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Vacuum Shear Force Microscopy Application to High Resolution Work

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A new technique—Vacuum Shear Force Microscopy (VSFM)—is introduced as a reliable method for maintaining a constant separation between a probe and sample. Elimination of many of the instabilities observed when applying the shear force mechanism to imaging under ambient conditions, allows for routine nanometer lateral and sub-nanometer normal resolution. In this paper this technique is applied, firstly, to the imaging of microtubules (biology) and, secondly, to the patterning and subsequent imaging of nanoscale metal lines (nanofabrication).

KEYWORDS: shear-force microscopy, nanofabrication, near-field optics, vacuum, scanning microscopy

1. Introduction

Significant economic benefits are to be obtained from the miniaturization. As a result there is a need for reliable nanoscale fabrication and imaging techniques. As past techniques, designed for the micro-scale, are often not applicable on the nano-scale, considerable research has gone into the development of new methodologies. In the area of surface imaging, the last two decades have seen first the discovery and then commercialization of a whole family of scanning probe microscopes. These include the atomic force microscope (AFM), scanning tunnelling microscope (STM), scanning near field optical microscope (SNOM or NOM) and shear force microscope (SFM). All members of this family provide sub-micron and, in certain cases, sub-nanometer imaging resolution.

Among the above techniques, that of SFM has proven to be the most controversial. Its well established use in NOM techniques has come under attack as a source of false optical contrast,[1,2] calling into question the results of many early experiments. In addition, the ability of the shear force (SF) images themselves to accurately represent surface topography has been questioned. It seems that the few nanometer thick surface layer may produce strange and non-reproducible results, including inverted topography,[3] resulting in the overall conclusion being that *constant SF signal does not necessarily mean constant height*. Considerable work has been done to illuminate the various interactions that collectively comprise "shear force". Key among these are Gregor *et al*.[4] investigation of the probe-surface interaction at a single point under vacuum conditions, and Davy *et al*.[5]'s investigation of the rise, under ambient conditions, of a thin water and pollutant surface film. In concluding their paper, Davy *et al*.,[5] make the radical comment that SF images must be taken within 10′ of exposure to air in order to be correlated with the surface topography. In this letter, we will show that the placing of a high quality SFM in a vacuum not only overcomes many of the problems associated with its application under ambient conditions but also provides higher resolution, image clarity and stability in the control of probe-sample separation. [It should be noted in passing that Behme *et al*.[6] have made considerable efforts improving SF instrumentation for variable cryogenic temperatures with a view towards spectroscopic studies. In their work resolution in vacuum is a secondary concern.[7]]

In addition to imaging, VSFM has found application in the area of nanofabrication. One example of the need for reliable distance control is the new and promising technique of near field optical photochemical vapour deposition (NFO-PCVD).[8] In this technique, an optical near field brush is used to write a pattern on a flat surface. Just as in Shodo (the art of Japanese brush calligraphy), reliable distance control is crucial to maintaining a constant line width, as, for a given brush, the separation between the brush and the writing surface determines each stroke's width. In this letter we will also show that by using VSFM one can accurately control the separation between the brush and writing tablet, allowing for uniform lines of uniform thickness to be written.

Currently, lines as narrow as 15 nm and as thin as 1–4 nm can be reliably written and then imaged. This is limited by the properties of the optical field rather than that of the shear force feedback system. We believe that the VSFM will become the method of choice for work in this exciting new field of nano-calligraphy.

2. Experimental Conditions

A schematic of a typical combination vacuum shear force, near field optical microscope is shown in Fig. 1. For explanation purposes, we can consider the VSFM as being composed of four separate systems: vacuum chamber, microscope head, shear-force distance control, and scanning electronics. In our case, the vacuum system is an encapsulated type reaction chamber designed with photochemical vapour deposition in mind. As well as operating in the presence of a few hundred Pa of Argon buffered metal alkyl gases (used for NFO-PCVD), a turbo molecular pump allows evacuation below 1 mPa vacuum. In addition to standard valves and ports, two aspects of the design deserve special mention. A special port passes an optical fibre, the tip of which is used both as a SF probe and near field optical source, through a sealed gasket, while a special window allows the approach of the sample to the SF tip to be observed using an external CCD camera.

The second key component is the microscope head. It must provide relative movement between the sample and probe tip in three dimensions while at the same time minimizing the effect of external vibrations and temperature fluctuations on probe-sample separation. In our initial prototype, the head was designed in a concentric configuration (Fig. 1), utilizing two coaxial 4 quadrant piezoceramic tubes (PZT). The inner

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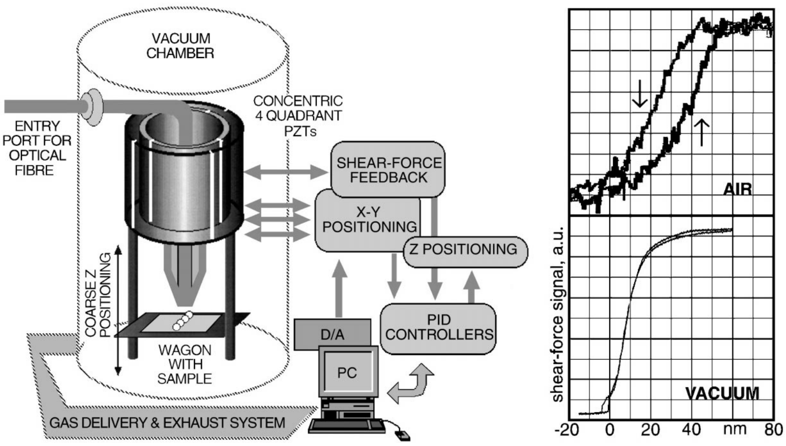

Fig. 1. Left: chematic of the vacuum shear force microscope. Right: typical shear force approach (arrow down) and retreat (arrow up) curves. These curves are taken under ambient and vacuum conditions (after 50′ at 2.7 Pa).

tube holds the sharpened optical fibre, while the outer supports the sample. Coarse lateral positioning is sacrificed in order to allow for higher mechanical stability. Coarse vertical positioning is provided by a piezoelectric-induced slip-stick motion scheme[9] utilizing the outer PZT. The typical amplitude of the tip lateral dithering did not exceed 1 nm, while the gap distance was 2–3 nm for imaging experiments and ∼5 nm for deposits patterning.

The third aspect of a VSFM concerns the excitation and detection of the SF signal. In our setup we have used both optical and non-optical methods to monitor the separation dependent SF signal. In the first case, the inner tube served to vibrate the fibre—its four quadrants to allowing bistability to be suppressed[10)]—and the SF amplitude was monitored using reflected laser diode light. A photo diode was used for detection. In the second, non-optical case, a tuning fork, mounted on the end of the inner tube, provided the detection signal. In both cases, the changes in the amplitude of the SF signal, after passing through a lock-in amplifier, were used in either an analogue or digital feedback loop to maintain a constant probe-sample separation.

A separate system provided the voltage to the outer PZT for raster scanning (imaging mode) or movement along a defined trajectory (NFO-PCVD write mode).

In actual operation, the sample is placed on the wagon far from the probe tip, the vacuum chamber closed, and air evacuated. After withdrawing the inner PZT, a coarse approach is made by alternately applying cycloid pulses to the outer PZT (slip-stick motion) to move the sample slightly less than 10 μm (the scan length of our PZT) up towards the fiber tip, and scanning the outer PZT its entire length. This procedure is repeated until a drop in the SF signal is detected (usually after a cup of coffee). At this point the voltage on the inner PZT is adjusted to position the outer PZT close to the center of its operating range thus minimizing PZT drift. An image is obtained, or a line written by moving the sample laterally, while using the feedback loop to keep the probe-sample separation constant.

3. Results and Discussion

The work described here involved three different components: observation of changes in the SF detection system in moving to low pressures, a comparison of ambient SFM and VSFM imaging of a well-known biological sample and, finally, a demonstration of the use of VSFM in conjunction with NFO-PCVD for nanofabrication. In the latter case, VSFM is used both to maintain a constant separation during patterned prenucleation as well as to provide a high-resolution topographical image of the sample after the completion of deposition.

The shear force signal as a function of probe-sample separation as the probe is first approached to the sample surface and then retreated away at both ambient and vacuum conditions is shown in Fig. 1 (right). In both cases, the shear force mechanism is only active at separations of a few tens nanometers or less. Of crucial importance to the matter at hand are the significant differences between operation under ambient and vacuum conditions. 1. As a result of the decrease of the damping coefficient at lower pressures, is a significant increase in the Q factor of the probe resonance. In the case of an optical fibre glued on one prong of a 33 kHz piezoelectric tuning fork, the Q factor at 2 mPa was 30% higher than that under ambient conditions ($Q = 1900$). 2. There is a slight blue shift of the resonance frequency.[6)] This shift ranges from a few Hz for high-Q probes to a few tens of Hz for low-Q probe assemblies. 3. There is significantly less noise seen in the approach and retreat curves taken under vacuum conditions which allows for much more stable operation under feedback. One possible explanation for this is the absence of the surface layer of water and contaminates under vacuum. Further elucidation of the shear force mechanism will be published elsewhere.[11)]

Our initial VSFM studies involved the observation of microtubules—a well studied biological polymer that forms the basis of the mammalian cytoskeleton and information expressway. These 25 nm diameter (TEM measurement) hollow polymer tubes are many micrometers in length. In noncontact AFM measurements[12)] by Vaters *et al*. (1995), microtubules appeared to have a diameter of 60 nm. Authors pointed out a structural broadening due to the well known effect of AFM tip-sample convolution (tip profile acts as a higher spatial cut filter), reducing resolution. In our previous work under dry ambient conditions microtubules were measured to be about 40 nm in diameter and 15 nm in height[10)] which we contributed to the slightly oval shape taken by these tubes when affixed to a glass cover slip. NOM measurements confirmed that the MTs have a diameter of less than 40 nm.[13) Figure 2 is a typical VSFM image of microtubules, demonstrating the enhanced image quality of VSFM over both published AFM[12)] and SFM measurements[10)] done under ambient conditions. In the upper left a large microtubule bundle shines brightly while in the lower right a single 25 nm diameter microtubule crosses the picture from left to right. In comparing images (cf ref. 10) taken under ambient and vacuum conditions, it is readily apparent that individual microtubules are much more clearly visible along their entire length and at a higher signal-to-noise ratio. This is despite the fact that vacuum conditions have caused the tubes to collapse from 15 nm to about 3–4 nm in height. Without going into any further discussion of the distortion of microtubules under vacuum conditions, we can very conservatively estimate our horizontal resolution to be well in excess of 5 nm, making VSFM a highly competitive alternative to non-contact AFM and even TEM in certain cases. But more important than just this resolution is the improvement in image quality at low pressure. This indicates that the vacuum shear force mechanism allows the sample-probe separation to be kept in a stable manner at a given distance. We attribute this not only due to the removal of the surface water layer and other impurities and the higher *Q*-value of the system but also the elimination of most air induced noise.

Encouraged by the quality of these images we decided to apply vacuum shear force as the distance control mechanism in NFO-PCVD. NFO-PCVD is a new technique that enables maskless production of nanometric structures with controllable size, chemical composition and morphology.[8)] In this two step technique an illumination mode NOM is placed inside the reaction chamber for PCVD. In the first step, prenucleation, a pattern is written on the substrate using the NOM in vacuum. In the second step, pattern formation, the optical tip is moved away from sample surface and the pattern grown using standard PCVD techniques. As the optical field density falls away exponentially as one moves away from the fibre tip, precise sub-nanometer control of the fibre tip—sample separation is required to ensure a constant line width. As images taken with NOM/SFM usually involve only a low number of pixels, image clarity, as shown by power spectral analysis,[14)] provides good evidence of high resolution. To be more precise, pixelation is a technical problem of the digital image acquisition on this specific system, rather than a fundamental problem of the analogue technique. In contrast to a conventional NOM, in the case of NFO-PCVD, the role of the optical detector is performed by highly sensitive metal alkyl molecules, selectively rupturing their chemical bonds under the induced optical field. An example of NFO-PCVD shown in Fig. 3. Although the probe used to draw this line had a 100 nm aperture, it is really the sharp exponential decay of the optical near field which most greatly impacts the width variations during patterning. Writing a line of constant width thus requires that the probe-sample separation to be held constant at a sub-nanometer scale. As can be seen by the even width of the Zinc line drawn in Fig. 3, VSFM is clearly up to the task of providing the high level of distance control necessary for NFO-PCVD patterning. In addition, the 15–20 nm wide, 2–4 nm high lines clearly attest to the superior imaging abilities and high resolution of the VSFM technique.

It is interesting to note that of the currently used techniques to maintain a constant gap separation, only SFM and possibly NOM can meet the requirements of NFO-PCVD. STM methods are inappropriate since the substrate is an insulator. While AFM can provide surface topography images of high resolution, its reliance on cantilever-based vertical dithering make it unsuitable for NFO-PCVD. Moreover, with digital signal processor controlled fast phase-tracking data acquisition,[15)] when thousands of pixels can be gathered within seconds, VSFM can be a very attractive method for a wide range of applications.

4. Summary

In conclusion, we have presented a new technique of probe-surface distance control and imaging Vacuum Shear Force Microscopy (VSFM) that overcomes many of the limitations and instabilities associated with the shear force mechanism operating under ambient conditions. It thus obtains unprece-

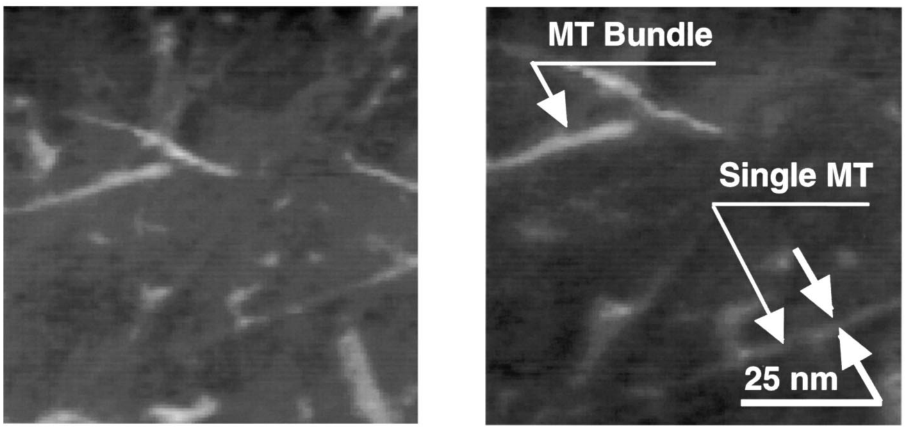

Fig. 2. Two consequent VSFM images of a microtubule taken at 2 mPa. Areas of the images on the left and on the right are $1028 \times 1028\,\text{nm}^2$ and $720 \times 720\,\text{nm}^2$ respectively. The microtubule was assembled from fluorescent rhodamine labelled tubulin using conventional techniques. A carefully cleaned hydrophilic quartz cover slip served as the substrate. A drop of poly-L-rezine (1 mg/m*l*) was placed on the substrate for 15 minutes and then washed off. The solution containing microtubules was then placed on the quartz cover slip and washed off after 30 seconds. A bunble of microtubules is indicated in the upper left portion of the image while a single microtubule crosses the lower part of the image. Single microtubules affixed using this method have an apparent full width half maximum FWHM of 25 nm in agreement with TEM measurements.

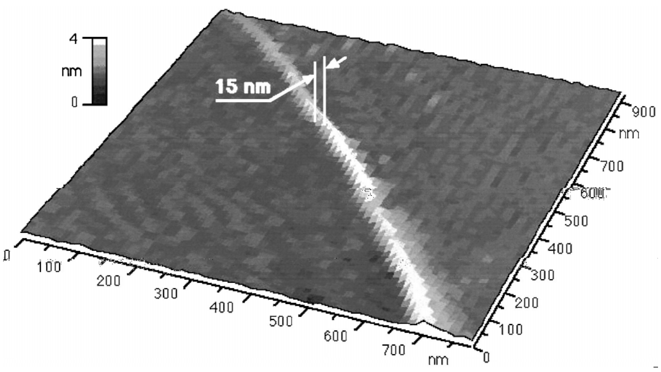

Fig. 3. VSFM image of Zinc lines written on Corning glass substrate by means of NFO-PCVD. The vacuum shear force mechanism is used not only to image the Zinc deposit but also to maintain a constant separation between the substrate and the NFO probe during patterning. The probe used in this case had a 100 nm light emitting aperture.

dented resolution and image clarity. The high vertical and horizontal resolution inherent in this technique enables its use in a wide array of applications, ranging from topographic imaging, nanofabrication, and surface energy transfer studies. This should make it a ready competitor to standard AFM and even to TEM in some cases. In addition, we have shown its unique applicability as a distance control mechanism in NFO-PCVD and its subsequent application in imaging lines deposited using this technique.

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