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Probe chip nanofabrication enabled reverse tip sample scanning probe microscopy concept and measurements

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Abstract

We introduce a new scanning probe microscopy (SPM) concept called reverse tip sample scanning probe microscopy (RTS SPM), where the tip and sample positions are reversed as compared to traditional SPM. The main benefit of RTS SPM over the standard SPM configuration is that it allows for simple and fast tip changes. This overcomes two major limitations of SPM which are slow data acquisition and a strong dependency of the data on the tip condition. A probe chip with thousands of sharp integrated tips is the basis of our concept. We have developed a nanofabrication protocol for Si based probe chips and their functionalization with metal and diamond coatings, evaluated our probe chips for various RTS SPM applications (multi-tip imaging, SPM tomography, and correlative SPM), and showed the high potential of the RTS SPM concept.

Supplementary material for this article is available [online](#)

Keywords: RTS SPM, multi-tip SPM, Si tip, metal tip, diamond tip, probe chip, RTS scalpel SPM

1. Introduction

A sharp tip traces the surface to probe and record its local interactions with it; this simple concept led to the invention of scanning probe microscopy (SPM), a breakthrough characterization technique that enabled the exploration and manipulation of surfaces at the nanometre and atomic scale [1]. Indeed, SPM has had a strong impact on the research in various fields, most notably in materials science [2, 3], nanotechnology [4], and biology [5, 6]. Such versatility stems

from the technique's constant evolution. From its origins dating back to the 1980s, and first prototypes operating in quantum tunnelling [7] and atomic force regimes [8], many flavours of SPM methods have been developed to probe now a plethora of different tip-sample interactions, such as physical, chemical, electrical, near-field optical, and thermal [9]. Even though the SPM technology has made remarkable progress over the years, it still suffers from its fundamental limitation: the measurement will be only as good as the tip is. In other words, scanning with high-quality tips is crucial for obtaining accurate and reproducible SPM data. The issue here is that in most cases the integrity of the tip is not preserved during the measurements [10]. The otherwise sharp apex of the tip can change its shape, become blunt, contaminated, or break off completely. This leads to the appearance of the



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well-known visual artifacts in the SPM data [11, 12]. For their detection and deconvolution from real data, several research groups have proposed the use of machine learning algorithms [13–17].

Still, a better, and more direct solution is to simply replace the bad tip. However, this approach has several drawbacks in terms of efficiency and applicability. First, changing the tip is a manual and time-consuming process in most conventional SPM instruments. Second, it is difficult to relocate the same sample position after the tip change, resulting in the loss of the area of interest. Third, it impedes the further development of SPM technology [18]. Based on the recent trends in SPM correlative research [19, 20], a high-throughput automated SPM platform that can perform multimodal correlative SPM measurements would be a desirable innovation for both industrial and academic purposes. However, this would require a fast and automated system for changing the tips, which from the engineering standpoint is a challenging task. Alongside tool manufacturers, researchers are now exploring clever solutions to overcome this challenge and prevent the SPM field from reaching the point of technological stagnation [21–23]. Therefore, we introduce in this paper another conceptual iteration of SPM, with which we shift the paradigm from a tip scanning the sample to a sample scanning the tip. We coin this novel approach as reverse tip sample scanning probe microscopy (RTS SPM). At the core of this method lies a nanofabricated probe chip that hosts, from the SPM user's perspective, nearly an infinite number of sharp Si-based tips. It can be placed on a sample stage of a standard atomic force microscopy (AFM) instrument without hardware modifications, and without imposing limits in terms of operational modes and working conditions. In RTS SPM the sample is mounted at the end of a tipless cantilever. Upon engaging, the cantilever will start to scan one of the tips on a probe chip. Changing a worn-out tip is a quick and simple task in the RTS configuration, as it is only a matter of moving the cantilever onto a nearby tip.

To demonstrate the viability of the RTS SPM concept, we have developed a robust and time-efficient nanofabrication protocol for Si-based probe chips. Our probe chips are made by a plasma-based dry etching process, which is done in a single session and features a novel tip sharpening step. Furthermore, we applied additional metal and boron doped diamond (BDD) coatings onto the fabricated Si probe chips to test a broad application scope of RTS SPM. The quality of the resulting probe chips was evaluated by scanning electron microscopy (SEM), while their performance was tested by doing RTS SPM measurements on diamond nanoparticles, Si surfaces, and two-dimensional (2D) MoS₂ layers. The obtained results showcase the potential behind the RTS SPM concept.

2. RTS SPM method

In RTS SPM, both the experimental design and operation are inverted with respect to the standard SPM method. As figure 1(a) illustrates, the sample is now mounted on a

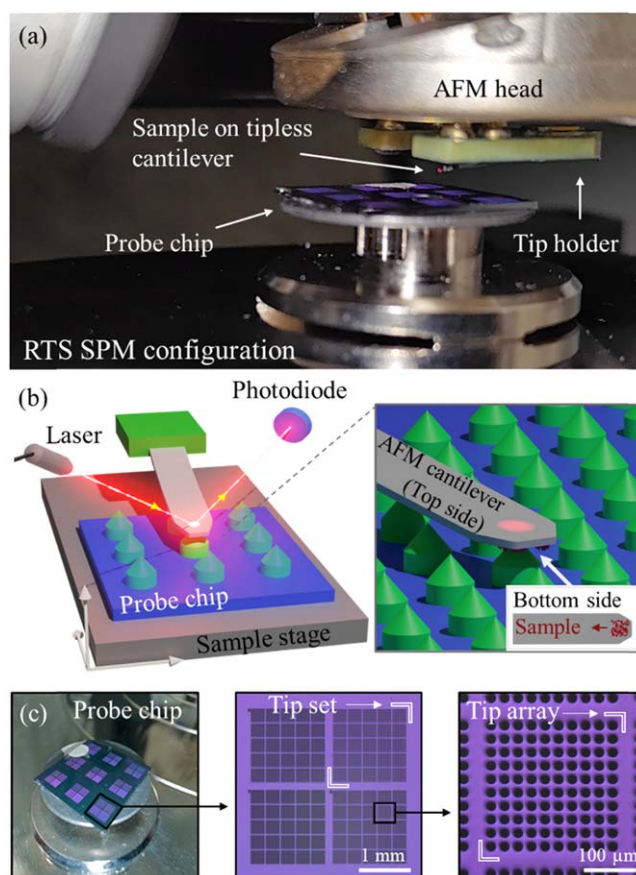


Figure 1. (a) Picture of an RTS setup. (b) Schematic diagram of the RTS SPM concept, showing the experimental layout of the RTS configuration together with the sample that was transferred on the tipless cantilever, now scanning one of the tips on the probe-chip. (c) Series of pictures of a probe chip at different magnifications.

commercially available tipless cantilever, while the tips are integrated into an in-house nanofabricated probe chip. The probe chip can be placed either on the sample holder or directly on the sample stage. To perform a measurement in the RTS configuration, the cantilever holding the sample engages with one of the thousands of identical sharp tips located on the probe chip (figure 1(b)).

The measurements proceed as usual after this point. Once the tip wears out, the user simply applies an adequate piezo XY offset to reposition the cantilever onto a fresh neighboring tip, a process that takes mere seconds to complete and allows for scanning to be resumed in the same area of the sample, without any disruptions to the experiment. This is in stark contrast to the standard SPM approach, where each tip replacement entails retracting the cantilever, taking off the old tip and putting on a new one, re-engaging the sample, and locating the same region of interest. The entire sequence can last anywhere from approximately 15 min in ambient conditions to several hours in a high vacuum environment. Therefore, in the application space involving frequent tip replacements, RTS SPM offers a significantly higher data-per-minute performance over the standard SPM method. It is also worth noting that the probe chip can be made compatible with any standard SPM system, and thus no structural

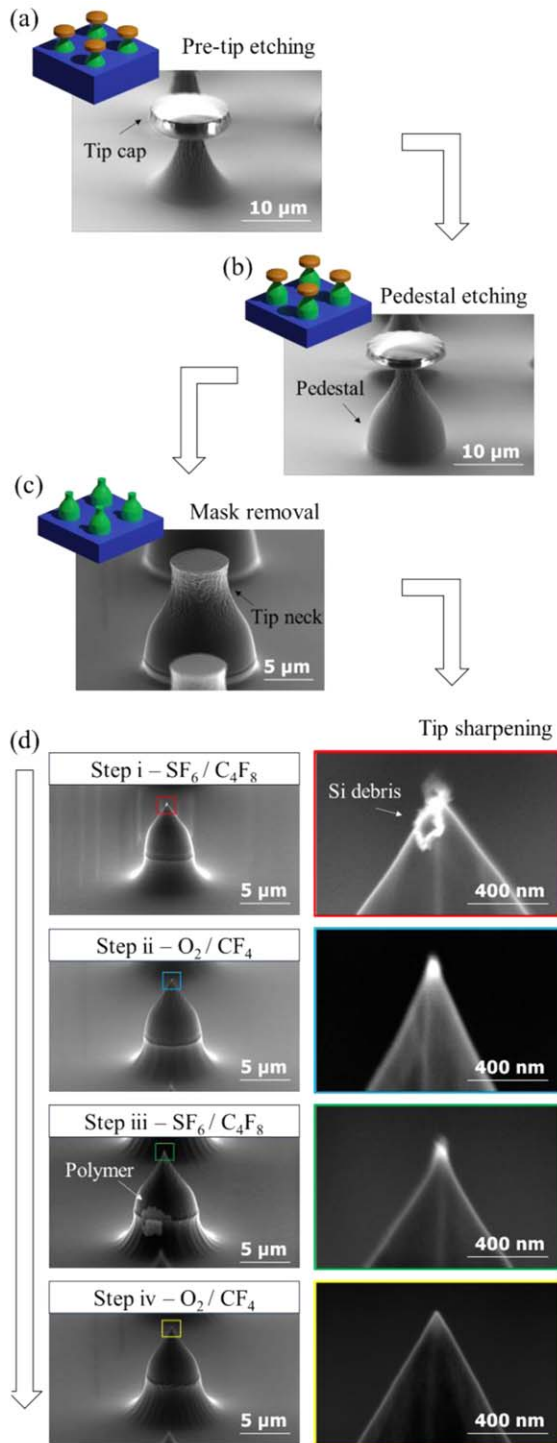


Figure 2. Probe chip nanofabrication protocol presented through a series of sequential SEM micrographs that cover every step of the process. (a) Isotropic etching of the pre-tip structure. Graphical inset is intended to highlight the photoresist tip-cap (orange) and the Si pre-tip structure (green) that was formed by isotropic etching of the p-type Si (100) substrate which now acts as the probe chip base (blue). (b) Increasing the height of the structure by creating the pedestal with anisotropic etching. (c) Removing the tip-cap. (d) A series of SEM overviews and red, blue, green, and yellow close-ups that show the simple four-step tip sharpening method, which includes: (i) neck separation etching step; (ii) debris removal step; (iii) tip sharpening etching step; (iv) clean-up step.

adjustments to the instrument are needed. Furthermore, the RTS configuration can be executed in any working environment (air, inert atmosphere, or vacuum) and at any operational temperatures (room, high, or cryogenic). Finally, and most importantly, all standard AFM modes are compatible with the RTS geometry, since it does not introduce new physics which could affect the interactions between the tip and the sample.

When designing an RTS SPM experiment, there are a couple of key considerations to bear in mind. First and foremost, there are constraints regarding the sample preparation such as sample size, cantilever stiffness and sample attachment method. The restricted sample area is contingent upon the dimensions of the tipless cantilevers available on the market (around $100 \times 100 \mu\text{m}^2$). Additionally, the sample should be as thin as possible to prevent it from acting as a lever arm, thus avoiding any potential unwanted torsion contributions that might affect the measurements. Next, a careful selection of the tipless cantilever stiffness is imperative for each experimental scenario, as it cannot be changed anymore after the sample has been attached to the cantilever. It is noteworthy, however, that probes featuring multiple cantilevers having the same (e.g. ARROW-TL8 by NanoWorld) or diverse spring constants (e.g. HQ:NSC36/tipless/No Al by MikroMasch) are commercially available. Therefore, it is possible to attach multiple identical, or even completely different samples onto each one of the cantilevers found on these kinds of tipless probes. Lastly, while there could be numerous ways to attach the samples (such as direct material deposition/growth, seeding, transfer, pick-up, etc), it is crucial to carefully select an appropriate method or even devise a new one tailored to a specific application.

The second set of considerations concerns the probe chip. Again, for a given application, it is important to choose the optimal layout, size, and shape of the tips on the RTS probe chip. In our case, the standard probe chips are comprised of 25 tip sets which formulate a 5×5 grid (see figure 1(c)). Each tip set hosts 100 tips arranged into a 10×10 tip array. The spacing between the tips within the tip arrays is $25 \mu\text{m}$. The reasoning behind this value is twofold. First, the tip spacing should be large enough to allow for the full width of the tipless cantilever to be scanned without unintentional side contacts with the neighboring tips. However, the spacing should still be kept as small as possible to incorporate as many tips as possible within the scan range of the AFM piezo driver. For our instrument (Bruker's Dimension Icon AFM) this is $90 \times 90 \mu\text{m}^2$, which means that we have, at least, 9 tips at our direct disposal without the need to withdraw the cantilever for repositioning using the course motors. Finally, the tips on the probe chip should be uniform high-aspect-ratio structures which end with an ultrasharp apex on the top. For our applications we determined that the height of the tips should not be less than $8 \mu\text{m}$, to allow for deep scanning of the cantilever without front contacting the base of the neighboring tip. All these requirements for the probe chips

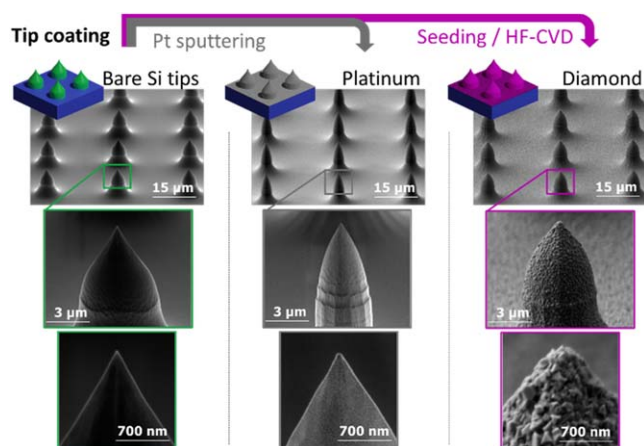


Figure 3. Results of the probe chip functionalization process. SEM images of bare Si probe chip, Pt, and BDD coated ones, from the left to the right, in their respective order and at different magnification levels.

can be satisfied by our nanofabrication process which is discussed in the next section.

3. Probe chip nanofabrication

Here, a detailed description of the probe chip nanofabrication process is provided. Over the years, wet anisotropic etching, and dry reactive ion etching (RIE) have become the two most prominent ways to fabricate Si tips. Even though, out of the two, wet etching is a significantly cheaper option, controlling the final shape of the tips is challenging due to the much higher etch rates. Furthermore, the aspect ratio of the end structures is constrained by the method's dependency on the Si crystal orientation [24]. For this reason, we opted for the dry etching method as it offers both isotropic and anisotropic etching capabilities, which are essential for creating densely packed high-aspect-ratio structures. We have developed a reproducible and highly efficient protocol, where all dry etching steps are performed in a single dry etching session and within a single working day. The first step of our process was the mask patterning. The mask is comprised of circular features which are intended to form 10 μm —wide 'tip caps' on a 4-inch diameter p-type $\langle 100 \rangle$ Si wafer (see figure 2(a)). This was accomplished by spin-coating a positive photoresist onto the substrate, followed by the subsequent UV lithography patterning. After the mask development, the wafer substrate was transferred into an Oxford Plasmalab System 100, where the inductively coupled plasma—reactive ion etching (ICP-RIE) took place. The dry etching process started with the isotropic etching of the pre-tip structure using a sulphur hexafluoride (SF_6) and perfluorocyclobutane (C_4F_8) gas mixture. The former is a common dry etching agent for Si [25], whereas the use of the latter, in a smaller fraction ($\text{SF}_6:\text{C}_4\text{F}_8 = 2:1$, in our case), is known to improve the formation of high-aspect-ratio structures by acting as a passivation-like CF_2 polymer film is formed on the sidewalls of the pre-tip

structure which is highly efficient at absorbing fluorine radicals. Since subsequent ion bombardment favours the polymer removal at the base of the structure over its sidewalls, one can have a better control of the taper angle [27]. As it can be seen from the SEM image presented in figure 2(a), the pre-tip structure was successfully etched below the tip-cap. However, the obtained tip structure was not yet compliant with one of the probe chip's design requirements—a minimum tip height of 8–12 μm is required to enable the deep cantilever scanning without frontal collisions with a neighboring tip. For this reason, we performed in the second step Pseudo-Bosch anisotropic etching by further increasing the amount of C_4F_8 in the gas mixture ($\text{SF}_6:\text{C}_4\text{F}_8 = 3:4$). This allowed us to fabricate a pedestal-like structure under the pre-tip to increase its height without significantly changing its morphology or the width of the base (please refer to the SEM image in figure 2(b)). Next, we removed the tip caps (figure 2(c)) as a lead up to the tip sharpening step. Even though a mask removal is readily done just with the use of an O_2 plasma, we used the recipe which incorporates a very small amount of CF_4 , because it is known to improve the etch rate [28]. From this point on, the standard protocol would be to use thermal oxidation as a mean to obtain sharper tips [29, 30]. However, a big drawback of this method is that it requires a specialized oxidation furnace, which makes the overall process more expensive and time-consuming. To overcome these challenges, we have devised a novel four-step tip sharpening protocol, which is incorporated into our single ICP-RIE session. As presented in figure 2(d), our protocol uses an alternating sequence of isotropic Si etching steps and clean-up cycles. (i) It is true that upon mask removal, performing just a single isotropic etching step would contribute to the sharpening of the tip apex, but note that the thinnest part of the pre-tip located between the apex and the pedestal structure, the 'tip neck', will be etched first. Consequently, a small amount of material above the tip neck is likely to collapse and form Si debris near the tip apex. This would have a negative impact on the overall tip quality and process reproducibility. (ii) Therefore, in our second step, we addressed the tip neck area with the use of a plasma generated from a mixture of O_2 and a modest quantity of CF_4 . SEM images indicated that there was no surface contamination on the tips, albeit the satisfactory results in terms of sharpness were still lacking. (iii) Thus, the additional tip sharpening was accomplished by performing yet another cycle of isotropic etching. (iv) Lastly, we removed any remaining polymer residues from our probe chips with the final O_2/CF_4 plasma clean-up session, and with that, we completed our probe chip nanofabrication process. The Si tips were found to be uniformly sharp across the produced probe chip as shown in figure S1, thus making the probe chip suitable for RTS SPM measurements. However, to further expand the application scope of the RTS SPM method, we functionalized our tips by coating them with platinum (Pt) and BDD. The high electrical conductivity of Pt enables the use of the RTS SPM method to measure the electrical properties of the samples. We used sputter deposition due to its isotropic coating characteristics, allowing us to uniformly deposit a 10 nm thick Cr adhesion layer and then a 30 nm thick Pt layer with a consistent

thickness onto the Si tips. Because metal-coated tips are more susceptible to wear, we also fabricated conducting BDD coated tips, which offer excellent mechanical and chemical stability, while still retaining good electrical characteristics.

This was achieved by depositing 200 nm of BDD with the use of hot filament CVD. For comparative analysis, in figure 3, we show the SEM images of the bare Si probe chip together with Pt and BDD-functionalized Si ones.

As expected, the deposition of either material caused the tip apexes to increase slightly and become decorated with a grainy texture. However, the best way to evaluate the quality of the obtained probe chips is to test their performance in RTS SPM measurements.

4. Evaluation of nanofabricated probe chips in RTS SPM measurements

4.1. RTS ensemble imaging with bare Si probe chip

To evaluate the uniformity of the bare Si tips on the nanofabricated probe chip, three parameters were tested: tip height, tip-to-tip separation, and tip quality. A few sets of nine Si tips were randomly selected across the probe chip. With the expected tip-to-tip separation of 25 μm , all the tips within the 3×3 grid were in reach of the AFM piezo driver; therefore, tip-to-tip switching could be done by simply applying piezo offsets in increments of 25 μm in X or Y directions. We conducted AFM measurements in tapping mode and in the RTS configuration on diamond nanoparticles which were transferred directly from a liquid ethanol/diamond nanoparticle suspension by so-called seeding onto a tipless Si cantilever (MikroMasch HQ:NSC35/tipless/No Al (5.4 N m^{-1})) without any additional processing steps [31]. We found these nanostructures to be suitable as testbeds for this type of study because of their small dimensions and intricate shape. Note that what we refer to as a ‘diamond nanoparticle’ is a cluster of aggregated 5 nm large nanodiamonds. To obtain comparative statistics, we used multiple tips to resolve the structural features of a single nanoparticle. The corresponding tip switches were done after each completed scan. Figure 4 illustrates the outcomes of the representative experiment. We used nine different tips in this experiment, enumerated as in the first AFM camera view. However, we will focus only on the six tips at the extreme ends of the piezo range, as they are crucial for testing the tip height and tip-to-tip separation. These parameters were important to ensure that the cantilever did not crash when moving from one tip to another, and that the offsets between the recorded images were minimal. Our results showed no signs of these negative effects, indicating that the tips had good uniformity in terms of height and separation. Among other experimental examples (see figure S2) we selected this one as it highlights statistical diversity. Figure 4 shows the AFM camera view of the cantilever positioned on each tip and the corresponding AFM images of the nanoparticle they produced. The AFM images reveal that tips 4, 7 and 9 generated the best results, as they captured the reproducible shape (in this case, a diamond

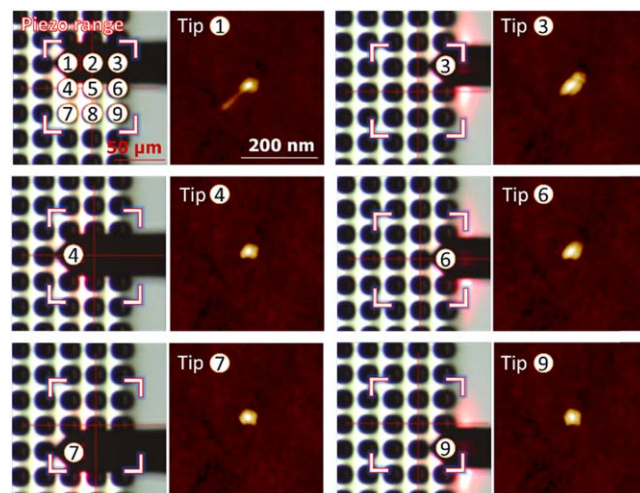


Figure 4. Dynamic mode RTS AFM measurement of diamond nanoparticles which were deposited on a tipless Si cantilever. A sequence of six scans conducted with six different bare Si tips (labelled 1, 3, 4, 6, 7, 9) on the same nanoparticle are presented in their respective order with camera view of the cantilever position and corresponding AFM topography image.

nanoparticle having 32 nm diameter and a measured height of 7 nm) and the fine details of the nanoparticle. On the other hand, tips 1, 3, and 6 exhibited some common tip-related AFM artifacts. For instance, the double-tip effect is evident in the image obtained by the tip in the top left corner of the piezo range, tip 1. This tip scans the nanoparticle twice, creating a ghost image.

Another tip-related AFM artifact is visible in the image recorded by the tip in the far-right corner of the piezo range, which is numbered as tip 3. The nanoparticle appears larger and distorted in its lateral dimensions, due to the tip convolution effect. This effect implies that the tip apex is blunt and irregular. The same is true for tip 6, but to a slightly lesser extent.

Altogether, the results presented here demonstrate one of the biggest advantages of the RTS SPM method. Scanning the same object with multiple tips reduces the data's dependence on AFM tip quality and thus, enables more accurate information extraction.

4.2. RTS scalpel SPM with BDD coated Si probe chip

The following experiment was conducted to evaluate the durability of the tips located on the BDD coated Si probe chip, but also the applicability of the RTS SPM method for SPM tomography measurements. SPM tomography is an innovative technique that uses a slice-and-view approach to obtain 3D images of nanoscale structures. This concept was first introduced by Xu *et al* [32], who exploited the hardness of diamond tips [33] in conjunction with the precise force control of AFM to remove thin layers of material (scalpel SPM) and subsequently probe the resulting surface in various AFM modes (e.g. electrical). Since then, this methodology has been employed to study various materials for different purposes [34–38], among which is also Si, because of its

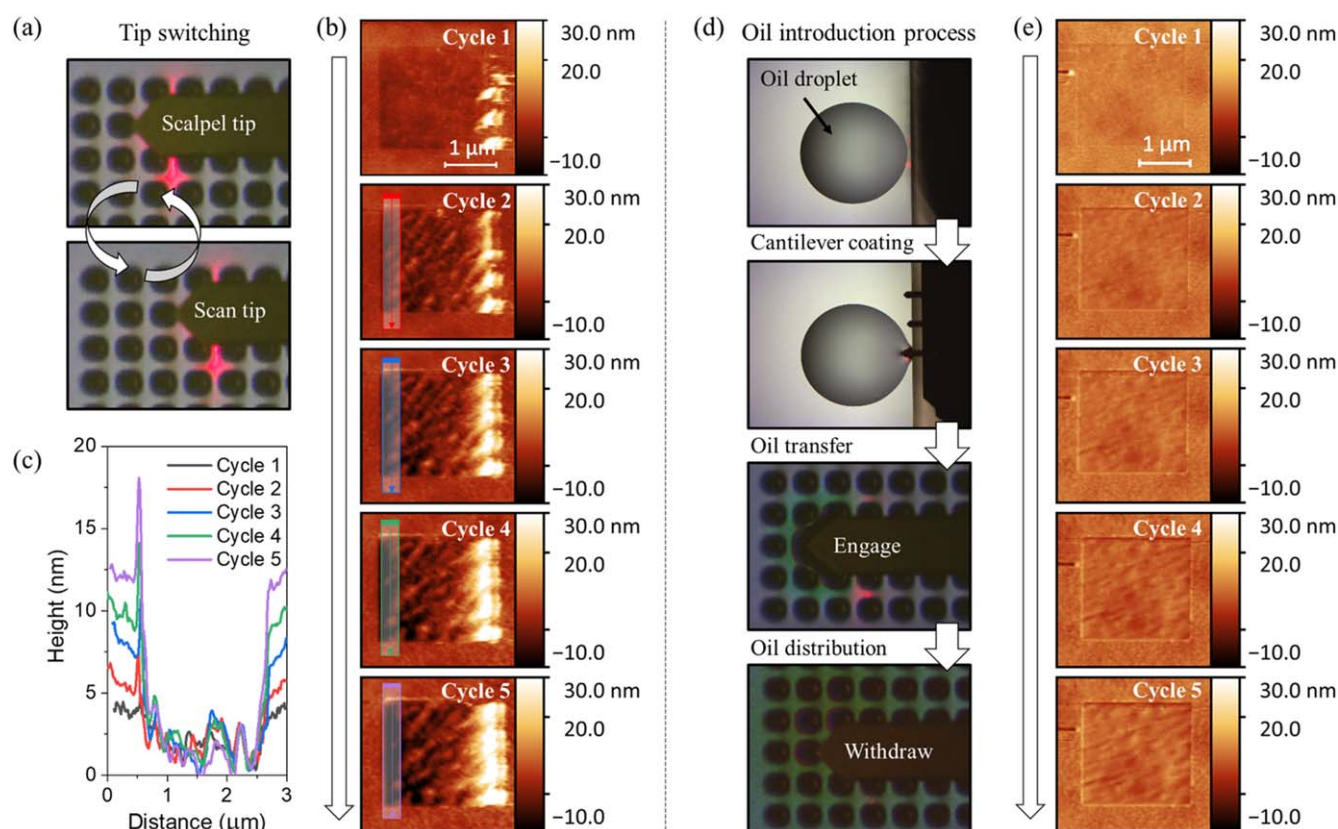


Figure 5. Scalpel RTS SPM with BDD coated Si tips. (a) Pictures taken with the AFM camera showing the cantilever moving from the tip used for material removal (scalpel tip) to the one used for imaging (scan tip). (b) Contact mode AFM images recorded after each material removal step. (c) Height profiles corresponding to each cycle, extracted along the paths indicated with arrows in panel (b). (d) Step-by-step oil introduction process represented through AFM camera captures. (e) AFM images acquired in contact mode after each one of the five cycles of scalpel RTS SPM which were performed with oil introduced.

dominance in the nanoelectronics industry [39, 40]. For this reason, we tested our probe chip on a Si surface, using a Si tipless cantilever (MikroMasch HQ:NSC35/tipless/No Al (5.4 N m^{-1})) as our sample. We performed scalpel SPM cycles in the RTS configuration, whereby each cycle had a material removal step and a subsequent topography imaging step. Both steps were performed in contact mode: the first one involved applying a high force (between 500 nN and 1 μN) to scratch the surface, while the second one used lower values of the deflection setpoint to avoid doing so. For each step we used one of two neighboring tips on the BDD coated Si probe chip. Figure 5(a) shows the AFM camera captures of the cantilever positioned above the designated tips i.e. scalpel and scan tips. Here the ability to switch tips seamlessly with the RTS SPM method is a key advantage over the conventional configuration. Although BDD-coated tips are robust enough to withstand the high contact pressures during the Si scalpel step, it should be noted that Si is a hard material by itself and hence even diamond tips are wearing off and may sometimes get damaged or contaminated by debris adhering to the tip apex [41]. This can affect the quality of the data obtained when imaging the state of the surface after the scalpel step. By using a different tip for each step, we ensure that the condition of the tip which is used to probe the material property (scan tip) is optimal, and thus the data obtained will be trustworthy. Moreover, the seamless

tip-switching capability also enhances the scalpel step itself. If the structural integrity of the scalpel tip becomes compromised in any way and the tip loses its ability to remove material, it can be easily replaced with a new one and the experiment can continue without any interruption. However, in our case, we achieved efficient material removal without damaging the tips, as shown in the series of images in figure 5(b) and the corresponding color-coded line scans in panel 5(c). In total, we performed five scalpel cycles and removed more material with each one. This can be seen in the topography images which reveal the change in the surface morphology after each cycle, but also from the height profiles where there is a clear depth increase throughout the series. By cycle 5, we created a 12 nm deep square-shaped crater. The material removal rate was about 2 nm per scan for all cycles except for the first one, where it was 3.5 nm per scan. One possible explanation for this phenomenon is that the tip undergoes some light, initial wear during the first scalpel cycle, which reduces its efficiency in removing material, but also increases its durability against further deterioration, resulting in a stable trend afterwards [42]. Another conceivable reason could be that the debris from the scratched surface reduced the initial material removal rate.

In reference to the work done by Noël *et al* [43], where the scalpel SPM experiments were conducted in an oil medium to reduce the effect of the debris on the material removal

rate, we also tested if this approach could be done in the RTS configuration. Through the series of pictures taken with an AFM camera and presented in figure 5(d) we propose the step-by-step protocol of the oil introduction for the RTS SPM experiments. First, we placed a small drop of oil (castor bean oil) on a flat surface. Then, we carefully dipped our tipless Si cantilever into the oil with the use of the AFM course motors. Next, we moved the oil-coated cantilever over the probe chip and engaged. The oil formed a localized droplet under the cantilever as soon as the contact was made. Finally, we withdrew the cantilever, leaving the oil on the probe chip. This caused the oil to spread over several tips in the area, now visible as a uniform light, green tint in the picture. In this way we coated multiple tips with oil which could now be used for our RTS scalpel SPM experiments.

In figure 5(e), we show results of five scalpel cycles through a series of AFM contact mode images. In contrast to the experiments performed in air, the dugout crater was free of debris and had a much smoother surface with an RMS roughness (S_q) of 1.5 nm. Indeed, this value is considerably lower than the 4.2 nm S_q of the crater which was created in air. This finding is remarkable, as it contradicts the lastly mentioned paper where the authors did not observe a variation in the surface roughness across the two media. However, they noted that to perform an accurate analysis of the surface quality they would require a fresh tip. This again shows the advantage of working in the RTS configuration, as it makes use of different tips for scanning and material removal.

4.3. RTS correlative SPM with BDD and Pt coated Si probe chips

As stated above, one of the major challenges when performing multimodal SPM measurements is the need to change tips for being able to measure in different AFM modes. The RTS configuration enables not only the fast and convenient tip switching but also allows to switch the probe chip. Figure 6(a) is a picture of a bare Si, BDD, and Pt coated probe chips resting on the AFM sample stage. The cantilever holding the sample can be easily navigated from one to the other with the use of the AFM course motors. To demonstrate this capability and to test the performance of our probe chips, we conducted a proxy of an RTS correlative SPM experiment which involved two different kinds of tips (Pt and BDD coated Si probe chips).

The sample was a chemical vapor deposition (CVD)-grown MoS_2 monolayer on sapphire substrate, which we transferred onto a Au-coated tipless Si cantilever by adopting a protocol used for transferring 2D materials onto transmission electron microscopy grids [44]. Figure 6(b) shows an optical microscopy image of the bottom side of the Au-coated Si cantilever after the 2D material has been transferred. The color mapping of the different regions is based on the image brightness employed for clarity. The isobright region of the image, colored in red, is a uniform MoS_2 monolayer as confirmed by Raman spectroscopy measurements (see figure S3). The homogeneity of the 2D layer was broken up just in the top right corner of the cantilever where it appears to have

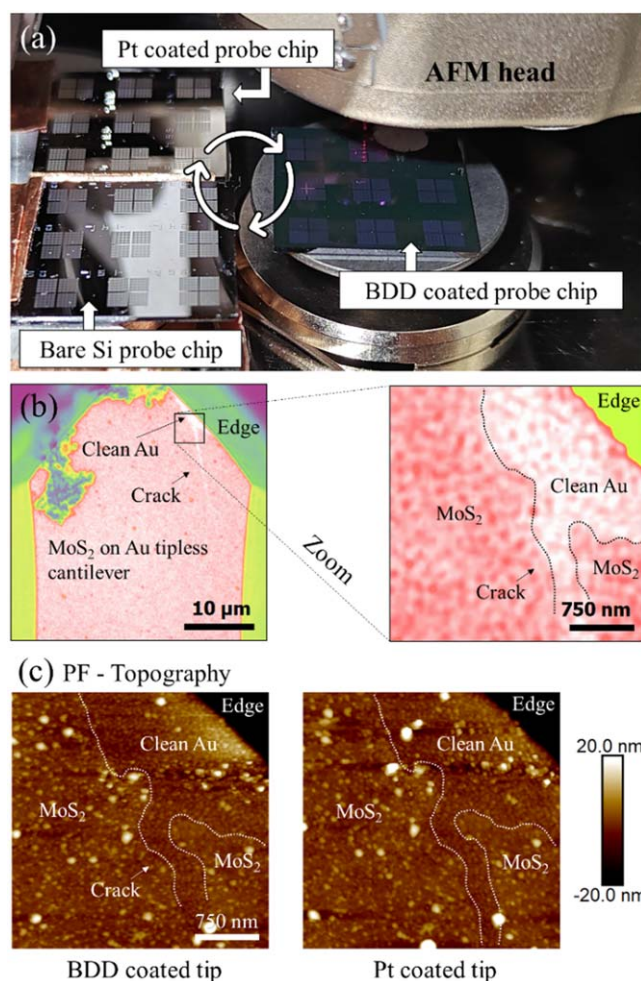


Figure 6. (a) Picture of RTS configuration with bare Si, BDD and Pt coated probe chips placed on a sample stage. (b) Optical microscopy image of the bottom side of the Au coated tipless Si cantilever, after 2D MoS_2 has been transferred. (c) Peak force topography images of the ROI recorded with BDD and Pt coated probe chips.

cracked (colored in white). The area around the crack was our area of interest (ROI) because it contains the exposed Au surface. Peak force (PF) Kelvin probe force microscopy PF-(KPFM) and PF quantitative nanomechanical mapping (PF-QNM) measurements in RTS configuration were carried out to simultaneously probe both mechanical and electrical interactions between the Pt and BDD coated tips with the Au and 2D MoS_2 surfaces. In this way, we could correlate the obtained data to the tip coating thickness, hardness, and conductivity. After scanning the ROI once with each probe chip and recording their coarse motor XY coordinates, we could quickly switch between them with minimal navigational errors (sub- $1\ \mu\text{m}$). These can be easily corrected by applying small adjustments with the use of the piezo driver. Figure 6(c) shows the topography images of the ROI recorded with both types of probe chips which were spaced centimeters apart. The pinpoint focus on the ROI illustrates the effectiveness of the probe chip switching process. Regarding the tip performance, both tips followed the surface corrugations closely, with minimal convolution effects. In the topography images, one can clearly distinguish the regions which were

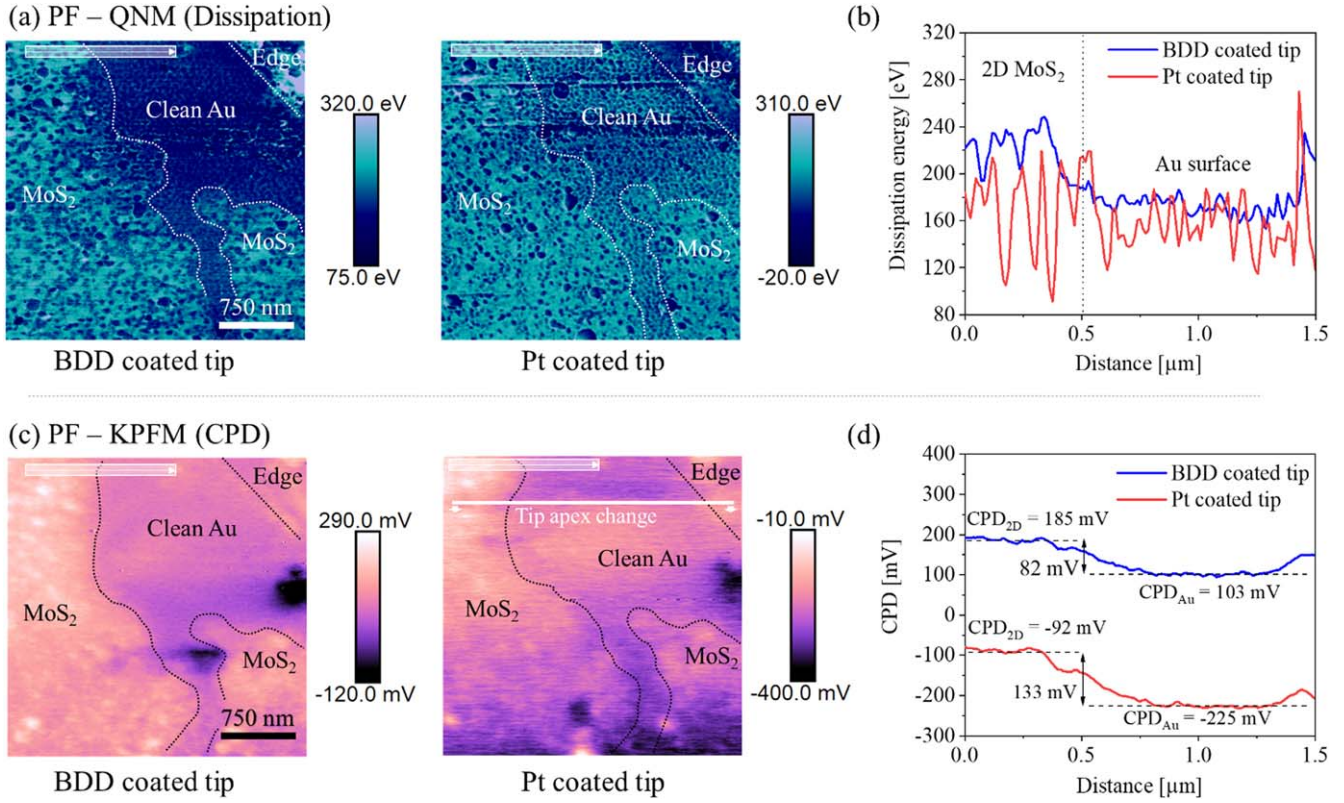


Figure 7. Examination of the Pt and BDD coated probe chip performance by qualitatively mapping mechanical and electrical responses of the surface in the ROI. (a) PF-QNM dissipation maps of the ROI. (b) Linescans extracted along the paths marked with white arrows in the dissipation maps. (c) PF-KPFM surface potential maps recorded in the same area. (d) Corresponding line scans from the images in panel (c).

covered with the 2D layer from those that were not. The clean Au surface displays a darker contrast and a grainier topography when compared to the MoS₂ monolayer. However, a closer look at the images revealed that Pt coated tips slightly outperformed the BDD coated ones in terms of resolution. This is to be expected, as the thinner Pt coating yielded a sharper tip apex, as shown in figure 3. On the other hand, based on the same SEM images, the BDD coating has a much more textured morphology. As a result, the small protrusions near the tip apex will act as highly localized interaction points between the tip and the surface. This explains why the BDD coated tips, despite having a thicker coating, still achieved a decent resolution. Figure 7(a) shows the PF-QNM dissipation maps of the ROI recorded by both tips whereby the image obtained with the Pt coated tips displayed again an overall sharper image. However, the larger apex of the BDD coated tips contributed to a better dissipation contrast across the ROI (visible in the line scans presented in figure 7(b)). This is because the BDD coated tips, being much harder, deformed the Au surface more. In such non-elastic deformations, the measured dissipation energy is dominated by the work of adhesion, which scales with the size of the contact area, i.e. the size of the tip apex [45]. Interestingly, the opposite trend is present in the KPFM surface potential maps of the ROI, which are shown in figure 7(c). We measured the contact potential difference (CPD) between two types of tips (BDD and Pt coated) and two types of materials (2D MoS₂ and Au surface). Note that the measured values are not absolute, as

the tips were not calibrated, but they are still useful for a qualitative comparison of the tip performance. This time, Pt coated tips provided more contrasting maps. The CPD difference across the MoS₂/Au boundary was 133 mV, compared to 82 mV with BDD coated tips. One reason for the better sensitivity of the Pt coated tips is the higher conductivity of the Pt coating. Another factor is the sharper tip apex, as the smaller area of tip-sample interaction is known to give a more spatially accurate measurement of the electrostatic response [46]. However, we also observed that the CPD map which was recorded with the Pt coated tip in the top-down direction had an inconsistent quality. The white line in the image marks where the morphology of the tip apex has changed. The soft metal coating can get damaged by even slight intermittent tip-sample contacts during the PF experiment. This can lower the tip conductivity and consequently, its performance. Lastly, it is worth mentioning that even though the detailed discussion of the physics behind this sample is beyond the scope of the paper, our RTS study is in good agreement with similar work done in a standard AFM configuration by Pullman *et al* [47] on a CVD MoS₂ monolayer which was transferred onto a Au surface.

5. Conclusions

In this paper, we introduced the RTS SPM concept as an innovative solution to a fundamental issue of the

conventional SPM technology: the need for frequent, tedious, and time-consuming tip replacements. To show that this idea is feasible, we have developed and demonstrated a nanofabrication process for making Si—based probe chips using ICP-RIE. Our simple and time-efficient protocol produces probe chips that contain high-quality tips thanks to a novel tip sharpening step that is integrated into it. Moreover, we expanded the potential uses of the RTS SPM concept by coating the Si tips with BDD and Pt. The performance of these coated tips, alongside the bare Si ones, was evaluated in three distinct RTS applications. Firstly, we performed RTS ensemble imaging using bare Si probe chips on diamond nanoparticles. This showcased the tip-switching capability of RTS SPM, enabling multiple-tip imaging which could prove to be invaluable for various SPM applications that demand precise data and extensive statistical analysis. Secondly, we demonstrated that the RTS tip-switching capability is a significant advantage and a critical enabler for SPM tomography measurements, a methodology of ever-growing importance in semiconductor nanoelectronics. This was achieved through scalpel RTS SPM experiments using BDD probe chips in both air and oil environments. The tips proved to be highly efficient in removing Si material across both media. Our study also highlighted that a quick replacement of worn-out or damaged tips enhances the effectiveness of the scalpel SPM method. Furthermore, the use of separate tips for scanning and material removal revealed phenomena that were previously obscured in similar measurements conducted in the standard AFM configuration. And lastly, we have demonstrated the potential of RTS SPM for correlative SPM measurements by doing PF-KPFM/QNM on 2D MoS₂ which was transferred onto a Au coated tipless Si cantilever. We switched between different types of tips (Pt and BDD coated ones) seamlessly in the same experiment without losing the area of interest. We also compared their performance and revealed how the interplay between tip apex size, hardness and conductivity affected the data obtained.

Like any early-stage technology, the RTS SPM concept faces some initial challenges, notably in refining sample preparation methodologies. Nonetheless, we are confident that investing additional efforts in its development is worthwhile, given its potential to open new horizons in SPM research and further advance the state-of-the-art in SPM technology.

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Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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