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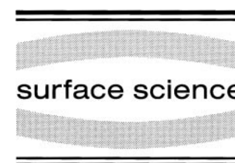

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The role of shear forces in scanning force microscopy: a comparison between the jumping mode and tapping mode

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Abstract

Shear forces are known to be one of the main causes of sample damage in scanning force microscopy. In this work, we compare two related imaging methods: jumping mode and tapping mode. These methods have been used in the imaging of two delicate samples such as DNA on mica and single-wall carbon nanotubes on silicon oxide. The results of these experiments show that while the tapping mode does not produce any visible modification, the jumping mode introduces irreversible sample damage. In the jumping mode case, the damage is explained assuming the presence of lateral force components derived from the normal force. In the tapping mode and under our usual experimental conditions, normal forces are found to be extremely weak, and thus, lateral contributions are negligible. From the experiments, we conclude that the absence of friction forces introduced by the tip scanning is not sufficient to obtain non-intrusive images. © 2000 Elsevier Science B.V. All rights reserved.

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Scanning probe microscopy techniques [1] have been shown to be a powerful tool in surface research (biology, nanotecnology, etc.) (see Ref. [2] for a review). The most widely used among these techniques is scanning force microscopy (SFM) because of its ability to obtain images of both insulators and conducting samples in different ambient conditions: vacuum, liquids [3,4] and ambient air [5,6]. One of the most common problems with this technique is the damage produced in soft samples during the scanning process [7]. Shear forces are known to be the main cause of damage when the SFM is operated in contact mode [8,9]. The development of non-destructive operating modes has been a breakthrough in the investigation of this technique [10]. Among them, the tapping mode (TM) is the most utilized because of its combination of a gentle scan and high spatial resolution.

In TM, the tip oscillates at a rather large vibration amplitude, that is, between 10 and 100 nm, near its resonance frequency. As the tip approaches the sample, the oscillation amplitude is reduced due to tip–sample interaction. This reduction is used as the feedback signal for the acquisition of topographic images. Initially, this amplitude reduction was attributed to repulsive forces during an extremely short contact time [11]. Since the contact time was so short, shear forces were negligible, thus avoiding damage to both the tip and sample [12–14]. More recent work shows

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TM regimes in which the amplitude reduction is due to attractive forces [15,16] or non-linear interactions [17]. In these regimes, amplitude reduction occurs without contact. A recent study [18] shows that in spite of the possibility of non-contact regimes, under the standard working conditions, the contact time represents about 10–30% of the oscillation period.

Another mode where shear forces are reduced compared to contact mode is the so-called jumping mode (JM) [19] also called pulsed force mode by other authors [20]. This technique basically produces a force versus Z displacement curve at each point of the image. With this method, the contact time and the applied force can be measured with a high accuracy. The important feature of this mode is that the sample is moved laterally with respect to the tip when the two are out of contact, thus minimizing shear forces during lateral motion [19]. As the shear forces are thought to be the main cause of damage while scanning, JM should also be a non-intrusive technique, similar to TM.

However, whenever the tip contacts the surface, a lateral force appears due to the average tip–sample angle. For typical tip–sample angles of $\sim 10^\circ$, this force is about 10% of the applied normal force. In any case, this lateral force must be present in any mode that implies tip–sample contact, as in JM or in the repulsive TM regime.

In order to compare both techniques, experiments were performed with two different kinds of samples: DNA adsorbed on mica and single-wall carbon nanotubes on silicon oxide. On the one hand, both are of similar dimensions (~ 2 nm diameter) and present an extremely weak interaction with the substrate; on the other hand, their mechanical properties are very different: while DNA is soft, carbon nanotubes are considered to be quite stiff [21].

Experimental SFM conditions for both samples are as follows: Olympus type cantilevers with a nominal force constant of 1 N/m, resonance frequency of 75-80 kHz and a tip curvature of 15–20 nm have been used. We have chosen these soft cantilevers because they can be utilized in both TM and JM. However, high spring constant cantilevers (40 N/m, 300 kHz resonance frequency) are useful for TM but useless for JM because of the large load/deflection ratio. Images were recorded at typical scan rates of 0.5 s per scan line. The free amplitude oscillation (a_{free}) was set to 60 nm. The reduction factor is chosen to be small, that is, the set point (a_{set}) is only slightly smaller than the free oscillation amplitude ($a_{\mathrm{set}}/a_{\mathrm{free}} \sim 0.9$). In this setup, any tip–sample interaction is minimized. JM images have been obtained by applying a maximum normal force of 10 nN with a tip excursion along the Z direction of 100 nm. The tip jump was chosen to be large enough to overcome adhesion forces during the scanning process. The contact time was set to 10 ms. This time has also been checked by direct measurement on an oscilloscope screen. Experimental set-up conditions are summarized in Table 1.

DNA samples were prepared following Hansma et al. [22]. In Fig. 1a a TM image of a DNA strand is shown. Several consecutive images were obtained with no change in topography, as expected. In order to compare both methods TM images are acquired until thermal drift effects are negligible. Then a JM image is taken. After taking this image, we returned to TM to check the effects of the JM scan on the surface. In Fig. 1b and d several types of manipulations are reported: geometrical shape modifications of the DNA (Fig. 1b); a break in the DNA strand (Fig. 1d) and some displacements of material (Fig. 1b and d).

Single wall nanotubes were deposited on a flat silicon oxide surface following Burghard et al. [23]. Fig. 2a shows a TM image of the nanotubes. As is the case with the DNA sample described

Table 1
Comparison between TM and JM

	Tapping mode	Jumping mode
Force constant (N/m)	1	1
Resonance frequency (kHz)	75	75
Contact time (μs)	$4^a/0^b$	10^4
Minimum tip–sample distance (nm)	$0^a/2.5^b$	0
Sample damage	No	Yes

[a] Theoretically calculated in Ref. [18].
[b] Estimated from Fig. 4.

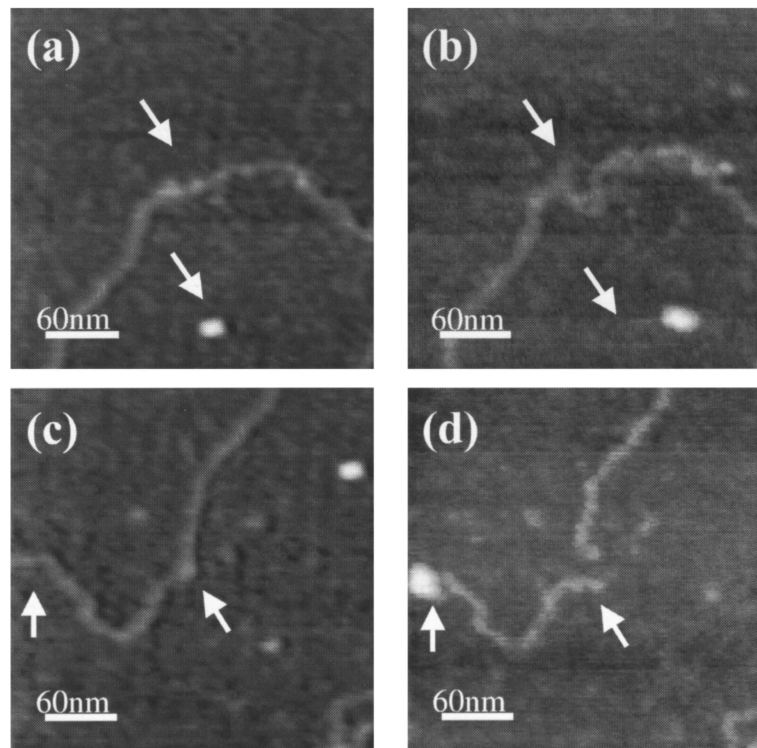

Fig. 1. Effects of JM on a DNA sample. (a) TM image of a DNA strand. Olympus type cantilevers with a nominal force constant of 1 N/m, resonance frequency of 75–80 kHz, and with a tip curvature radius of 15–20 nm have been used. Images were recorded at typical rates of 0.5 s per scan line. The free amplitude oscillation was set to 60 nm. The reduction factor is chosen to be small, that is, the set point is only slightly smaller than the free oscillation amplitude ($a_{set}/a_{free} \sim 0.9$). These parameters minimize the tip–sample interaction. For JM, the applied force was 10 nN with a tip excursion in the Z direction set high enough to overcome adhesion forces (about 100 nm). The contact time measured for JM is a few milliseconds. (b) TM image of the same area where the effects of the previous JM scan on this soft sample can be observed. Geometrical modifications of the DNA are seen in the middle of the scan area, and displacements of material are noted in the lower part of the image (see white arrows). It can be seen that the resolution of the image is not reduced after JM scanning. (c, d) Another area that has undergone the same process described above. Here, some new modifications can be found. A break in the DNA is clearly visible in (d) as well as some deposition of new material image (see white arrows).

above, consecutive images over the same area did not show any visible damage due to imaging in TM. In Fig. 2b and c, JM images of the same area are shown with similar parameters to those given above. After imaging in JM, TM is used to reveal any changes due to imaging in JM. Fig. 2d shows these changes.

Our experiments show that the two modes used, both of them developed to minimize shear forces, yield images with different quality. While TM images are obtained routinely without any appreciable sample damage, JM imaging of soft samples shows irreversible damage. Therefore, we conclude that, although a necessary condition, the absence of shear forces induced by the scanning is not sufficient to obtain images non-intrusively.

The origin of JM's intrusive behavior can be explained by taking into account the drawings depicted in Fig. 3. The upper diagram represents the force between the tip and sample. It can be seen that these forces may have lateral components, depending on the surface orientation. In the lower part of the figure, a three-dimensional sequence of a jumping mode scan is modeled. It is important to remember that in JM, the tip is moved perpendicular to the average surface plane

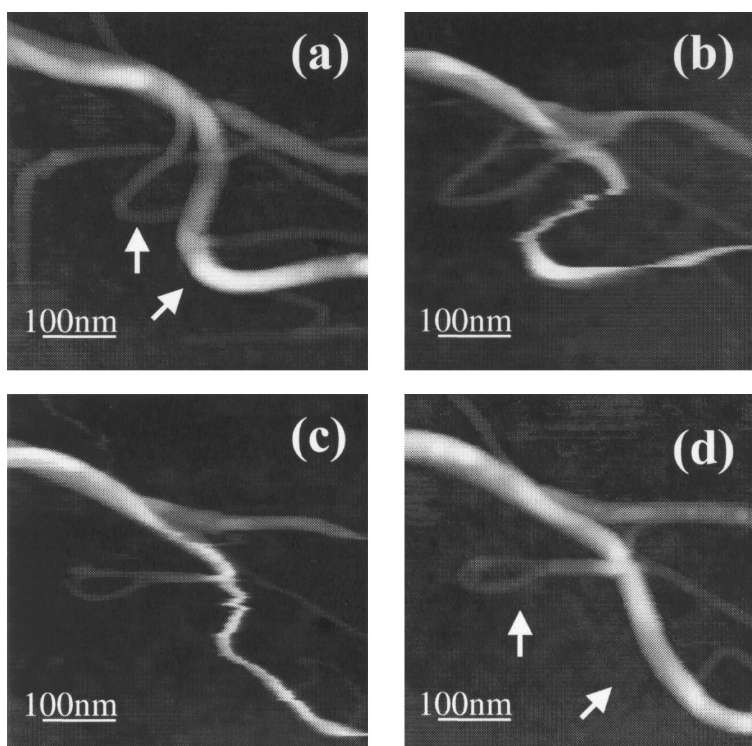

Fig. 2. Effects of JM on single-wall nanotubes. (a) TM image of the sample for use as reference. (b, c) Two consecutive images obtained in JM. Note the motion of the nanotubes during scanning. Comparison of (d), a TM image, with (a) shows the changes induced by the JM scan (see white arrows).

and parallel to it only when the two are out of contact. Depending on the relative position of the molecule line and tip axis, irreversible damage occurs. Fig. 3b shows the case where the molecule line and tip axis coincide. In this case, the force does not produce any lateral movement of the molecule. However, when the molecule line and tip axis do not coincide (Fig. 3c), the normal force with respect to the substrate creates a non-negligible lateral force on the molecule that will produce an irreversible displacement (Fig. 3d). A recent work by Falvo et al [24] shows that nanotubes can be moved laterally when a force, typically of 10 nN is applied. Since our maximum normal force is 10 nN, the maximum lateral force induced is also in this range. Thus, our experimental data support the hypothesis that these lateral force components, resulting from the normal force, are the principle cause of the irreversible damage observed in the JM images.

However, if tip–sample contact occurs in TM [11,26] lateral forces, as described in Fig. 3, should be present. We, however, do not observe any surface modification after scanning in TM. In order to investigate this contradiction, we have performed a new experiment combining both jumping and tapping modes. The idea is as follows. With the microscope operating in TM, under the conditions described above, a spot in a clean, flat region is selected on the sample; the X–Y scan is then stopped, and JM is enabled. This allows the dependence of the cantilever oscillating amplitude, as a function of the distance to the sample, to be analyzed. In this combined mode, the tip motion is controlled by the small piezo, upon which the cantilever is fixed, and the jumping motion is performed by the scanning piezo tube. The period of jumping cycles is in the order of milliseconds. As the resonance frequency of the cantilever is 80 kHz, the tip oscillates more that 1000 times

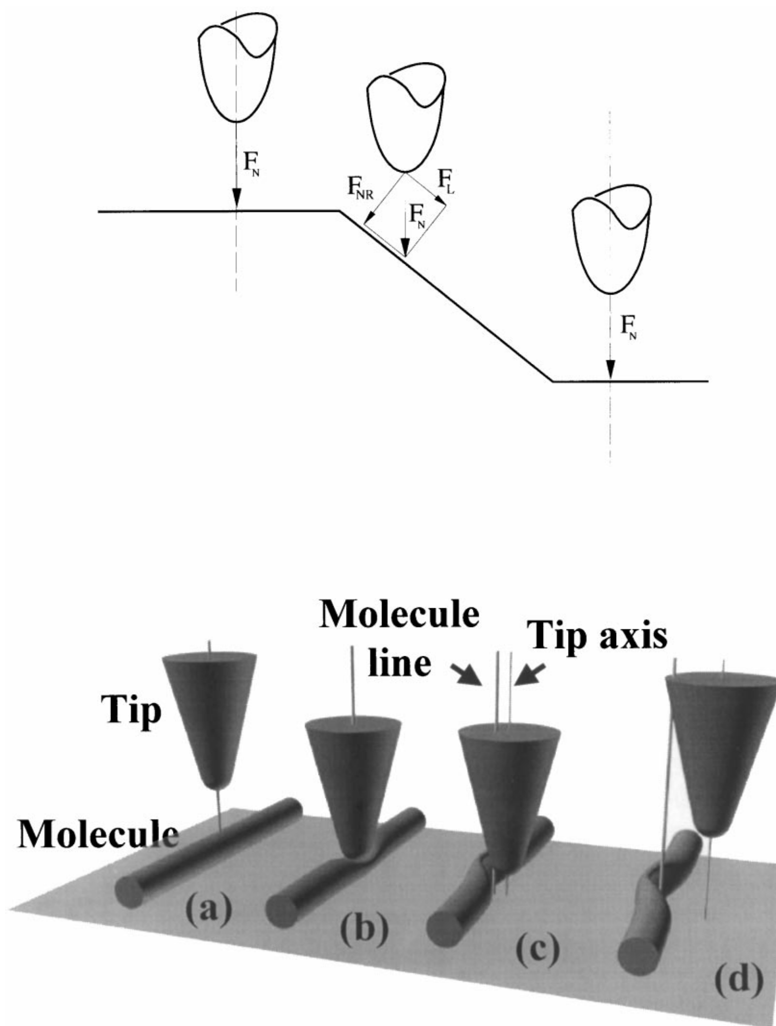

Fig. 3. Lateral forces induced by tip–molecule interaction. Schematic representation of JM interaction during an approach cycle. The upper diagram represents the force between the tip and the sample. Depending on the relative tip–surface orientation, lateral force components can be induced. The lower image shows this effect applied to a long molecule (a). When the tip axis and the molecule line coincide, no lateral displacement is produced (b). (c) Snapshot of the tip touching the molecule from the side. Here, the normal force creates a non-negligible lateral force component capable of producing an irreversible displacement in the molecule (d).

during a single jumping cycle. The data represented in Fig. 4 was collected using an oscilloscope in the X–Y mode. The sample motion is connected to the horizontal channel of the oscilloscope, and the normal cantilever deflection, detected by the photodiode, is fed into the vertical channel. The oscilloscope image is then recorded using a digital video camera connected to a computer.

To facilitate discussion, several labels have been added to Fig. 4 to mark points of interest. Point A is arbitrarily chosen as the starting point of the force versus distance cycle. Here, the tip is in the furthest position from the sample, and the cantilever is free to oscillate with maximum amplitude. At point B, an interaction appears between tip and sample, causing a change in the resonance frequency of the cantilever, which results in a decrease in the amplitude of oscillation. The ampli-

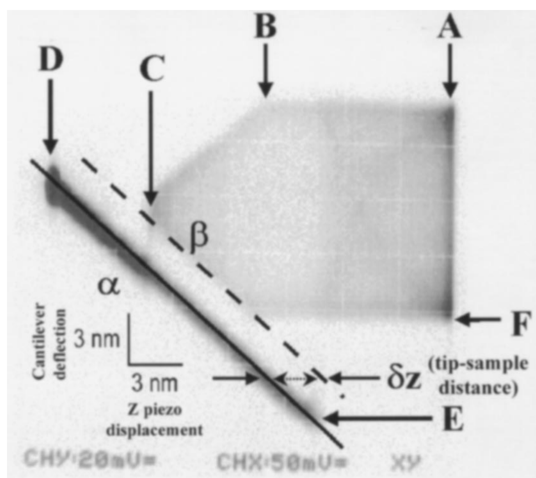

Fig. 4. Jumping and tapping combination, showing the oscilloscope trace of the cantilever deflection vs. Z-piezo position. The relevant points for one approach and withdraw cycle are shown. The tip is seen to go through several states. Between A and B, the cantilever is free, and the oscillation amplitude is maximum. From B to C, the amplitude decreases approximately linearly with respect to Z. At point C, the tip snaps to contact with the surface. As the Z-piezo continues to approach the surface, the cantilever deflects upwards until the set point of the normal force is reached (point D). After a few milliseconds, the piezo is withdrawn, and the tip jumps off the sample surface at point E. The cantilever can then oscillate freely again as it approaches the maximum tip–sample distance, F. Lines α and β are shown as guidelines. α (solid) defines the position of the surface, and β (dashed) marks the lower turning point of the oscillation. The horizontal arrow shows the distance, δz, between the surface and the tip at any given point. The data of this figure have been taken in 20 ms.

tude reduction is linear with the Z sample position and becomes zero at the point of contact (C). At point C, the contact force increases until the feedback set point is reached (point D). After a few milliseconds, the Z motion is reversed, and the tip is released (point E). As can be seen, this point does not coincide with point C because of the adhesion forces [25]. The cantilever can then oscillate freely again, as it approaches the maximum tip–sample distance, (point F). The lines α and β are drawn as guidelines. Line α (solid) goes through points where the tip is in contact with the surface and, consequently, defines the position of the sample surface. The key point in the argument is that this line defines the sample surface position [26]. Line β (dashed) is defined by the lower turning point of the cantilever oscillation. The fundamental feature observed in the figure is that, as the vibration amplitude of the cantilever decreases between points B and C, the tip does not reach the line of contact α between B and C. Thus, from this experiment, we conclude that, for the TM parameters used, the tip does not touch the surface. The results support the argument that, for TM operation under the given conditions, there is no tip–surface contact, thus avoiding lateral forces resulting from normal forces, as described in Fig. 3. We have also measured phase versus Z displacement in a similar manner to that shown in Fig. 4. We have found an inflexion point in the phase versus Z plot that corresponds with point B of this figure. A detailed study of the phase behavior in TM using the technique shown will be the topic of a future publication. The experiments were repeated on both samples for different oscillation amplitudes ranging from 10 to 100 nm, and the results remain essentially the same. It is important to note that, in spite of the absence of contact, the images present a resolution comparable to the tip diameter (~20 nm). TM experiments performed using SuperSharpSilicon®-Tips (Nanosensors® ref SSS-SEIH-8) show a resolution of 5 nm, again comparable with the tip diameter. Thus, in our case, non-contact TM cannot be attributed to the effect of large radius tips as in Ref. [16].

Following Ref. [18], the standard working conditions with high spring constant cantilevers would imply contact times of about 10–30% of the oscillation period. Hence, larger lateral forces would be induced by stiff cantilevers during this contact time. This would increase the observed sample damage. However, this type of cantilever is frequently used in TM to scan fragile samples. This apparent contradiction could be reconciled following two different hypotheses. First, the estimated contact time is so short (<1 μs) that the force between the tip and the sample is somehow negligible. The second hypothesis is that even for these cantilevers, there must be some working conditions where tip–sample contact does not take place, and when scanning extremely soft samples, these are the only possible working conditions to obtain reproducible images.

Summarizing, we have compared JM and TM as gentle imaging methods. Both techniques present, in principle, weak lateral forces. However, we observe that while TM does not produce sample damage when scanning, JM introduces surface modifications during the imaging process. This can be explained by assuming lateral forces induced by the normal force, which is always present, during contact time, in JM. To investigate the absence of damage in TM, we have combined TM and JM. The results indicate that under the standard experimental conditions given herein, the tip and sample do not come into contact during TM. Therefore, tip–sample forces in TM are much smaller than the force present in JM, allowing lateral force components to be neglected in TM. This is the underlying reason that extremely gentle TM scanning has been realized in these experiments. In conclusion, we believe that in order to image fragile samples with high-resolution TM, tip–sample contact must be avoided.

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References

[1] G. Binning, C.F. Quate, Ch. Gerber, Phys. Rev. Lett. 56 (1986) 930–933.
[2] C. Bustamante, D. Keller, G. Yang, Curr. Opin. Struc. Biol. 3 (1993) 363–372.
[3] Y.L. Lyubchenko, L.S. Shlyakhtenko, Proc. Natl. Sci. USA 94 (1997) 496–501.
[4] H.G. Hansma, D.E. Laney, M. Bezanilla, R.L. Sinsheimer, P.K. Hansma, Biophys J. 68 (1995) 1672–1677.
[5] C.F. Quate, Surf. Sci. 299–300 (1994) 980.
[6] J. Hu, X.D. Xiau, D.F. Ogletree, M. Salmerón, Science 268 (1995) 267.
[7] C. Bustamante, D. Keller, G. Yang, Curr. Opin. Struc. Biol. 3 (1993) 363–372.
[8] N.A. Burnham, O.P. Behrend, F. Oulevey, G. Gremaud, P.-J. Gallo, D. Gourdon, E. Dupas, A.J. Kulik, H.M. Pollock, G.A.D. Briggs, Nanotechnology 8 (1997) 67.
[9] J.P. Spatz, S. Sheiko, M. Möller, R.G. Winkler, P. Reineker, O. Marti, Langmuir 13 (1997) 4699.
[10] Y. Maeda, T. Matsumoto, T. Kawai, Appl. Surf. Sci. 140 (1999) 400–405.
[11] J. Tamayo, R. Garcia, Appl. Phys. Lett. 73 (1998) 2926.
[12] Q. Zhong, D. Inniss, V.B. Elings, Surf. Sci. 290 (1993) L688.
[13] M. Radmacher, R.W. Tillmann, M. Fritz, H.E. Gaub, Science 257 (1992) 1900.
[14] H.G. Hansma, J. Vesenka, C. Siegerist, G. Kelderman, H. Morrett, R.L. Sinsheimer, V. Elings, C. Bustamante, P.K. Hansma, Science 256 (1992) 1180.
[15] B. Anczykowsky et al., Phys. Rev. B 53 (1996) 15485.
[16] G. Bar, R. Brandsc, M.-H. Whangbo, Surf. Sci. 422 (1999) L192–L199.
[17] L. Nony, R. Boisgard, J.P. Aimé, J. Chem. Phys. 111 (1999) 1615.
[18] R. García, A. San Paulo, Phys. Rev B 60 (1999) 4961.
[19] P.J. de Pablo, J. Colchero, J. Gómez-Herrero, A.M. Baró, Appl. Phys. Lett. 73 (1998) 3300.
[20] A. Rosa-Zeise, E. Weilandt, S. Hild, O. Marti, Meas. Sci. Technol. 8 (1997) 1333–1338.
[21] M.M. Treacy, T.W. Ebbesen, J.M. Gibson, Nature 381 (1996) 678.
[22] H.G. Hansma, I. Revenko, K. Kim, D.E. Laney, Nucleic Acids Res. 24 (1996) 713–720.
[23] M. Burghard, G. Duesberg, G. Philipp, J. Muster, S. Roth, Adv. Mater. 10 (8) (1998) 584–588.
[24] M.R. Falvo et al., Nature 397 (1999) 236.
[25] P.J. De Pablo, J. Colchero, J. Gómez-Herrero, A.M. Baró, D.M. Shaefer, S. Howell, B. Walsh, R. Reifenberger, J. Adhesion 71 (1999) 339–356.
[26] C.A.J. Putman, K.O. Van der Werf, B.G. De Grooth, N.F. Van Hulst, J. Greve, Appl. Phys. Lett. 64 (1994) 2454.