



PAPER

Automated real-space lattice extraction for atomic force microscopy images

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

Abstract

Analyzing atomically resolved images is a time-consuming process requiring solid experience and substantial human intervention. In addition, the acquired images contain a large amount of information such as crystal structure, presence and distribution of defects, and formation of domains, which need to be resolved to understand a material's surface structure. Therefore, machine learning techniques have been applied in scanning probe and electron microscopies during the last years, aiming for automatized and efficient image analysis. This work introduces a free and open source tool (AiSurf: Automated Identification of Surface Images) developed to inspect atomically resolved images via scale-invariant feature transform and clustering algorithms. AiSurf extracts primitive lattice vectors, unit cells, and structural distortions from the original image, with no pre-assumption on the lattice and minimal user intervention. The method is applied to various atomically resolved non-contact atomic force microscopy images of selected surfaces with different levels of complexity: anatase TiO₂(101), oxygen deficient rutile TiO₂(110) with and without CO adsorbates, SrTiO₃(001) with Sr vacancies and graphene with C vacancies. The code delivers excellent results and is tested against atom misclassification and artifacts, thereby facilitating the interpretation of scanning probe microscopy images.

1. Introduction

In recent years giant leaps have been made in scanning probe microscopy, particularly in atomic force microscopy (AFM) [1–3]. Atomically sharp tips, often functionalized with simple molecules (e.g. CO-terminated tips) [4–7] allow not only for the structural identification of material surfaces at the atomic level but also for precise manipulation of single atoms and molecules [8, 9].

Improved imaging techniques, aided by artificial intelligence (AI) [10–12], lead to a vast abundance of atomically resolved images. Acquisition rates have reached a point where the analysis of individual images by humans, albeit computer-assisted, becomes a new bottleneck in the advancement of related surface science fields.

Machine learning (ML) can represent a viable alternative to accelerate the processing of these data. Supervised learning approaches based on deep neural networks (DNNs) have been applied to a variety of different tasks [13], ranging from tip functionalization [14, 15] and lattice recognition [16–19], all the way up to automated molecular structure discovery [9, 20]. Despite these promising results, the applicability of DNN for efficient interpretation of experimental images is limited by the available databases [21]. While

electron microscopy [22–24] and scanning tunneling microscopy (STM) [25] image databases have been recently published, we are not aware of publicly available ones of atomically resolved experimental AFM images for surfaces (a database on simulated AFM images for molecular identification has been recently collected [26]). Augmenting an image database using both measured and simulated data is a sub-optimal option for practical purposes: (i) although AI optimization methods have been proposed [11], it is complicated to measure high-quality images and define a reliable reproducibility protocol; (ii) simulating atomically resolved images using computational surface science methods requires the precise knowledge of the structural model at the atomic scale, which might be accessible for bulk-terminated surfaces but highly complex in the case of surface reconstructions; the vast diversity of defects that can be present in a surface complicates this task even further. Last but not least, supervised ML also heavily relies on the network architecture, which forbids non-expert users to play with different architectures to achieve better accuracies. Open-source packages like pycroscopy [27] and AtomAI [28] can overcome this initial difficulty, although they remain dependent on a database. These reasons hinder the adoption of supervised ML in the field of atomically-resolved AFM microscopy.

In the absence of large databases, unsupervised ML methods constitute a convenient alternative to data-hungry supervised ML techniques and slow manual analysis of individual images. Several successful attempts based on unsupervised algorithms have been made. Atomap [29] analyzes scanning transmission electron microscopy (STEM) images by using 2D Gaussian fitting, differentiating atomic columns by their different brightness; different approaches based on clustering and principal component analysis were also proposed [30, 31]. In a recent work focused on the automated identification of local structures in atomically resolved images, Laanait *et al* [32] have shown that the well-established computer vision algorithms scale-invariant feature transform (SIFT) [33] combined with clustering algorithms (CAs) [34] are capable of recognizing and labeling atoms according to their local environment. SIFT has also been applied to other microscopy images such as confocal scanning laser microscopy ones (CSLM) [35]. An advantage of this approach is that it does not involve any pre-assumptions on the underlying periodicity of the lattice. Reciprocal-space analysis can be used to analyze periodic structures in images and, eventually, remove image artifacts. However, some prior knowledge of the system and the intervention by an expert user are required. Restricting the analysis to real space is expected to provide better stability under such features, thereby expanding the degree of analysis.

In this work, inspired by [32], we propose an open-source tool, AiSurf ('Automated identification of Surface images'), developed to inspect and classify crystalline 2D phases in atomically-resolved images via Scale Invariant Feature Transform and CAs. With no pre-assumption on the lattice symmetry and minimal human intervention, AiSurf can be applied to the analysis of regular structures, as well as for images showing complex structural correlations involving the formation of structural domains, structural and chemical defects, and recognition of adsorbates. To assess the performance and transferability of the code, we analyze a variety of surface structures characterized by different features, specifically: (i) Defect-free anatase $\text{TiO}_2(101)$; (ii) SrO-terminated $\text{SrTiO}_3(001)$ with Sr vacancies; (iii) Rutile $\text{TiO}_2(110)$ with O vacancies; (iv) CO adsorbates on rutile $\text{TiO}_2(110)$; (v) Simulated graphene with C vacancies. Additionally, to further test the capabilities of the algorithm against experimental noise, we show the results obtained for an $\text{SrTiO}_3(001)$ image affected by noise and other artifacts. All images have been acquired via non-contact AFM experiments, apart from the graphene image, which has been simulated using the probe particle model [36, 37].

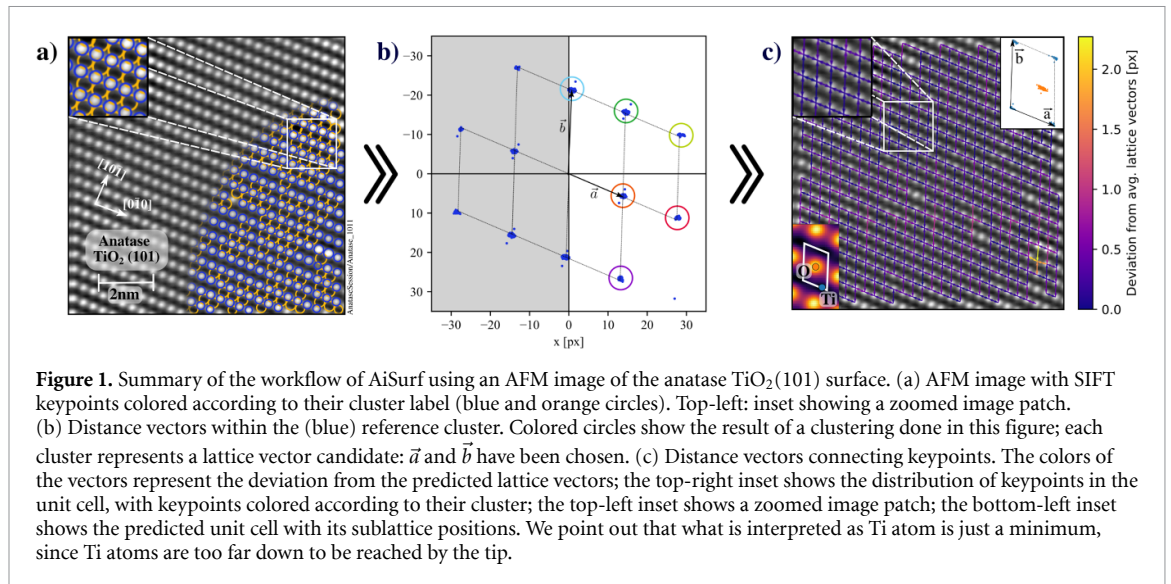
Our focus on AFM images, at variance with [32], is due to the presence of typical artifacts that do not occur in images obtained with other techniques (like STM and STEM), giving an additional performance test under not-previously-shown artifacts. Among them, we have contrast and intensity variation due to (i) insufficiently compensated drift correction, (ii) insufficiently compensated tilt correction between the image's left and right part of the image, (iii) unstable tip termination leading to an atomic rearrangement on the AFM tip during scanning [38].

The algorithm and computational protocol are presented in the next section. The experimental details on the acquisition of the non-contact AFM images and their automated analysis are presented and discussed in section 3. Data, code and documentation are available as indicated in section Data availability statement.

2. Method

This section describes the workflow of AiSurf, graphically schematized in figure 1.

First, we apply SIFT (as implemented in the library 'opencv' [39]) to an input image to extract atomic features ('keypoints') as bright and dark blobs, in analogy with the work of Laanait *et al* [32]. We specify that 'atomic features' may also refer to adsorbates on the surface or, in general, to minima or maxima present in the image. Before detecting local minima or maxima (with sub-pixel resolution), SIFT applies a



difference-of-Gaussians (DoG) filter to the image. The DoG filter is the difference of two images obtained by low-pass filtering with Gaussian kernels having two different widths. This results in a bandpass filter in the Fourier domain; rejecting high-frequency noise as well as low-frequency background variations. In the implementation used here, the DoG filter comes close to a Laplacian-of-Gaussian filter, i.e. a low-pass filtered version of the second derivative [40]. For optimal detection, the input parameters have to be adjusted (depending on the scale and contrast of the features in the image), AiSurf default values can handle the images satisfactorily. Every parameter is described in the documentation as detailed in section Data availability statement. Ideally, all visible features should be identified while no keypoints should be assigned to background noise. To further enhance the quality of keypoints, we exclude a thin area on the border of the image (by default, its width is equal to the median size of the features), and we ignore keypoints showing a significant deviation in size from the median value. This constraint should ensure that only atomically-sized features remain.

As stated in the original work [33], the keypoints detected by SIFT are characterized by a feature description based on image gradients that are brightness, scale, and rotational invariant. We break rotational invariance to distinguish keypoints belonging to different sublattices if they show the same local environment but different orientations. This can be the case of atoms of the same species but in different sublattices (e.g. graphene lattice, see figure 2), or atoms next to a point defect (e.g. vacancy) but in different orientations.

The keypoints are then clustered based on their SIFT descriptors by using agglomerative clustering taken from the scikit-learn library [41]. Agglomerative clustering categorizes features with a bottom-up approach: initially each feature belongs to its own cluster, which will be merged with the others until the specified number of clusters is reached. The merging procedure depends on the chosen linkage criterion (Ward's method [42] has been chosen in this work). This method is deterministic and only uses the number of clusters as an input parameter.

The optimal number of clusters is chosen by calculating the silhouette score/coefficient [43]. The silhouette score quantifies the goodness of a clustering process; for each feature, we call a the mean intra-cluster Euclidean distance and b the mean nearest-cluster one: for each sample the silhouette score is defined by $s = (b - a) / \max(a, b)$. What is commonly referred as 'silhouette score' (also in this work) is the average of the scores of each sample, which spans from -1 to $+1$. A value approaching $+1$ corresponds to a perfect cluster separation, while a negative value approaching -1 occurs when each feature is assigned to the wrong cluster. After finding the optimal number of clusters, each feature is labeled according to the cluster it has been assigned to.

Alternative methods, such as density-based clusterings are difficult to use here because they are typically parameterized by a characteristic distance, which is case-specific and usually non-intuitive to estimate. As an example, we show the results obtained for a defect-free AFM image of the anatase TiO₂(101) surface [44]. The experimental image overlaid with the clustered keypoints is shown in figure 1(a). The clustering labels allow us to select a reference cluster that will be used to extract the lattice vectors. It should contain only keypoints on the same sublattice since they have similar local environments and, thus, similar SIFT descriptors. We point out that AiSurf is currently not designed to distinguish different domains; for example, translational or rotational domains [45, 46] will be recognized as a single one. The reference cluster is by

default the one with most keypoints, but users can change it at their preference. In order to obtain the lattice vectors, the nearest neighbors are computed for every keypoint of the reference cluster. These nearest neighbors are grouped into sub-clusters, used to calculate the distance vectors from the reference point. The centers of mass of these sub-clusters represent possible lattice vector candidates. The two shortest, linear independent candidates are chosen as lattice vectors \vec{a} and \vec{b} . Finally, a Bravais lattice is generated by linear combinations of these lattice vectors. Figure 1(b) shows a visual representation of this process. We note that the identification of lattice vectors and Bravais lattice is based uniquely on the reference cluster, and all other clusters are not verified to hold the same vectors: this choice allows to exclude spurious effects arising from defects, i.e. non-periodic features' distributions.

After determining the lattice vectors, it is possible to calculate the sublattice positions by processing all detected keypoints again. For each keypoint we compute its position within the newly-obtained unit cell; the resulting distribution of positions within the unit cell is then clustered. The center of mass of each cluster is calculated using the Bai-Breen method [47], which accounts for the periodic boundary conditions. The resulting distribution of keypoints in the unit cell is visible in the top-right inset of figure 1(c). Up to this point, the keypoints were labeled according to the clustering performed on the descriptors, which allowed for the selection of the reference cluster for the lattice vectors identification. Now, every keypoint is labeled according to its sublattice position.

AiSurf can also plot an average view of the unit cell by averaging the area around each keypoint belonging to the same atomic species. The predicted unit cell and its sublattice positions are then drawn. The bottom left of figure 1(c) shows the image extracted with this method.

3. Results and discussion

3.1. Experimental and simulated AFM images

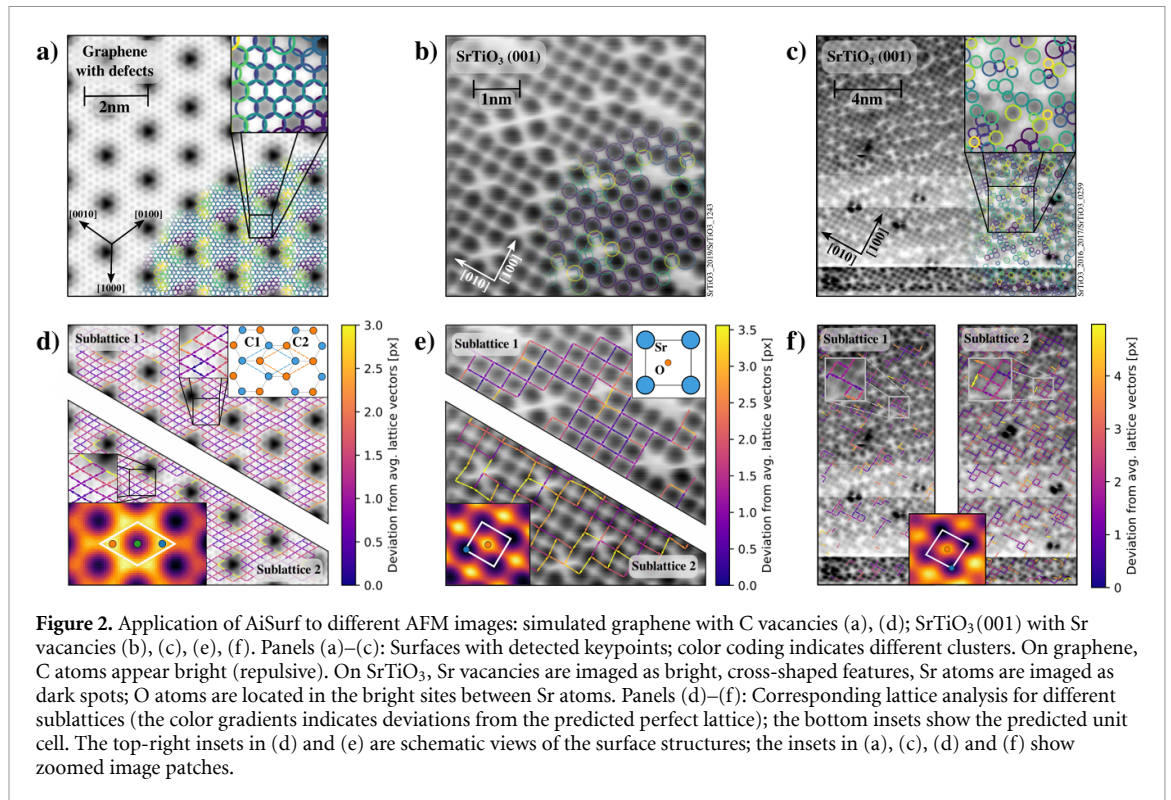
The images of anatase $\text{TiO}_2(101)$ in figure 1, the SrO termination of cleaved $\text{SrTiO}_3(001)$ in figures 2(b) and (c) and rutile $\text{TiO}_2(110)$ in figure 3 have been obtained using non-contact atomic force microscopy (nc-AFM) [2, 48] in constant-height mode and ultra-high vacuum, whereas the nc-AFM image of graphene was simulated. The signal displayed as grayscale image is the frequency shift Δf . Details on the experimental and computational setups are given below.

Figure 2(a) shows a constant-height nc-AFM image simulated using the probe particle model [36, 37] on free-standing graphene with C vacancies in a regular pattern, and three N atoms substituting C in the sites neighboring the vacancy. The simulation was performed using a CO-terminated AFM tip [8] (oriented with the O atom towards the surface) at a distance of 0.36 nm from the most-protruding atom in the surface slab, using an oscillation amplitude of 200 pm. The CO tip, with a negative $-0.05e^-$ charge on the protruding O atom, was characterized by a spring constant of 0.5 N m^{-1} . The simulation was performed over a $8 \times 8 \text{ nm}^2$ surface area, with 802×802 pixels.

Figure 2(b) shows an unreconstructed SrO-terminated region on a cleaved $\text{SrTiO}_3(001)$ surface [49, 50]. This termination consists of Sr and O atoms, organized in a (1×1) square pattern, with a characteristic concentration of 0.14 ± 0.02 monolayers of point defects in the form of Sr vacancies. The image was acquired in close vicinity to the surface using an O-terminated tip [51], such that Sr and O atoms are detected in the attractive (dark) and repulsive regime (bright), respectively; Sr vacancies are imaged as bright, cross-shaped features. The gradually decaying contrast from the bottom to the top of the image is due to the over-compensation of vertical drift caused by creep of the piezo scanner, moving the AFM sensor away from the surface. The image was obtained with no application of tip-sample bias voltage, with an oscillation amplitude of 100 pm, over a $5.4 \times 5.4 \text{ nm}^2$ surface region with 400×400 pixels. Imaging was performed in ultra-high vacuum with base pressure below 1×10^{-11} mbar, at a sample temperature of 5 K.

Figure 2(c) shows a lower-quality image of the surface displayed in figure 2(b). In addition to the intrinsic Sr vacancies imaged in the form of cross-shaped point defects, this surface also hosts protruding defects (in a concentration of less than 0.5%, displayed as dark regions) that typically appear after a surface is exposed to the residual gas in ultra-high vacuum for several days. The contrast differences in the four image regions are due to different tip-sample distances throughout the image since the nc-AFM tip was manually retracted/approached to the surface to avoid contact with the protruding defects. The image was obtained with an oscillation amplitude of 300 pm, without applying a bias voltage, over a $16.5 \times 16.5 \text{ nm}^2$ region with 400×400 pixels. Imaging was performed under the same conditions of figure 2(b).

The nc-AFM image of clean rutile $\text{TiO}_2(110)$ shown in figure 3(a) was acquired during a previous study [52]. This surface consists of rows of two-fold coordinated bridging O_{2c} atoms and five-fold coordinated Ti_{5c} atoms, running along the $[001]$ and alternating along the $[1\bar{1}0]$ direction; the rows of O atoms are occasionally interrupted by single point defects in the form of oxygen vacancies. Bright spots



indicate O atoms detected in the repulsive regime, whereas O vacancies are imaged as missing spots (the Ti atoms are too distant from the tip and are not detected by nc-AFM).

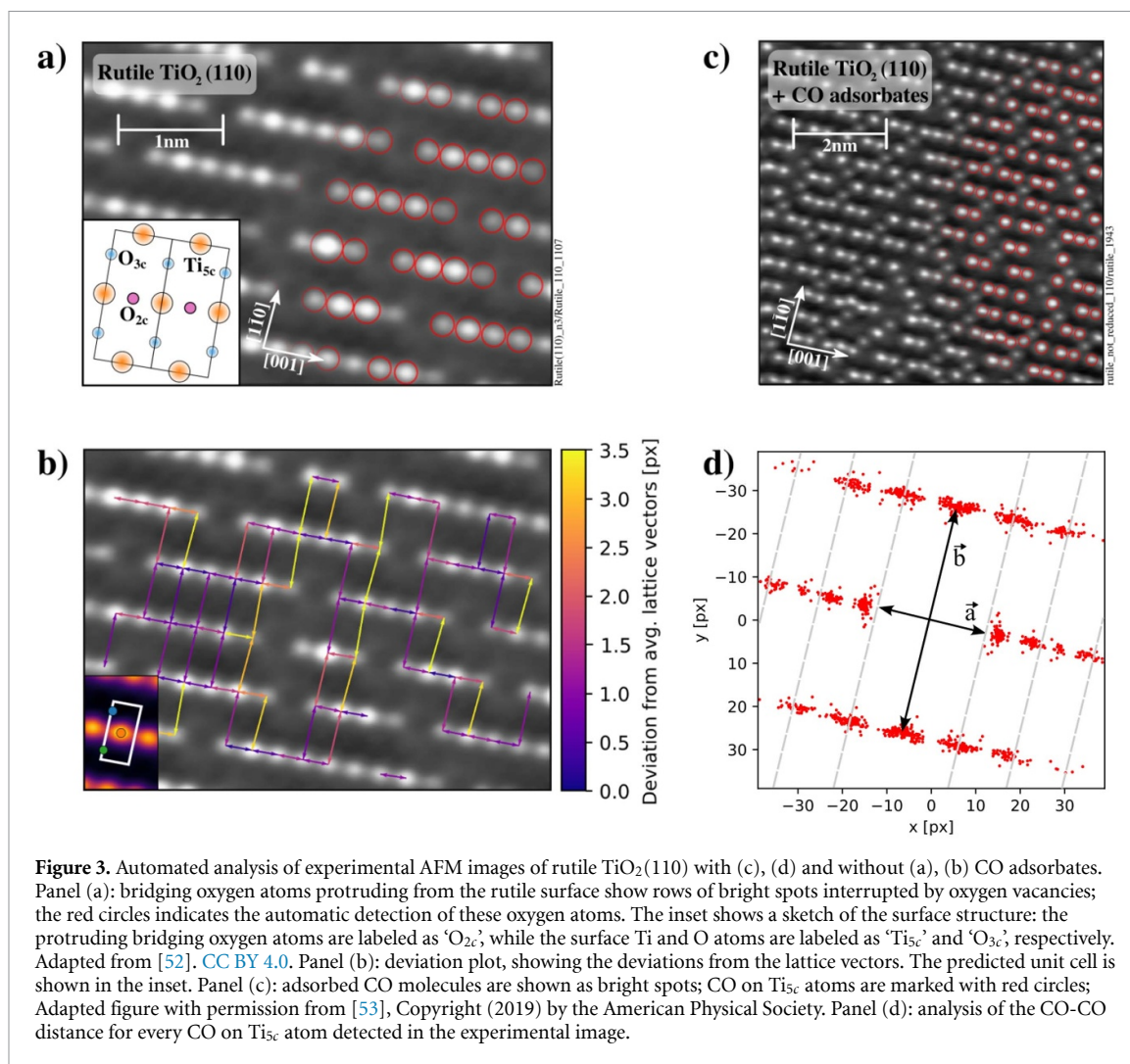
The constant-height nc-AFM image of a rutile TiO₂(110) covered by CO molecules shown in figure 3(b) was acquired during a previous study [53]. Imaging was performed with a CO-terminated tip that detects each adsorbed CO molecule as a bright spot; bridging oxygen atoms are not resolved because the adsorbed CO molecules protrude significantly more.

3.2. Automated analysis and discussion

The results obtained by AiSurf for graphene and SrTiO₃(001) are collected in figure 2. The parameters used for this analysis are included in the code repository. Three images were analyzed to test the capabilities of AiSurf under different conditions. Figure 2(a) shows a simulated image of graphene, used here to inspect the capability of our algorithm in fully controlled conditions; figure 2(b) shows a typical experimental AFM image of SrTiO₃(001) with high contrast, which represents a more realistic case; the experimental image in figure 2(c) shows multiple artifacts, and strong contrast variations; it has been used to test the capabilities of the algorithm under challenging conditions.

Figures 2(a)–(c) show the keypoints detected by AiSurf, marked with colored circles in a part of the image (each cluster with a different color). These are the keypoints left after the filtering process described in section 2. It can be noticed how the SIFT algorithm can accurately detect the centers of the features, even if they are only a few pixels in size, as the ones in figures 2(a) and (c). This is vital for the lattice recognition analysis since off-centered keypoints lead to inaccurate predictions. This first clustering process may not correctly detect all the different atomic species in the image but guarantees that the largest clusters contain features of the same type, which is needed for a successful analysis. Indeed, figures 2(a)–(c) show that atoms surrounding defects are typically assigned to different clusters for the same atomic species far from the defects; this apparent misclassification gets corrected during the sublattice recognition, where keypoints are labeled according to their sublattice position instead of the descriptor.

In all cases, the identification of lattice vectors and unit cells works with good accuracy. Since defects are neglected in the lattice vector extraction, they pose no challenge to the algorithm. However, some atoms are not detected, leading to gaps in the lattices shown in the bottom-row images. In addition, the breaking of rotational invariance leads to a separation of the carbon sublattices (see figure 2(a)) and a distinction between keypoints next to a defect (based on the direction the defect lies). The knowledge of the lattice periodicity allows us to identify any deviation from a regular pattern. Figures 2(d)–(f) show the vectors between keypoints belonging to the same sublattice, with a color coding depicting the vectorial difference from the predicted average. This distance-deviation plot helps detect displacements due to the presence of



defects and other sources of distortions. Figure 2(d) shows slight displacements only around the nitrogen impurities. The top-right inset depicts the ideal lattice and highlights the two inequivalent C atoms ('C1' and 'C2'). The bottom-left inset shows the average predicted unit cell; an orange and a blue circle represent the two carbon atoms. The algorithm has recognized the green circle at the center of the cell as a feature due to its strong contrast with the other ones, but this feature is not an atom, it rather marks the absence of an atom at the center of the ring of C atoms. All the images, except for the insets with white background, have been automatically generated by the algorithm.

Figure 2(e) shows the Sr sublattice and that of the fourfold sites between the Sr atoms (the O sites), as well as the vector deviations. As expected, deviations are present in the proximity of defects; 'Sublattice 2' shows some misclassified points but evident distortions. The square unit cell has been flawlessly extracted. Figure 2(f), used to test the algorithm's capabilities for images with more noise and experimental artifacts, shows overall positive results. Sublattice 1, defined by O lattice sites, presents several unrecognized areas, especially in the low-contrast regions. Distance deviations might not be reliable in this case. Sublattice 2 posed a lower challenge for its detection. The zoomed areas show that the algorithm can detect features even when barely visible. The unit cell has been successfully extracted.

Figure 3 shows the application of AiSurf to rutile $\text{TiO}_2(110)$ with and without CO adsorbates [53]. Figure 3(a) shows some detected bridging oxygen atoms, highlighted in red. The corresponding deviation plot is shown on figure 3(b): deviations near oxygen vacancies are visible; away from vacancy sites, no relevant deviations are present. This image is a good example of how defect-induced deviations can be easily highlighted with AiSurf. Such local structural distortions could play a role in chemical reactions between surface and adsorbates and might also facilitate the formation and identification of metastable configurations [54]. The inset in figure 3(b) shows the average unit cell. Other than the central oxygen atom (labeled as O_{2c} in figure 3(a) inset), two other features are present in the cell at the position of the three-fold coordinated oxygen atoms (O_{3c}).

Figures 3(c) and (d) show the analysis of CO molecules adsorbed on rutile $\text{TiO}_2(110)$. CO molecules adsorb both on five-fold coordinated Ti atoms and at oxygen vacancies. CO on Ti atoms are highlighted with red circles in figure 3(c). The CO-CO distances distribution on these sites is shown in panel (d). In the latter one can easily visualize details barely visible from the original AFM image: neighboring CO along \vec{a} ([001] direction) tend to repel each other by slightly tilting away from the surface normal, unlike second nearest neighbor CO or CO belonging to different rows. This can be noticed by observing the different shape of the two clusters at the center of panel (d), which are differently distributed than the others, marking an absence of short-distance neighboring CO. This result agrees with density functional theory calculations [55, 56]. Without computer tools, such details can be overlooked in content-rich images like figure 3(c).

While AiSurf is an aid for finding features in atomically resolved AFM images, determination of unit cells, and deviations from a regular arrangement, the interpretation is mostly left to the user. First, it has to be noted that the SIFT algorithm is based on detection of local minima and maxima. A minimum or maximum is not necessarily a physical feature, as exemplified by the hollow sites in the hexagonal graphene rings in figures 2(a) and (d) or the minima in figure 1(c) (blue dot in the bottom inset, no atom is detected in this position, labeling this site as a Ti atom is an interpretation). In the SrTiO_3 case, the maxima between the dark Sr sites (sublattice 1 in figures 2(e) and (f)) are at the positions of the O atoms, but this is a mere coincidence. Areas with Sr vacancies show that the O sublattice is not resolved; these areas appear with roughly constant brightness and do not show the O atoms as maxima.

When analyzing displacements, it is important to be aware of the influence of the tip on imaging. Especially tips with a rather wobbly termination such as CO-functionalized tips can easily deform due to (electrostatic) forces on the tip [36]; this can lead to apparent displacements of the surface atoms [37]. Oxygen-terminated tips tend to be stiffer, but even in the absence of tip deformation the minima or maxima in Δf images may not exactly coincide with the atomic positions. In our SrTiO_3 example, the Sr vacancies are charged defects, thus they will distort the electrostatic field in their vicinity. Since the negative frequency shift above the Sr atoms is due to electrostatic attraction between the O-terminated tip and the positive Sr atom, the field of a neighboring vacancy will influence the position of the Δf minimum. Thus, the displacements around the Sr vacancies (yellow and orange vectors in figure 2(e)) do not necessarily reflect the atomic coordinates, and they may be also related to the imaging process.

4. Summary and conclusions

In this work, various atomically resolved non-contact AFM images with different surface symmetries and degrees of complexity were analyzed in an automated way using the open source tool AiSurf. At its present state, AiSurf allows detecting the distribution of interatomic vectors in distinct sublattices and extracts primitive lattice vectors, unit cell, and deviations from ideal lattices. We have shown results obtained on simulated graphene with vacancies, as well as experimental images of anatase and rutile TiO_2 (the latter with and without CO adsorbates) and $\text{SrTiO}_3(001)$ with Sr vacancies. The distance-deviation plot proved to be useful for the detection of lattice distortions caused by oxygen vacancies, and the CO tilting (see figures 3(b) and (d) respectively). This could be applied to the analysis of any sources of lattice distortions. The algorithm has been also tested on a lower-quality $\text{SrTiO}_3(001)$ experimental image affected by artifacts. Since the underlying SIFT algorithm detects both maxima and minima, atomic features can be detected independent of whether they appear with repulsive or attractive contrast. Furthermore, the algorithm makes no assumption about the atoms showing circular symmetry; it can classify atoms consistently imaged as asymmetric features due to an asymmetric tip.

Such robustness suggests further implementations for real-time analysis, parallel to the image acquisition process. In addition, the application to atomically resolved images other than AFM ones (e.g. STM) is believed to be possible and will be further tested. Future developments are already planned, such as the identification and analysis of grain boundaries, domains and detection of defects. Another interesting feature to address is the automatic identification of atomically resolved areas, extracted from overall poorly-resolved images.

AiSurf is a user-friendly, documented unsupervised-ML tool that requires minimal user intervention and no need to provide any image database. Developing a robust tool for analyzing atomically resolved images is still an ambitious goal for both supervised and unsupervised techniques. The most adopted ones deliver successful results for limited types of images, which usually present a good atomic contrast and/or are acquired with a particular technique, such as the one for which they were designed. We aim to design a protocol that relies on little or no prior knowledge of the physical system, hoping to achieve a wide degree of flexibility. To conclude, we believe that providing intuitive and open access tools will help material scientists

accelerate time-consuming tasks like image selection and analysis, and assist the detection of elusive features in atomically-resolved images.

The AiSurf tool along with the necessary input data is publicly available on GitHub [57].

Data availability statement

The data that support the findings of this study will be openly available following an embargo at the following URL/DOI: <https://github.com/QuantumMaterialsModelling/AiSurf-Automated-Identification-of-Surface-images>. Data will be available from 2022 December 31.

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