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Towards easy and reliable AFM tip shape determination using blind tip reconstruction

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ABSTRACT

Quantitative determination of the geometry of an atomic force microscope (AFM) probe tip is critical for robust measurements of the nanoscale properties of surfaces, including accurate measurement of sample features and quantification of tribological characteristics. Blind tip reconstruction, which determines tip shape from an AFM image scan without knowledge of tip or sample shape, was established most notably by Villarrubia [J. Res. Natl. Inst. Stand. Tech. 102 (1997)] and has been further developed since that time. Nevertheless, the implementation of blind tip reconstruction for the general user to produce reliable and consistent estimates of tip shape has been hindered due to ambiguity about how to choose the key input parameters, such as tip matrix size and threshold value, which strongly impact the results of the tip reconstruction. These key parameters are investigated here via Villarrubia's blind tip reconstruction algorithms in which we have added the capability for users to systematically vary the key tip reconstruction parameters, evaluate the set of possible tip reconstructions, and determine the optimal tip reconstruction for a given sample. We demonstrate the capabilities of these algorithms through analysis of a set of simulated AFM images and provide practical guidelines for users of the blind tip reconstruction method. We present a reliable method to choose the threshold parameter corresponding to an optimal reconstructed tip shape for a given image. Specifically, we show that the trend in how the reconstructed tip shape varies with threshold number is so regular that the optimal, or Goldilocks, threshold value corresponds with the peak in the derivative of the RMS difference with respect to the zero threshold curve vs. threshold number.

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1. Introduction

The atomic force microscope (AFM) is a versatile and powerful tool for the analysis of topographical and interfacial properties of surfaces with nanoscale resolution. Quantitative AFM is limited by the geometry of the probe tip used to scan the surface, which means that accurate knowledge of the AFM tip shape is critical for accurate dimensional [1], nanomechanical [2], frictional force [3], chemical force [4], electrical force [5], and magnetic force measurements [6]. Typical scanning probe imaging modes allow a tip of finite geometry to move over a sample surface while maintaining a nominally constant tip-surface separation in order to render a topological image of the surface. In the language of mathematical morphology, the topographical image produced is the dilation of the surface by the tip (more specifically the tip reflected through its apex) [7]. Note that dilation is not the same as convolution, since convolution is a linear mathematical process, and the process of dilation, by which an image is created by the physical interaction of tip and sample, is non-linear [8].

As is shown in Fig. 1, a tip that is sharper than the surface features will more accurately reproduce those features in an image (Fig. 1a) than a blunter tip (Fig. 1b). Images produced with a tip whose geometrical features are of the same order as or larger than the surface features (Fig. 1b) will produce image features that can be significantly broadened relative to the true surface geometry. Such measurements therefore cannot be relied upon for accurate lateral dimensional measurements of nanoscale surface features. While conventional AFM imaging exhibits some fundamental limitations, including the inability of the tip to image undercut features (as exemplified by a tip imaging a nearly spherical feature in Fig. 1), some techniques have been developed to minimize these limitations [9,10]. Nevertheless, the measured height of imaged features in conventional AFM will usually be accurate even though the image widths may not be, provided that the AFM is in dimensional calibration and the sample is sufficiently stiff to withstand imaging forces.







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Fig. 1. Representations of AFM image profiles (bold line) produced for a surface with sharp and spherical features when imaging with (a) a sharp tip or (b) a blunt two-peak tip.

Ever since the AFM was invented, users have sought sharp, robust, reproducible tips [11] of many different materials, including carbon nanotubes [12] or hard materials such as ultrananocrystaline diamond [13]. While quantification of tip size by the manufacturer is useful, even the most robust tip can experience wear or mechanical shearing due to continuous or intermittent contact with a surface during an AFM experiment [14,15], especially if that experiment is performed in contact mode [16,17]. The tip can also pick up contamination, such as nanoparticulates or surface moieties, which can alter the chemistry and shape of the tip. Hence, for the most accurate tip shape determination, characterization is required before, during, and after an experiment.

There are two main methods used to characterize an AFM probe tip shape [1] (a) ex situ direct imaging of the tip (typically using electron microscopy) and (b) in situ indirect analysis that leverages the fact that an AFM image is the mathematical dilation of the sample by the tip shape. Direct imaging is typically accomplished using either a scanning electron microscope (SEM) or a transmission electron microscope (TEM). An SEM operated in backscattered mode can produce images of an entire tip (including apex, shank, and cantilever). Even some of the details of the tip apex on the order of tens of nanometers [18-20] can be determined, and resolutions on the order of nanometer may be achieved with high resolution SEMs [21]. For nanometer and subnanometer resolution of AFM tip features, a TEM is often used, although a TEM is typically limited to two-dimensional (2-D) shadow imaging unless multiaxis rotation stages within a TEM sample holder are available. Other disadvantages of electron microscopy include the potential for contamination by electron beam-ionized residual gas [22-24], the time-intensive and impractical necessity of removing the tip from the AFM, the difficulty in determining the exact region of the tip that would contact the surface, and the inability to image electrically insulating AFM cantilevers and tips since material charging may prevent stable high resolution imaging. In addition, immediate access to high quality electron microscopy instruments may be limited for some users. Additional complications can occur when extracting the exact tip shape from electron microscope images because of intrinsic distortion and convolution effects in the images [25].

Indirect tip shape determination methods use AFM images to determine the tip shape from samples whose surface geometries may be known or unknown [1]. Examples of tip characterization samples of assumed or known shape include, but are not limited to: gold spheres and polystyrene spheres [26,27], cone-like structures [28], holes and trenches [29], and carbon nanotubes [30]. In these cases, deviations in the sample features from their ideal

size and shape, especially for nanoscale structures, will introduce errors in the tip shape estimation. One must be particularly careful that reference samples do not change significantly with use (e.g. wear or contamination) [1], or that a method to re-characterize the sample geometry exists.

If the surface geometry is unknown, an upper bound to the tip shape can be extracted from the image in a process known as blind tip reconstruction [7,31,32]. Any sample can be used, but a sample with sharp features (e.g., TipCheck or NioProbe [MicroMasch, San [ose, CA]) provides blind tip reconstruction algorithms with more complete tip shape information, leading to more accurate tip reconstructions. Section 2 describes the mechanisms of the blind tip reconstruction method in detail. Blind tip reconstruction is advantageous because it can be performed in situ. On the other hand, commercial blind tip reconstruction algorithms can appear intractable to a user who seeks to quickly and easily determine their tip shape, specifically since the blind tip reconstruction process depends on a number of input parameters whose values are not obvious or intuitive and yet have a large impact on the resulting tip reconstruction. There are currently few guidelines on how these parameter values should be chosen.

Here, we introduce a methodology that enables simple and reliable tip shape determination when using a blind tip reconstruction method for a given surface. Our main objective here is to present clear protocols for the use of blind tip reconstruction algorithms as a straightforward and reliable method to determine tip shape, which could form the basis of an international standard under the auspices of ISO/TC 201/SC 9 in scanning probe microscopy. The algorithms used for this work are developed from those first published by Villarrubia [7], and they are implemented in MATLAB (MathWorks, Natick, MA). The MATLAB code used for the results in this paper will be available as open source code on the online Nanoprobe Network Software Library. In this paper, these algorithms are applied to simulated AFM images (generated from known simulated surfaces and tips) from which the tip shape is then reconstructed through the variation of relevant parameters. We thus demonstrate how a general user can reliably determine the optimum reconstruction parameter values for their images.

2. Summary of the mathematics of blind reconstruction

There have been many contributions to the theory and practical implementation of blind reconstruction [7,8,31–39], and many of these and related methods have been reviewed here [1]. In this paper we focus on the blind tip reconstruction algorithms developed by Villarrubia [7]. Although improvements of these algorithms have been made since Villarrubia's initial publication of his code [1], the fundamental core of these methods has remained the same. The mathematics of blind tip reconstruction depends on a number of input parameters that must be optimized to generate a best estimate for the tip shape. For these reasons, a physical description of the pertinent mathematical algorithms and related parameters is presented here for completeness and in order to make the mathematics involved more intuitive.

Within blind tip reconstruction, the tip shape is determined from an AFM image using an iterative process [7], which is represented schematically in Fig. 2 and will be described in the paragraphs that follow. While the original theoretical development of blind tip reconstruction uses the language and symbolism of mathematical morphology, the implementation of these ideas within Villarrubia's work (i.e., as implemented in the code) [7] can also be described in terms of the geometry of the tip and sample and their modification in the tip reconstruction process. We present a description of this "geometric representation" of the blind tip reconstruction to illustrate, from a slightly different



Fig. 2. A flowchart representing the key features of the blind tip reconstruction process. After initializing the tip shape estimate and reconstruction parameters, an image coordinate x' is selected, and then a tip coordinate x is selected. For each permissible value of d (as dictated by Eq. (1)), the value of *temp* is calculated. The minimum absolute value for *temp* is determined and set equal to *dil*. If the variation of the image geometry near the tip contact point is sufficiently large compared with *thresh* (Eq. (3)), then the tip is modified at point x. This process is iterated for all possible x and x' values until convergence occurs.



Fig. 3. Inverted tip and representative image profiles, indicating the application of the criterion of Eq (1). (a) A representative snapshot of one possible position where the tip contacts the surface at tip coordinate *d* to produce the image point at *x'*. In this location, tip estimate modification does occur because the apex of the tip profile, located at $x = x_c$, is below the image profile (as dictated by Eq. (1)). (b) A representative snapshot of a position where it is not possible for the tip to contact the sample to produce the image profile, as the apex ($x = x_c$) of the inverted tip profile lies above the image profile. This case represents the situation where the upright tip apex would penetrate the sample, which is physically impossible. Therefore this tip coordinate *d* cannot be used to modify the tip shape at *x*.

perspective, the main features of these algorithms. In this paper, we use the same mathematical nomenclature as in Ref. [7].

The reconstruction procedure is shown schematically in Fig. 2 and progresses as follows. First an initial estimate for an upper bound for the tip shape is chosen. We refer to the estimated tip shape hereafter as the tip estimate. Typically, a "square pillar" with a flat top is used, where the surface of the tip consists of a matrix of zeros with dimensions that match the number of pixels chosen for the size of the tip estimate. For example, a 20×20 pixel tip estimate is a 20×20 matrix of zeros. Since by definition the height of the tip apex is fixed at zero height, the entire surface of the tip lies at the same height as the apex. As the tip estimate evolves

during the tip reconstruction process, tip heights may change, but all will remain less than or equal to zero. A 2-D profile of a representative matrix of zeros, i.e., a square pillar tip, is shown as a rectangle in Figs. 3 and 4. Other initial tip estimates may be used, but this is the most general starting point.

Note that in Fig. 1b the tip geometry appears in the image as if the tip were reflected through its apex, i.e., as an inverted tip; this is necessarily the case because the image is the dilation of the surface by an inverted tip [7]. Therefore, for the purposes of tip reconstruction, the tip estimate is inverted (shown in Fig. 3 and in the center of Fig. S1a in the Supplementary Material). After the initial tip estimate is defined, the tip estimate is refined through comparison of the current tip estimate to the image geometry at every point in the image.

Two-dimensional profiles of a representative image and an initial tip estimate are shown in Fig. 3 and also in more detail in Fig. S1. (An extension to three dimensions is straightforward.) Coordinates on the image are given as x' and image heights are represented by the function I(x'). There are three coordinates of interest for the tip: a general tip coordinate x, the (fixed-height) center point of the tip x_c , and the tip-image contact point d. This point d on the tip corresponds with a chosen point x' on the image as a possible tip-sample contact point. P(x) is the height of the tip at any coordinate x, and is always less than or equal to zero, since P(x) represents the inverted tip.

After the tip estimate and reconstruction parameters are initialized, the algorithm iterates through all possible values for x' (contact point) and x (tip refinement position). To determine if refinement can occur, the tip estimate is compared to the image geometry for all possible contact points (all possible d values). Modification of the tip estimate at x occurs only when

$$P(d) > I(x') - I(x' + x_c - d)$$
(1)

Geometrically speaking, the criterion described by Eq. (1) allows for modification only if the tip apex coordinate x_c is below the image profile for the particular *d* being considered. This is illustrated in Fig. 3, where two representations of potential *d*

contact points are shown. For the profile in Fig. 3a, refinement occurs because the tip apex is below the image profile; whereas refinement does not occur for the profile shown in Fig. 3b because the tip apex lies above the image profile. If Eq. (1) is satisfied, the tip apex never deviates from a zero height value, i.e., $P(x_c)=0$.

Next, the extent of tip estimate modification at tip coordinate x is determined. For every allowable tip refinement position, with contact point at d on the tip and x' on the surface (represented in Fig. 4a), the value of

$$temp = I(x' + x - d) - I(x') + P(d)$$
(2)

is calculated. The variable *temp* represents the difference between the height of the image at x, i.e. I(x' + x - d), and the height of the image at x', i.e., I(x'). The value of *temp* is computed for all allowable d values and the minimum absolute value of *temp* is saved as the variable *dil* (Fig. 4b). Once *dil* is found, the threshold criterion is applied to determine if tip estimate modification actually occurs. The height of the tip at x is modified if *dil* is large enough compared to the threshold value, *thresh*. This criterion for tip estimate modification at x is represented mathematically as

$$dil < P(x) - thresh \tag{3}$$

If Eq. (3) is true, then the modification occurs (as is shown in Fig. 4c, where thresh=0 for simplicity) and the tip height at x is modified to

$$P(x) = dil + thresh \tag{4}$$

In the case of Fig. 4, initially thresh=0 and P(x)=0; therefore since it is always true that dil < 0, then Eq. (1) is true. Eq. (4) gives the extent of the modification at x, which in this case is P(x)=dil. If *thresh* was non-zero and *dil* is not large enough compared with *thresh* (i.e., Eq. (3) is false), then the point P(x) would have remained unmodified.

The process described above is applied at every tip coordinate x for the single image position x'. If the threshold value *thresh* is not zero, the modification will be less pronounced. For example, let the value of *thresh* be 1 nm. Then P(x) will be modified as long as



Fig. 4. The process of modifying or refining the initial tip estimate. (a) The tip-surface image contact point x=d is varied among all permissible points. The variable *temp* records the difference in height between the contact point and the height of the image above tip location x. (b) *dil* is determined as the minimum absolute value of *temp* found by varying d, or *dil*= – mini*temp*!. (c) The tip height at x is then modified to a new height so that it is lower than the previous height by the magnitude of *dil*. In this example x is located at the rightmost edge of the tip, and hence the modified portion of the tip is one pixel wide.

dil < -1 nm. This criterion makes it less likely that P(x) will be modified because the variation of the image profile over the distance of x-d will need to be larger than 1 nm to produce a large enough dil for the criterion $dil = \max(I(x' + x - d) - I(x'))$. Then when the modification does occur (Eq. (1) is satisfied), the new value of P(x), which is equal to dil+thresh, will not be as large (in absolute value), since dil < 0 and thresh > 0 (1 nm in this example case, so P(x) is 1 nm smaller in absolute value). The process is repeated at every image location x' within the image, and then the process iterates through the image again until the tip estimate P(x) converges.

As demonstrated in the above discussion, the input parameter *thresh* (the threshold value) is critical for the resulting tip reconstruction. If an image has no noise, then that image and the resulting tip reconstruction only contain information related to the tip and sample geometries. However, if the image contains noise, that noise could be mistaken in the tip reconstruction algorithms as pertaining to the tip geometry. The threshold parameter accounts for noise through the algorithms discussed above and discussed in reference [7].

The tip reconstruction user controls the threshold parameter, as well as the size of the initial tip shape estimate, namely its width (the size of the tip matrix). From a practical standpoint, some clear guidelines are necessary for selecting appropriate values of *thresh* and tip matrix size in order to produce an optimum tip reconstruction. Some useful suggestions for choosing these values have been given [7,20,35,37], but we wish to provide a more concrete methodology for determining these parameters. Regarding tip matrix size, there is a fairly large permissible range for this parameter over which the tip reconstruction will be essentially independent [7]. Specific suggestions on how to choose the tip matrix size are described in detail in Section 4.2.

The second major implication from the Villarrubia work is that there is a reproducible trend in the shape of the tip reconstruction as a function of threshold value. If the threshold value is too low in magnitude, the tip reconstruction is dominated by high frequency noise from the image. If, on the other hand, the threshold value is too high, no features on the image are sharp enough to allow for a modification of the initial "square pillar" tip geometry. Consequently, the reconstructed tip becomes unrealistically blunt. The optimum *thresh* value falls between these two extremes and is referred to here as the "Goldilocks" threshold (neither too big nor too small!).

3. Generation of simulated samples, tip, and images

3.1. Initial development of simulated sample surfaces, tip, and images

To test the capabilities of these blind tip reconstruction algorithms and determine criteria for selecting tip reconstruction parameters, a set of simulated images were produced by mathematically dilating a simulated sample with a simulated tip. This process assumed non-deformable contacts. The simulated tip is \approx 38 nm wide at the base with a maximum height of 254 nm. This tip also features two protrusions at right angles to each other, with their respective apexes positioned at 19 nm and 29 nm below the primary tip apex (Fig. 5). This simulated tip has no reflectional or rotational symmetry to provide the most general tip shape. Blind tip reconstruction using simulated images, where the tip shape is known, helps us determine optimal tip reconstruction parameters when knowledge of the tip shape is not available.

A number of distinct simulated surfaces were used; a representative set of these simulated surfaces and their corresponding images are shown in Fig. 6. These surfaces were designed to be analogous in shape to typical AFM samples which could be used to characterize the AFM tip shape, including: spikes (S); ridges (R), such as those formed by carbon nanotubes; and pitted, anodized alumina (P). The surfaces are not optimized to give tip reconstructions that most accurately reproduce the actual tip shape, but are simply meant to represent relevant tip reconstruction surface geometries. All images consist of 512×512 pixels, where each pixel is 1 nm in length along the *x*- and *y*-directions. In the *z*-direction, the maximum peak-tovalley height for all images was ≈ 290 nm.



Fig. 5. The process of creating simulated images for evaluating the MATLAB-based blind tip reconstruction algorithms. The features of the S surface (top left) are mathematically dilated by the simulated tip shape (top right) to produce the S image (top center). From this image, various tip reconstructions are produced and the Goldilocks tip reconstruction is identified (bottom).

The process of dilating the simulated sample and the simulated tip is represented schematically in Fig. 5. From these simulated images, tip reconstructions were produced by varying the threshold parameter over a sizeable range. For each image and set of tip reconstructions, the Goldilocks tip reconstruction was first identified by comparing the resulting tip reconstructions with the original simulated tip. The procedure for determining the Goldilocks tip reconstruction in cases where the tip shape is not known is explained in Section 5.

3.2. Addition of noise to the simulated images

Tip reconstructions were generated from simulated images with added noise so as to use the tip reconstruction algorithms to their full extent. Varying amounts of noise were incorporated into the simulated images, examples of which are shown in Fig. S2 in the Supplementary material. A 512×512 pixel white noise image was generated with a root mean square (RMS) value of 1 nm, and this noise image was scaled by a factor proportional to each simulated image's z-range and then added to that image. For example, to create an image with a signal-to-noise (S/N) ratio of 160 where that image has a z-range of 200 nm, an image containing only white noise with an RMS height value of (200/160) nm was added to the original image. In this paper, each image is referred to by its identifying letter (corresponding to the samples and images shown in Fig. 6) and the signal to noise ratio of the added noise. For example, an image produced using the R sample with an S/N ratio of 160 is referred to as R-160. For the analysis



Fig. 6. Representative simulated surfaces and resulting images used for evaluation of MATLAB-based blind tip reconstruction. The numbers indicated on the vertical scale bar in this figure and all following figures have units of nanometers. (a) Spikes (S): Flat-top spikes with an in-plane apex radius of 3.5 nm and side walls of slope 95 nm/nm (analogous in shape to NioProbe from Aurora Nanodevices or TGT1 from NT-MDT). (b) Ridges (R): Ridges with a roughly parabolic profile (analogous in shape to TipCheck from Aurora Nanodevices). (c) Pits (P): A series of paraboloidal pits (similar to anodized alumina).

that follows, tip reconstructions were performed on both noisy and noiseless images.

4. Details of the MATLAB-based blind tip reconstruction algorithms

The blind tip reconstruction algorithms discussed in this paper were coded in MATLAB and its algorithmic structure is diagramed in Fig. 7. The core of these algorithms is the original code published by Villarrubia [7]. This code, written in the C programming language, is directly accessed by the MATLAB algorithms. The original implementation in MATLAB was accomplished by Todd and Eppell [35] and allowed the user to produce one tip reconstruction at a time based on one set of tip reconstruction parameters. We augmented these algorithms to produce a set of tip reconstructions for a range of threshold values, and to enable real-time or off-line visualization analysis and facilitate the process of determining the Goldilocks tip reconstruction. The general process of the algorithms' implementation by users is as follows.

4.1. Import image into MATLAB

First the user imports an AFM image into MATLAB. The images imported into MATLAB for analysis can either be NanoScope data files (Bruker AFM, Santa Barbara, CA) or a text file. Multiple images can be loaded at the same time for rapid, serial image analysis. It is best if the user apply a low pass or median filter on the image before importing the image into MATLAB.



Fig. 7. The procedural steps of the MATLAB-based blind tip reconstruction from the user's perspective. The primary functionality added in this work is the ability to generate multiple tip reconstructions for multiple threshold values (bottom loop). These steps are described in detail in the sections indicated on the diagram.

4.2. Choose matrix size for reconstructed tip

The user then selects the matrix size for the reconstructed tip. As discussed in Section 2, the fidelity of the Goldilocks tip reconstruction to the known tip estimate is invariant to the tip matrix size within a certain range [7]. When optimizing for both accuracy and time efficiency, the user must choose a tip matrix size encompasses a sufficiently wide range of image features, but one that is not so large as to become overly time intensive. The general guideline is to choose a tip matrix size that has approximately the same lateral dimensions as the largest recognizable tip artefact in the image. For example, in Fig. 8 one can see in image S-80 the repeated feature resembling the

tip. The user chooses a tip matrix size equal to the dimension of one of these features; this corresponds to a square region of about 40×40 nm for the examples in Fig. 8. This choice of the tip matrix size sets the initial tip estimate. For the P sample, the tip matrix size is approximately the width of the ridges between holes, as seen in Fig. 8c.

4.3. Choose the threshold values

The user then chooses the threshold values to be used to produce tip reconstructions. Each threshold value will correspond to one tip reconstruction. Since the threshold values are typically



Fig. 8. Examples of how to choose the tip matrix size using (a) image type S, (b) image type R, and (c) image type P. By identifying the size of a representative image feature, one can determine the appropriate choice of approximate tip matrix size to input into the blind tip reconstruction algorithm. This size need not be exact, since the resulting tip reconstruction is insensitive to the tip matrix size within a reasonable range [7].

proportional to the image *z*-range, the user's choice is scaled relative to the image *z*-range. It is suggested that a first rough set of threshold numbers should be integer multiples of 5% of the image *z*-range. Each threshold value is then tracked with a designated threshold number to represent each threshold value. For example, if five threshold numbers were desired and the intervals were multiples of 5% of the image *z*-range, the threshold values would be 0%, 5%, 10%, 15%, and 20% of the image *z*-range, and would be designated with threshold numbers of 0, 1, 2, 3, and 4, respectively. Image S-80 has a *z*-range of 214.7 nm; hence, these threshold numbers would correspond to threshold values of 0 nm, 10.7 nm, 21.5 nm, 32.2 nm, and 42.9 nm, respectively.

For maximum time efficiency of the tip reconstruction algorithms, threshold numbers are applied in reverse numerical order, starting with the largest threshold number. The first refinement begins with the initial zeros matrix (square pillar) and applies the largest threshold value. The tip shape estimate corresponding to the largest threshold value is then used as the initial tip shape estimate for the next highest threshold number reconstruction. This process is repeated until all threshold numbers have been applied and their tip reconstructions completed.

4.4. Choose additional thresholds if necessary

After the tip reconstructions are complete, the program displays profiles of the tip reconstructions, and the user may analyze individual, three-dimensional tip reconstructions of interest. This first pass is made intentionally coarse to quickly assess the range of approximate values for the Goldilocks threshold parameter. The user is then given the option to produce additional tip reconstructions with a finer step (perhaps 0.1 threshold number steps, or steps of 0.5% of the image *z*-range, for the example given above) to isolate a particular set of tip reconstructions to refine further. We show in Section 5 that the blind tip reconstructions are similar within a range of threshold values, so the exact size of the threshold step is not critical.

5. Determining the Goldilocks threshold value and tip reconstruction

Determining the optimum value for the threshold parameter for a given sample is the most important part of the blind tip reconstruction process whereby the most accurate tip reconstructions are obtained for that sample. By applying *a priori* knowledge of the tip shape used to produce our simulated images, we have developed a method to guide the user in determining the Goldilocks threshold value when *a priori* knowledge of the tip shape is not available. We first discuss this methodology in the rare case where the actual tip shape is known (Section 5.1), and then discuss the more common case when the actual tip shape is not known (Section 5.2). We have applied these procedures to all the surfaces in Fig. 6, with images produced using a range of noise values (from no noise to an S/N ratio of 20). We have also analyzed several other sets of simulated images, as well as real AFM images. The trends discussed below are similar to the trends seen for all the images analyzed.

5.1. Determining the Goldilocks tip reconstruction when the actual tip shape is known

Since the simulated images were used in this study, the resulting tip reconstructions were compared to the known actual tip shape and also the tip reconstruction generated from a noise-less version of each images set. We use the actual tip shape for comparison since the main goal of a tip reconstruction user is to obtain the tip reconstruction that most accurately represents the actual tip shape. Since the simulated samples have finite geometry, even the most ideal tip reconstructions (those generated from images without noise) are not expected to exactly match the actual tip shape. For this reason, we present a comparison to the ideal noiseless image tip reconstruction, as others have previously done [7,39].

Fig. 9a shows an example set of *x*- and *y*-direction profiles of reconstructed tips produced from one of the images, in this case P-80. Each tip profile along a given axis was constructed using a



Fig. 9. Representative example of the result of a blind tip reconstruction, here from the P-80 profiles. (a) Twenty separate blind tip reconstruction profiles are shown, with threshold values ranging from 0 nm to 107 nm. (b) Selected blind tip reconstruction profiles with threshold numbers close to the Goldilocks threshold number are shown for clarity.

different threshold value. Note that, as expected, for very large threshold numbers, the tip shape estimate is identical to the original square pillar, and for small threshold numbers, the tip reconstructions are dominated by image noise and therefore tend to be unrealistically sharp. For clarity, some of the profiles have been removed in Fig. 9b to show only the profiles that most closely match the known tip profile (which is shown in black in Fig. 9). By visual inspection, the largest tip with reasonable geometry that matches the known tip profile can be chosen as the best tip shape estimate. For this particular set of tip reconstructions generated from P-80, the best visual match occurs at threshold number 1.4 (corresponding to a threshold value of 15.03 nm).

Although visual matching is sufficient when the actual tip shape is known, a more quantitative and systematic method to determine the Goldilocks tip reconstruction is necessary, since in most cases the actual tip shape is not known. A comparison of the profiles using the RMS difference between the known and reconstructed profiles is used to accomplish this task, which was previously used in [7]. Various other quantitative measures exist and could be used, such as the tip profile area [20], tip shape volume [10,39], or a tip radius approximation [38]. Each technique comes with its own advantages and disadvantages. For the case of tip profile area or tip shape volume, one must be careful when comparing areas or volumes, since the same cross-section area or volume could result from drastically different profile shapes. For example, a jagged double peaked tip shape could have the volume as a smooth parabolic shape. Therefore the area or volume does not constitute a unique measure of the relative accuracy of profiles or 3D tip shapes. The use of the radius of curvature to quantify the accuracy of a tip shape is not general enough for our purposes, because a parabolic fit that clearly identifies a tip radius may not always be possible or appropriate, as the tip may not resemble a parabolic shape. The RMS difference, on the other hand, gives a quantitative measure of comparison, which indicates how much a particular profile deviates from a reference profile, without making any assumptions about the tip shape. The RMS difference comparison can be made even when the tip reconstruction shape vary drastically or when no well-defined tip radius can be determined. The RMS difference, as will be shown in this section, is a reliable measure of deviation of a reconstructed tip profile from another tip profile (such as the known tip profile). Based on the definition of the RMS difference, it is expected the threshold value for the Goldilocks tip reconstruction corresponds to a minimum RMS difference between either the known actual tip shape profiles and reconstructed tip profiles or the noiseless tip reconstruction profiles and the noisy tip reconstruction profiles.

Fig. 10a shows the RMS difference between the reconstructed *x*- and *y*-profiles and the respective known tip profile as a function of threshold (threshold number on the bottom horizontal axis, threshold value on the top horizontal axis) for the P-80 image as an example. A definitive minimum in the RMS difference data is not apparent for both *x*- and *v*-profiles even though the best visual match for this set of tip reconstructions occurs at threshold number 1.4. The lack of a welldefined minimum in the RMS difference curve of Fig. 10a occurs because the reconstructed tip geometry does not closely match the known tip geometry far from the tip apex. This deviation arises from the fact that an AFM-type image will only include a relatively small percentage of image points that contain information about the lower points along the tip shaft, as has also been seen in other work [20]. Deviation in the reconstruction between the two peaks of the actual tip is also expected for the same reason [20]. Therefore, estimated tip shape information far from the tip apex can be misleading, especially in context of the RMS difference curve of Fig. 10a. This motivates a restriction on the range of profile comparison to points near the tip apex. This restricted range lies between the vertical dashed lines in Fig. 9. Thus, the restricted range coincides with the point at which the actual tip profile and the reconstructed tip profiles cross over one another. When this restriction is applied, the resulting RMS difference curve (Fig. 10b) shows a distinct minimum occurring at the threshold value corresponding to the Goldilocks tip reconstruction. A minimum also occurs at the same Goldilocks threshold value when plotting the RMS difference relative to the noiseless image tip reconstruction vs. threshold. (See Supplementary material for more detail.)

However, in reality the actual tip shape and the noiseless image are unavailable to the user, so these examples are only for proof of concept. Therefore a methodology is needed for cases where neither the actual tip shape nor the noiseless image is available.



Fig. 10. Plots of RMS difference between the actual tip shape and each tip reconstruction as a function of threshold value for image P-80. These plots are used to determine the Goldilocks threshold for the case when the true tip shape is known. On these graphs and the ones that follow, the lower horizontal axis gives the threshold number, and the upper horizontal axis gives the threshold value in nm, which is proportional to the threshold number. (a) RMS difference relative to the known tip profile as a function of threshold. (b) RMS difference relative to the known tip profile with restricted profiles.

5.2. Determining the Goldilocks tip reconstruction when the tip shape is unknown

Since the reconstructed tip profiles change as a function of threshold value in a systematic way, the trend in the RMS difference relative to a reference profile as a function of threshold number can be used to determine of the Goldilocks tip reconstruction when the actual tip shape is unknown. Any of the various tip reconstruction profiles could be used as a reference, but the profile corresponding to a threshold value of zero is used. In other words, the RMS difference curve in Fig. 11a is tracking the deviation of the tip reconstruction profile shape relative to the tip reconstruction profile produced when all of the image information is used (including image noise). Specifically, the RMS difference between

a given profile and the zero threshold profile is calculated for each threshold number.

The methodology to find the Goldilocks threshold value is described here using the representative examples in Fig. 11, and this methodology was found to work for all the images analyzed. Starting at zero threshold number, the RMS difference relative to that at zero threshold increases fairly slowly with threshold value (with a relatively modest slope from threshold number 0–1.2). As the threshold increases, the slope of the curve drastically increases as the threshold number 1.2–1.4). The Goldilocks threshold (with a high slope from threshold number 1.2–1.4). The Goldilocks threshold occurs reliably after an abrupt (high slope) transition region, e.g., just after this high slope region (threshold number 1.4) in Fig. 11a. As the threshold increases past the Goldilocks threshold,



Fig. 11. Plots of RMS difference and its derivative between the zero threshold profile and each tip reconstruction as a function of threshold value for image P-80 for (a) & (b) and P-20 for (c) & (d) as examples. These plots are used to determine the Goldilocks threshold without directly referring to the true tip shape. (a) RMS difference relative to the zero threshold profile as a function of threshold for P-80. (b) Derivative of RMS difference (derivative of plot in (a)) as a function of threshold. The Goldilocks threshold corresponds to the global maximum of the derivative for P-80. (c) RMS difference relative to the zero threshold profile as a function of threshold for P-20 showing a less step gradient than that of (a). (d) Derivative of the RMS (derivative of plot in (c)) as a function of threshold for P-20. Even when the changes in slope are less dramatic in the RMS difference plot, the derivative plot (d) clearly shows a peak that corresponds to the Goldilocks threshold for this image.

the slope of the RMS difference curve reduces again (with a low slope again from threshold number 1.4 onward). In general, the Goldilocks threshold is consistently identified as the threshold for which the greatest change in slope occurs for the RMS difference relative to the zero threshold. This trend in the RMS curve is consistent regardless of surface type or level of noise (including, but not limited to, the simulated images shown in Fig. 6 & S2 and real AFM images), and is also consistent with the trends seen by others [20,35]. The physical explanation for this phenomenon is that the abrupt transition between unphysically sharp tips to the Goldilocks tip shape occurs when the threshold is just large enough to exclude image noise in order to produce the largest tip shape consistent with the true dilation of tip and sample [35].

An alternative way to find the Goldilocks threshold value is to calculate the derivative of the RMS difference relative to the x-direction and y-direction zero threshold profile. Such derivative plots are shown in Figs. 11b, d, and 12b. The derivative is calculated by taking the slope of the interval directly preceding a particular data point on these graphs. For example, in Fig. 11b the derivative indicated for threshold number 1 is the slope of the RMS difference vs. threshold value curve over the interval of threshold number 0 to threshold number 1. The peak in the derivative curve indicates the threshold number for which the tip reconstruction is optimized. For example, in Fig. 11b, the derivative plot exhibits a global maximum at threshold number 1.4, indicating that 1.4 is the Goldilocks threshold number, in agreement with the Goldilocks value determined using the best visual match and using the minimum of the RMS difference with respect to the actual tip shape in Section 5.1. For some graphs of the RMS difference relative to the zero threshold, the transition in slope is not as dramatic (for example, see Fig. 11c). In that case, the Goldilocks threshold is not obvious from the plot of the RMS difference, but instead can be easily identified from its derivative (Fig. 11d).

How can one be sure that one has found the Goldilocks tip reconstruction? It is possible that the transition to Goldilocks occurred for a threshold number between 1.3 and 1.4? One way to address the ambiguity for the choice of Goldilocks threshold from Fig. 11a and b would be to investigate other possible threshold numbers between the threshold numbers 1.3 and 1.4. As shown in Fig. 12, threshold number steps of 0.01 instead of 0.1 yield more precise values for the Goldilocks threshold. With this smaller interval between thresholds, the transition region from low threshold number (1.2) to higher threshold number (1.4) contains two distinct abrupt changes in RMS difference: one transition around threshold number 1.23, and another transition around threshold number 1.35. These transition points are much more clearly identified as spikes in the derivative (Fig. 12b). The reason for the double transition in this case is that the left hand side of the tip reconstruction profile makes the transition up to the Goldilocks shape first (around a threshold of 1.23) and then finally at 1.35 the right side of the tip reconstruction profile then transitions to the Goldilocks shape. For this reason, when a double transition occurs, the last abrupt transition in the RMS graph should always correspond with the Goldilocks tip reconstruction. Hence, in this case the Goldilocks threshold corresponds with the threshold number of 1.35. The minor peaks in Fig. 12b are caused by small variations in the tip reconstructions as the threshold changes. These variations are a natural consequence of the minute variations in the tip reconstructions, which occur due to noise of various frequencies in the image.

So which is the Goldilocks tip reconstruction? Fig. 9b shows profiles from P-80 with Goldilocks threshold numbers of 1.3, 1.4, and 1.5. It can be seen that little quantitative difference exists between the tip reconstructions 1.4 and 1.5. The same is true for the tip reconstruction profiles from threshold numbers of 1.35 and 1.4 (not shown). In other words, any threshold value between 1.35 and 1.5 would produce a reasonably accurate tip reconstruction. Calculating the difference between each of these profiles, we find that the RMS difference from the actual tip profile deviates by at most 6 nm between the profiles for threshold numbers 1.4 and 1.5 (see Fig. 10). This insensitivity to threshold over a range of the Goldilocks threshold provides leeway as the user attempts to find a reasonable estimate for their tip shape. Hence, once there is an unambiguous sharp peak in the derivative plot that is consistent for both the x- and y-profiles, no further refinement of the threshold step is needed.

Using the methodologies described in this section, we demonstrate a reliable approach for determining the Goldilocks tip



Fig. 12. Plots of RMS difference and its derivative between the zero threshold profile and each tip reconstruction as a function of threshold for image P-80. In this case, the threshold numbers are separated by increments of 0.01, instead of increments of 0.1 as shown in Fig. 11. (a) RMS difference relative to the zero threshold profile as a function of threshold. (b) Derivative of RMS graph (derivative of plot in (a)) as a function of threshold. The Goldilocks threshold is identified as the highest threshold value for which there is a peak in the derivative plot for both *x*- and *y*-profiles.

reconstruction, even for noisy images and for cases where the Goldilocks threshold may be initially ambiguous.

6. Conclusions

The blind tip reconstruction method is a very powerful tool to determine the shape of an AFM tip using a sample of unknown surface geometry, which could simply be the primary sample to be studied in the AFM experiment. Here, an overview of the blind tip reconstruction process was presented to make the process more intuitive. We developed an augmented MATLAB-based implementation of blind reconstruction that allows for easy variation of the key input parameters: threshold value and tip matrix size. This algorithm was used to investigate the role of these parameters, as implemented using a set of simulated images. It was found that, if there are recognizably dilated features in the image, then the tip matrix size is straightforward to determine. Specific guidelines for determining the optimum threshold number (the Goldilocks threshold number) were developed. If the tip shape is known, then the Goldilocks threshold number identifies the tip reconstruction that occurs when the RMS difference with respect to the known tip shape or the noiseless reconstruction is minimized. If, as is typically the case, the actual tip shape is not known, then it was found that the Goldilocks tip reconstruction occurred at the maximum of the derivative of the RMS difference between profiles at a given threshold and those at zero threshold.

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Appendix A. Supporting information

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