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Intricate stepline artifact can mimic true atomic resolution in atomic force microscopy

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In atomic force microscopy, many artifacts are known to arise from sample elasticity, although its true atomic resolution capability has been definitely proven in liquid and UHV at optimally minimized loading forces during the past several years. In this paper the way in which a marginal but particularly shaped multiple tip can artificially produce an apparent sharp step line in the atomic scale image, due to elastic sample deformation at only slightly high loading forces, is demonstrated experimentally and illustrated. A real mono-atomic step with almost true atomic resolution was revealed simultaneously with the false apparent step line in one image, which has to be distinguished from the well-known ‘‘ghost image’’ artifact, although it is related. This effect, which could mimic true atomic resolution, is explained by attributing the x/y-lattice periodicity resolution and an additional locally ‘‘switched on’’ offset in the z signal to two different characteristic regions of a multiple tip. The measurement of the height of such a ‘‘false’’ crystal step is too small, whereas the ‘‘giant atomic corrugation’’ effect in contact mode atomic force microscopy is here suggested to be due to the same elastic sample deformation by a multiple tip.

I. INTRODUCTION

Sample elasticity is responsible for often observing the correct atomic scale periodicity by contact mode atomic force microscopy (AFM),[1–3] while being in fact far away from true atomic resolution.[2] A multiple tip being pressed into the sample is summing up many images of the same periodicity, thus retaining the correct periodicity while averaging over many unit cells and thus obviously showing an arbitrary structure within the unit cells. Interestingly, when increasing the loading force and thus through further sample deformation increasing the number of contributing (atomic scale) minitips, the signal to noise ratio for the periodicity picture can even increase, since (i) the lever's thermal vibration is obviously reduced as the lever becomes more strongly ‘‘clamped’’ at its free end (‘‘shifting’’ its thermal vibration more strongly to the first harmonic and higher), and because of (ii) the superimposition of the (possibly many) periodic pictures, which are contributed by (possibly many) minitips (e.g., tip atoms) at fixed relative phases (atomic spacings at the tip) when the whole ‘‘macrotip’’ is elastically indenting the sample surface. In the simplest approximation, mainly for high repulsive load, (ii) can be qualitatively illustrated as follows: Linearly summing up n sinusoidal line traces with identical amplitude A (e.g., along the fast scan direction x) with fixed relative phases (tip atom spacings) would lead to the resulting line trace:

$$A_n(x)=\sum_{j=1}^{n} A_{j-1}(x)=\sum_{j=1}^{n} A\sin[2\pi/\lambda_s x+2\pi(j-1)\lambda_t/\lambda_s],$$

where $A_n(x)$ will retain the correct periodicity but aquire an arbitrary amplitude; λ_s is the sample atom spacing, λ_t is the tip atom spacing, and n the number of contributing minitips (tip atoms). If atomic spacings on sample and tip surface are roughly equal, the amplitude of $A_n(x)$ can become much larger than A, at least for $n|\lambda_s-\lambda_t|<0.5\lambda_s$. This also holds for an amorphous tip (random relative phases) as long as the variation of λ_t is smaller than $|\lambda_s-\langle\lambda_t\rangle|$. Also for much larger n, a mean increase of the observed atomic corrugation amplitude can occur.[4] In addition, the sample can sometimes appear much stiffer when being indented by the macroscopic tip of a certain geometrical shape than upon expansion,[5] and the minitip line trace contributions can thus be expected to be ‘‘rectified.’’ Summing up those $(\Sigma_{j=1}^{n}|A_{j-1}(x)|)$ can lead to strongly altered unit cell shapes.[4]

This introduces only a very much simplified illustration. The superimposition of ‘‘minitip’’ images leading to the AFM micrograph from a nonideal probe tip is nonlinear [nonlinear elastic sample deformation at high loading forces $(>10^{-9}N)$ through the entire tip in contact with the sample; strongly nonlinear distance law of the electrostatic or dispersion force interaction between sample atoms and single tip atoms at low forces $(\leqslant 10^{10}N)$.]

Such artifacts due to sample elasticity such as the well-known ‘‘giant atomic corrugation’’ have long been explained by STM.[6] They are less understood in AFM, although they do exist there as well, as is commonly known particularly pronounced on elastic layered compounds like highly oriented pyrolytic graphite, mica, MoS_2. The effect (ii) of the superposition of many periodic pictures, as described above, could be one simple qualitative illustration for it. To my knowledge, giant atomic corrugation is not seen in true atomic resolution AFM images. For instance, in Fig. 5 of Ref. 2, the measured heights of the atomic (oxygen) sites on the calcite surface are roughly correct, as expected from the crystal data; the same approximately holds for the true atomic resolution images in Ref. 7. However, elasticity effects similar to the ones described in Refs. 5 and 6 should lead to at least slightly enhanced apparent heights, even on the true atomic scale. Generally, calibration of atomic force microscopy (AFM) data with respect to sample elasticity is often needed regarding the height information of the three dimensional topography map that scanning probe microscopes are capable of providing.[5] It is noted that lateral force effects also have been employed to explain the giant atomic corrugation in AFM.[8]

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Calcite is known as an extremely efficient standard sample for testing true atomic resolution in water. The AFM tip mainly detects the oxygen sites on the cleaved surface with lattice parameters $|\mathbf{a}|=8.1\,\text{Å}$, $|\mathbf{b}|=5.0\,\text{Å}$, $\gamma=90°$ (see, e.g., Ref. 2). Calcite is very slightly soluble in water (solubility product 4.7×10^{-9}), and so it apparently provides the precisely suitable ionic conditions near the calcite surface between tip and sample, such that the imaging forces can be well balanced. Then the effective load, which is exerted on the sample atoms by the front atom(s) of the tip, can be minimized below values of 10^{-11} to $10^{-10}\,\text{N}$.[2] On the other hand, the dissolution process is so slow that the AFM's imaging rate can still clearly reveal atomic steps, which were found to move only at roughly 2 nm per minute and even below, after the calcite-water system has been allowed to sufficiently equilibrate.

II. MATERIALS AND METHODS

A homebuilt atomic force microscope[9] was operated using commercial Si_3N_4 cantilevers with integrated tips (sharpened microlevers™, spring constant $k_{\text{lever}}=0.5\,\text{N/m}$, PSI, Sunnyvale, CA). Calcite crystals were freshly cleaved immediately prior (a few minutes) to imaging in tridistilled water. The minute solubility of $CaCO_3$ calcite (4.7×10^{-9}) may actually favor a cleavage surface clean enough for atomic scale surface studies in water.

III. RESULTS AND DISCUSSION

Figure 1 shows the intricate step line artifact, basically caused by sample elasticity. Since a significant artifact is intended to be clarified here, which, however, can only be recognized by relatively subtle deviations from a true step line, the AFM image naturally is not of the highest quality due to an incidental but characteristic imperfection of the tip, as will be analyzed. Two steplines are seen on the calcite surface imaged in water, where one of them has roughly the correct height ($3\,\text{Å}+/-0.2\,\text{Å}$, crystal data: 3.14 Å), whereas the other one is measured much too low with less than 1.5 Å [Fig. 1(b)]. Due to the marginal imaging conditions, the expected ‘‘phase shifts'' of the atomic rows between the upper and lower terrace when crossing the ‘‘3 Å high'' monoatomic step line, are only clearly resolved here for the rows parallel to lattice vector **a** (upper rows just pointing into the middle of the lower rows[2]). Crossing the shallow (<1.5 Å) apparent step, no such phase shifts are observed. Hereby, the fast scan direction was horizontal and its line frequency was roughly 25 Hz. At a usual thermal drift of a few Å per minute, no significant artificial phase shift should be expected between the ‘‘left'' and the ‘‘right'' step in the figure (less than 0.05 seconds ‘‘apart''), which are almost perpendicular to the fast scan direction. Only a slight overall bending of all the rows in parallel would represent such drift.

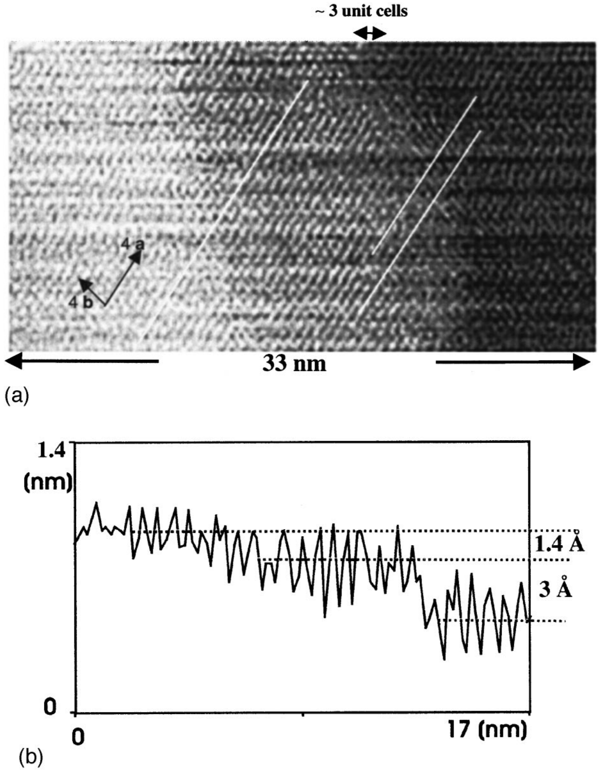

FIG. 1. (a) Atomic scale image of calcite recorded by AFM (constant force contact mode) in water. Two nearly parallel step lines are visible, where the higher step line (~3 Å, nominal crystal data: 3.14 Å) on the right side is revealed with almost true atomic resolution, just averaged/washed out over very few unit cells. Along lattice vector **a**, the atomic rows (5.0 Å spacing between them) on the upper terrace just point in the middle between the according rows on the lower terrace, as expected. The left step line, which appears even sharper, represents an artifact, which has to be clearly distinguished from a simple ghost image, since then it would have to be an exact copy of the other step. But the height of this artificial step line is much too small (<1.5 Å) and the atomic rows are in line across the apparent step edge. An explanation can only be derived by taking into account the elastic sample deformation in combination with a characteristically shaped multitip. The imperfect image quality is mainly due to the marginal but specific tip conditions, which are under investigation here. A nonorthogonality of the scanner caused the angle between **a** and **b** to slightly divert from 90°. (b) Line trace across the step in (a).

From this, it can be concluded that the higher step is the ‘‘real'' monostep imaged at the correct position in the micrograph, i.e., with nearly true atomic resolution, just averaging over very few unit cells, as indicated by the slightly washed out step line. However, these imperfect imaging conditions are necessary to demonstrate the above mentioned tricky artifact: The shallow ‘‘step'' is very much like a ghost image of the same step, imaged by a secondary minitip. Ghost images as such are a well-known phenomenon in scanning probe microscopy. However, a more complicated artifact is clarified here: It can be explained by a spatial separation of *x*-*y* lattice resolution and a secondary *z* signal (‘‘local'' *z* offset) between two different regions on a multiple tip (Fig. 2): The original primary (multiple) tip is delivering atomic periodicities, although very close to true atomic resolution. That the real step (3 Å high) is visible here, just averaged over a few unit cells. During a scan from right to left, when the secondary minitip ‘‘stumbles'' over the same step line, it contributes a small offset (less than 1.5 Å) to the vertical signal without contributing anything significant to the lateral lattice resolution. In the hypothetical model (Fig. 2) it would be, for instance, less than 17% of the ‘‘periodic

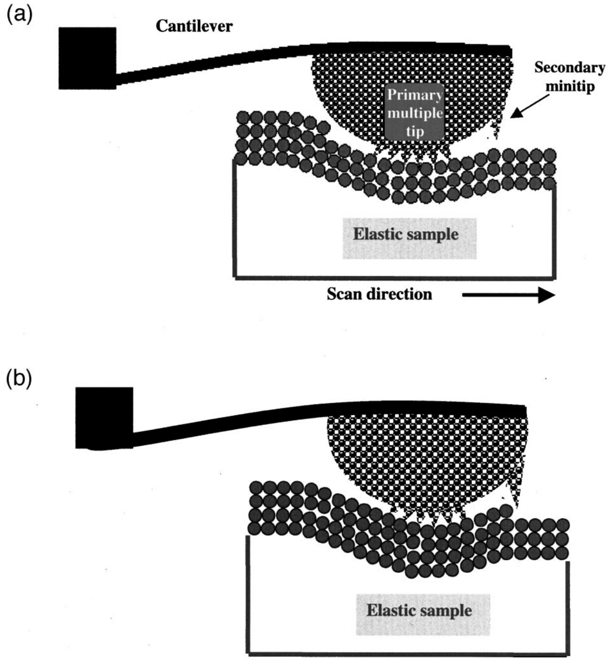

FIG. 2. Hypothetical model illustration of the tip-sample configuration responsible for the image formation in Fig. 1, arbitrarily chosen but certainly very close to the real situation. However, attractive noncontact forces are neglected here (contact mode), which is a strong simplification. A primary multiple tip (here six minitips) provides the image of the lattice peridodicity with close to true atomic resolution, just averaging six images by elastically deforming the sample (a). This primary tip provides an image of the step washed out by a few unit cells. The secondary minitip may not contribute at all until it follows the primary multiple tip crossing onto the higher crystal terrace. At this instance (b), it will get ''switched on,'' contributing mainly just a sharp but small z offset, appearing as a shallow but sharp step line, while the x/y lattice periodicity image is still formed to $>83\%$ at the primary multiple tip, i.e., by 6 of 7 minitips away from the actual step line on the defect-free terrace. It should be noted, that usually in contact mode, the overall force setpoint is attractive while the tip's front end is still being slightly pressed against the sample, since longer range attractive tip-sample interactions are ultimately determining the load (see, e.g., Ref. 2).

signal'' (six primary tips vs one displaced secondary tip). The apparent pronounced sharpness of the false step can be explained by the fact that the primary multiple tip continuously carries most of the load ($>83\%$ in the model in Fig. 2) applied to the sample during the whole image formation, while only a tiny additional load is exerted by the secondary tip when it passes the elastically deformed crystal step. Here, we have the merely incidental case of seeing both, the real step slightly washed out (which could as well lie outside the scan frame, depending on the distance between primary and secondary tip) and the sharp ''false ghost'' in one image. Of course, the same effect is possible, if atomic periodicities are imaged far away from true atomic resolution, i.e., averaging over some tens of unit cells by pressing the tip deeply into the elastic sample, as can be the case when imaging in air. Then, we would never see the real step, as the load of the primary tip would either elastically completely wash out the step line or would simply wipe it away. The much smaller load at the secondary tip could, even in air, allow it to ''stumble'' over this or another step line, which may induce a sharp ''jump'' in the vertical z signal, while the lateral periodicities are mainly ''generated'' perhaps some 10 nm away at the blunt primary (multiple) tip. This artifact is recognized by a measured apparent step height which is much too small, and there is no phase shift at all between atomic rows on apparent upper and lower crystal terraces when crossing the apparent step line in the image. The lesser minitips contribute to the primary multitip (probably only very few in Fig. 1 as illustrated in Fig. 2), the more the observed apparent unit cells may change in the direct vicinity (within the averaging regime of the multiple tip) of this false stepline, when the secondary tip gets ''switched on'' by passing onto the higher terrace, i.e., if this secondary tip's contribution to the x-y-periodicity image is no longer negligible. Such change in the unit cell's appearance could be eventually misinterpreted as a phase shift, but the atomic rows are practically perfectly in line further away from this ''ghost line'' (Fig. 1). Note that a regular ghost image without elastic sample deformation would have to be an exact copy of the primary image details, i.e., here, the phase shifts along the ghost step would have to be visible as well.

It should be noted that although the real step is measured with its correct height, at the same time the atomic corrugation appears already significantly enhanced [>2 Å instead of the expected (see, e.g., Ref. 2) value of about 1 Å]. Apparently, averaging over just a few unit cells (i.e., adding periodic traces from only very few minitips) can already lead towards the ''giant atomic corrugation'' artifact in AFM as hypothetically described above, which apparently is not observed in the true atomic resolution images in.[2,7]

As another illustration, a simpler manifestation of a comparable effect, i.e., a separation of the x/y lattice resolution on one hand and an additional local z offset on the other, could occur when imaging more soluble crystals in water, where step lines are moving much faster than the AFM's image frame rate. Atomic periodicities may still be imaged even with a blunt tip at high load, perhaps averaging over many unit cells. If a dissolving step line rushes through underneath the imaging tip, one will see a step line that may even appear relatively sharp, however, in this case, more or less parallel to the fast scan direction.

IV. CONCLUSIONS

In summary, this paper emphasizes that very intricate artifacts can mimic atomic resolution in AFM: besides the well-known effect of revealing atomic scale lattice periodicities while a multitip can actually be averaging over several unit cells, even sharp apparent step lines can be falsely visible on the atomic scale. The latter is proven by Fig. 1. Both effects are caused by elastic (and sometimes even plastic) sample deformations, the latter effect being additionally due to a characteristically shaped multitip as modeled in Fig. 2. Thus, a step line can unambiguously prove true atomic resolution, only if both step height *and* phase shifts of the atomic rows across the step are observed correctly. In contrast, observing giant atomic corrugation in AFM can be a good indication for merely seeing the lattice periodicity folded with a multiple tip, which is elastically deforming the surface, i.e., it can perhaps be a good indication for not having true atomic resolution. Reliable true atomic resolution in contact-mode AFM is most likely only possible if the relevant loading forces at the tip's front end have been minimized down to or below $10^{-10}N$.[2] Thus, the most reliable choice is obviously noncontact atomic scale imaging.[2,7] In particular, oscillatory noncontact techniques,[7,10] detecting a cantilever's frequency shifts in UHV, have been proven to enable true atomic resolution in feedback-controlled constant interaction mode. Further, due to their higher sensitivity, dynamic force microscopy techniques[7, 10–13] make possible the usage of stiffer (>10 N/m) levers, and furthermore they allow dynamic minimization of the relevant loading forces between the sample and the front end of the tip by adjusting the lever amplitude towards larger values. Therefore, using DFM techniques, true atomic resolution in noncontact mode with feedback control may be also possible in a liquid,[12] perhaps even on strongly corrugated samples such as biomolecules. The same concept may hold for imaging in air.[13]

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