samples during observation.

2. Vibrating fiber tip: calculation of the drag force

To understand better and quantify the interaction force between the tip and the sample, a simple model is briefly presented for the vibration of the tip, more details can be found in standard text books [4]. The fiber tip can be considered as a cylindrical cantilever which acts as a spring, with a static spring constant $k_{\text{stat}}$. When the fiber tip is excited at a frequency $\omega$, the motion $u_L(t)$ of its extremity is governed by the equation

$$m_0 \ddot{u}_L + F_D + k_{\text{stat}} u_L = F \exp(\text{i} \omega t) + \text{c.c.},$$

where $m_0$ is an effective mass to be determined and $F$ the mechanical driving force imposed to the fiber. $F_D(t)$ represents a drag force opposing the movement of the tip. This drag force is assumed to be the sum of the forces resulting from the tip-sample shear force interaction, $F_{\text{int}}$, and the viscous losses inside the fiber, $F_k$, or produced by the external medium, $F_M$ (air or liquid). Assumption is made here that this drag force is proportional to the instantaneous velocity of the tip and can be expressed by a phenomenological parameter $\gamma$ as

$$F_D = F_k + F_M + F_{\text{int}} = m_0 \gamma \dot{u}_L.$$

Note that when the tip and the sample are in close vicinity, non-viscous interaction forces can influence the vibration of the tip and also slightly modify the spring constant $k_{\text{stat}}$. These effects are, however, neglected in the following discussion. The solution of Eq. (1) is

$$u_L = \frac{F_0 \delta_0}{k_{\text{stat}}(\omega_0^2 - \omega^2 + \text{i} \gamma \omega)} \exp(\text{i} \omega t) + \text{c.c.},$$

where $\omega_0 = (k_{\text{stat}}/m_0)^{1/2}$ is the resonance frequency of the harmonic oscillation of the fiber tip. $|u_L|$ is a Lorentzian shaped peak function of frequency with a maximum at $\omega = \omega_0$. Using the measure of the full-width at half-maximum (FWHM), $\Delta \omega$ of this peak function, we can define a quality factor, $Q = \omega_0/\Delta \omega = \omega_0/\sqrt{3\gamma}$ for the vibration of the tip.

By combining Eqs. (2) and (3) we get a simple expression for the amplitude of the drag force $|F_D|$ when the fiber is oscillating at the resonance frequency:

$$|F_D| = \frac{k_{\text{stat}} |u_L|}{\sqrt{3Q}}.$$  \hspace{1cm} (4)

This equation shows that the drag force $|F_D|$ is proportional to the spring constant $k_{\text{stat}}$ and to the amplitude of vibration $|u_L|$ of the fiber tip, whereas it is inversely proportional to the quality factor, $Q$. By measuring $Q$ and $|u_L|$ when the tip is far from the sample, one can calculate the sum $F_k + F_M$ of the drag forces coming from the internal losses and from the friction of the surrounding medium (air or water). The tip-sample interaction shear force, $|F_{\text{int}}|$ can then be obtained by measuring the attenuated amplitude of vibration $|u_{\text{int}}|$:

$$|F_{\text{int}}| = \frac{k_{\text{stat}} (|u_L| - |u_{\text{int}}|)}{\sqrt{3Q}}.$$  \hspace{1cm} (5)

Eq. (5) allows to deduce a quantitative value of the interaction force between the tip and the sample, even though its nature is not exactly known. The only assumption made here, is that it is proportional to the instantaneous velocity of the tip and does not depend on the instantaneous position, $u_L(t)$ of the tip. With this assumption, the system remains harmonic.

3. Measurement of the quality factor of a tip immersed in water

In order to calculate the magnitude of the drag force exerted by the liquid environment on the tip, measurements are performed of the amplitude of vibration of the fiber tip versus the excitation frequency for different depths $d$ of the tip in water. Results are displayed in Fig. 1. The tip has a length $L = 3.3$ mm, the diameter of the fiber is $D = 125$ $\mu$m. The vibration amplitude is measured by diffracting a probe laser beam onto the fiber [5,6].

The data of Fig. 1 can be fitted by Eq. (3), which allows to determine quality factors. $Q = 240$ is obtained for the tip in air, it falls to the value $Q = 52$
when the tip is immersed by only $d = 50 \mu m$ in water. The shoulder near the maximum of the curve for the tip in air is due to a second-resonance peak. This shoulder is not apparent anymore when the tip is immersed in water and the data is well fitted by Eq. (3). The amplitude of vibration $|u_L|$ decreases in the same proportion as the quality factor. If one assumes an amplitude of vibration of $|u_L| = 0.5 \text{ nm}$, (detected by our experimental system with a signal-to-noise ratio of 10), Eq. (4) allows to calculate the drag force, $|F_\text{D}| = 0.14 \text{ nN}$ in air. It increases to $|F_\text{D}| = 0.65 \text{ nN}$ when the fiber is $d = 50 \mu m$ in water. The difference between these two values is due to the higher viscosity of water.

Fig. 2 shows the quality factor and the calculated drag force of the vibrating tip as function of depth $d$ of the tip in water. We observe that the quality factor falls from $Q = 240$ when the tip is in air to $Q = 7.1$, when it is completely immersed in water. This implies that for a given amplitude of vibration $|u_L| = 0.5 \text{ nm}$, the drag force increases from $|F_\text{D}| = 0.14 \text{ nN}$ in air to $|F_\text{D}| = 3.5 \text{ nN}$ when completely in water, that is 25 times higher.

4. New experimental set-up to minimize water drag force

Fig. 2 shows that if one uses the experimental set-up described by Moyer and Kämmer [3], where the tip is completely immersed in water, quite a big drag force is acting on the sample. To control the tip-to-sample distance, the arbitrary criterion implemented here is $|u_\text{int}| / |u_L| = 0.5$, where $|u_\text{int}|$ and $|u_L|$ are, respectively, the amplitudes of vibration of the tip close and far away from the sample surface. Using Eq. (5), one can then calculate the interaction shear force $|F_\text{int}|$ between the tip and the sample. For a fiber tip completely immersed in water, $|F_\text{int}| = 1.75 \text{ nN}$.

For hard samples, like a metallic grating, such a “large” drag force is not a limitation and good images can still be obtained. However, for softer samples this force is too high and can damage the sample, while producing artifacts in the images. To decrease the interaction force, Eq. (5) shows that it is necessary to either decrease the spring constant $k_\text{stat}$, or increase the detection of the vibration, or minimize $(|u_L| - |u_\text{int}|)$, or finally increase the quality factor $Q$. This last parameter falls very rapidly when the tip is immersed in water.

Fig. 3 presents a new experimental set-up designed to maintain a small ($< 50 \mu m$) and constant depth of the tip in water to minimize the degradation of the shear force distance control in water. It guarantees at the same time to maintain a constant depth during the approach of the tip to the sample and during the scanning. This is necessary since any change would modify the amplitude of vibration of the tip, and thus interfere with the tip-sample
distance control. The smaller the depth of the tip in water, the more sensitive the amplitude to any change of depth. Thus, a very accurate control of the depth of the tip in water is needed.

The solution proposed here is to add a glass tube around the fiber tip. This tube is held tightly to the ferrule to which the tip is glued. The tip is positioned longitudinally so that a small length 50 µm is kept outside of the aperture plane of the glass tube. Once this system is immersed into water, the surface of the liquid remains in the plane of the aperture, the tube being filled with air and tightly closed at the top. This system consequently maintains a 50 µm constant depth of the tip in water, for any tip to sample distance. A system of communicating vessels allows to compensate for the evaporation in the liquid cell. An interferometer, whose beam is focused about 1 mm above the extremity of the tip measures the dithering of the tip. This interferometer is based on a laser diode, whose output is fed-back into its own cavity [7,4]. This system has the advantage to be compact and accurate.

5. Image of a soft hydrogel

This new system is used to observe a hydrogel (Poly(Ethylene Glycol 400) diacrylate), containing 70% of water. Fig. 4 shows the SNOM topographical (shear force) and near-field optical reflectivity images of this sample at λ = 670 nm. The optical image is obtained by shining the illumination light through the tip and measuring the reflected light through the same tip. These images are reproducible for forward and backward scanning, showing that the tip is able to sense the surface without damaging it. Fig. 4a shows that the system allows to map the topography of the surface with a vertical

![Image of a soft hydrogel](https://via.placeholder.com/150)

Fig. 4. SNOM topographical (a) and near-field optical reflectivity (b) images (λ = 670 nm) of a hydrogel (Poly(Ethylene Glycol 400) diacrylate), containing 70% of water. The gray scale represents the relative intensity of the light coupled back into the fiber in arbitrary units.
precision of 13 nm. This precision corresponds to the root mean square (RMS) of the difference between the two images obtained by the forward and backward scans. Although the surface of the hydrogel is expected to look like the surface of a sponge, we observe a quite regular surface with almost no structure. This means that the feature size of the structure is smaller than the mechanical size of our tip (about 200 nm). However, the optical reflectivity shows brighter and darker regions of sizes between 100 nm and 1 µm which could be interpreted as regions of different densities of the hydrogel. Note that such an image is impossible to obtain with a tip completely immersed in water because the tip to sample interaction force is too large and results in damage of the sample surface.

6. Conclusions

A scanning near-field optical microscope (SNOM) system specifically designed to work in water has been proposed and optimized. This system uses the shear force technique for the tip–sample distance control. A measurement of the attenuation of the vibration amplitude of a fiber tip when immersed in water shows that the tip quality factor decreases from $Q = 240$ in air to $Q = 52$ when the last 50 µm of the tip are immersed in water. A further decrease to $Q = 7.1$ is obtained for complete immersion of the tip. In this latter case, it is impossible to obtain a SNOM image of a soft hydrogel sample (70% of water), because of the strong tip–sample interaction force responsible for dragging the tip onto the sample and damaging its surface. A new liquid cell is proposed, which allows to maintain a small and constant depth of the tip in water. This system is based on a glass protection tube which is tightly attached to the upper part of the fiber tip. With this liquid cell, a quality factor of $Q = 52$ could be maintained for the dithering of the tip. Such a fine interaction allows to sense the surface of the hydrogel in water without inducing any damage. This system is easy to implement and could be further optimized to increase the sensitivity of the tip-sample distance control in a liquid.

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References