Breaking the Limits of Scanning Tunneling Microscopy Using Image Super Resolution

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Abstract— **A Scanning Tunneling Microscope (STM) is a type of microscope that harnesses quantum tunneling to process images of surfaces at the atomic level. STMs are used to study particles at nanoscale, enabling detailed investigations at the level of individual atoms. However, the resolutions of STM images are often limited by realistic experimental conditions.**

In this paper, we investigate image super-resolution methods empowered by deep learning to break the experimental limits of STM to obtain high-resolution STM images. We first train an object detection model to detect and segment target molecules from the raw STM images. Then, an STM Super-Resolution (STM-SR) program based on Super-Resolution Generative Adversarial Networks (SRGAN) is developed and trained on a high-resolution STM dataset to learn to convert the low-resolution STM images into high-resolution ones. Our benchmark results on the test STM set show that STM-SR leads to significant resolution improvement measured by Peak Signal to Noise Ratio (PSNR), compared to the bicubic interpolation method. This super-resolution breaks the experimental barrier and reaches a higher resolution level. Finally, a Localization Scanning Tunneling Microscopy (Localization-STM) method is developed to reconstruct and further enhance the resolution of the STM image beyond the STM tip-radius limitation. As a case study, we apply our machine learning approach to generate super-resolution STM images that resemble the supramolecule, which matches its theoretical chemical structure well. The machine learning-based super-resolution approach enables STM images to reach a similar quality under less restrictive conditions and brings new insight into atomic-level physics. In particular, this method can be applied to the STM study of large, complex molecular structures, which requires stringent experimental conditions.

Keywords— Scanning Tunneling Microscope (STM), Image Super Resolution, Object Detection, Super Resolution Generational Adversarial Network (SRGAN), Localization Scanning Tunneling Microscopy (Localization-STM)

I. INTRODUCTION

Scanning Tunneling Microscope (STM) forms atom-level images by scanning the sample surface using a physical probe. STMs are key tools of nanoscience. Since their invention, STMs have been responsible for many breakthroughs in nanophysics, semiconductor science, and biochemistry [1-3].

The resolution of STM images is critical to precisely measure the physical properties of target molecular samples, including surface topography, electronic properties, strength of chemical bonds, dielectric and magnetic properties, and contact charges, as well as to study the subtle effects in physics phenomena, such as conformation change, friction, lubrication, vibration, and molecular manipulation, in great details. STMs have a lateral resolution up to 0.1nm and a depth resolution of 0.01nm. However, an STM is only able to achieve high-resolution atomic-level images under ideal experimental circumstances, at cryogenic temperature and perfect flatness. Oftentimes, due to limitations imposed by the target samples, experimental conditions cannot achieve high resolution. In particular, STMs have a difficult time in getting atomic resolution when scanning large molecules at elevated temperatures, primarily because the high thermal energy causes significant mobility and movement of the molecules. Recently, experimental methods to enhance STM resolution have been developed by functionalizing the STM tip [4, 5] with an atom or a molecule that significantly contributes to the tip-sample interaction, which can reveal the internal structure molecules adsorbed on surfaces. These experimental resolution enhancement methods have become successful in certain classes of molecules, and many astoundingly clear STM images have been generated. However, these experimental resolution enhancement methods based on functionalizing tips complicate the experiment and are not universally applicable to any molecules of interest.

The purpose of this paper is to demonstrate that novel methods, empowered by physics experiments, image processing, and machine learning technology, can computationally enhance the resolution of STM images and overcome the physical limitations of STM experiments. In this study, we model the STM image super-resolution problem as a Single Image Super Resolution problem, and we attempt to train machine learning models with many STM images on a variety of molecules obtained from high-resolution STM experiments to enhance the low-resolution STM images where the high-resolution version cannot be obtained from experiments. Our STM image super-resolution procedure includes three major components: a target molecule detection program, an STM super-resolution (STM-SR) program, and a localization-STM program. The target molecule detection program trains a Yolov5 program [10] to detect target molecules with high accuracy from a large STM scan. Using a large set of high-resolution STM images across many molecule systems available as the training set, the STM-SR program adopts a Super-Resolution Generative Adversarial Networks (SRGAN) [11] architecture to transform low-resolution STM images into high-resolution STM versions. Based on the high-resolution STM images generated by STM-SR, inspired by the microscopy localization methods [12, 13], a localization-STM program is designed to further improve the

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resolution of the STM images beyond the STM tip-radius limitations. We use the supramolecule as a case study to demonstrate the effectiveness of our STM image super-resolution methods. Our results show that a clearer view of the atomic world can be achieved with the help of modern machine learning-based image super-resolution technologies.

II. Background

A. Scanning Tunneling Microscope (STM)

An STM is a microscope capable of taking images of atomic topography [1-3]. The STM has two key components, the scanning tip and the scanning surface. STMs utilize the electron tunneling current between the surface and the tip of the scanning needle to gather data. This usage of the electron tunneling current requires the scanning surface to be extremely flat and the scanning needle to be atomically pointed. STM measures the tunneling current and generates tunneling current images by scanning an extremely sharp metal wire tip over a surface of material samples with precise, angstrom-level control, taking advantage of the piezoelectric effect [14]. When the atomically sharp tip is sufficiently close to the surface within sub-nanometer distance, the voltage bias between the tip and the scanned surface enables electrons to tunnel through the vacuum in between to form tunneling current, due to the quantum tunneling effect. As the top encounters sample features of different heights, tunneling current changes correspondingly. Monitoring the tunneling current and coordinating the current with the positioning of the tip, the sample surface is topographically imaged at the atom scale, resolving the conformations of individual atoms in the material sample.

B. STM Image Super-Resolution

The efforts to enhance STM image resolution can be classified into experiment and computation-based methods.

The rationale behind the experiment-based methods is that the resolution of the STM images crucially depends on the chemical nature of the sharp STM tip apex. Hence, functionalizing the STM tips with a molecule or an atom can trigger the interaction between the tip and the specific structure of the target sample. Intentionally picking up the functionalizing molecule or atom amplifies the detected signal, which is the key to dramatically enhancing the STM resolution. For example, functionalizing the STM tip with a single carbon monoxide (CO) molecule improves the resolution of molecular orbital STM images [6, 7, 15]. Another example is the scanning tunneling hydrogen microscopy (STHM) with STM tip functionalization with H2, D2, and a variety of other atomic and molecular particles [8, 9], which allows the STM to resolve the atomic structures of large organic adsorbates in a direct imaging experiment. Although many STM images of different classes of molecules within sub-atomic resolution have been generated recently, the functionalizing tips method increases experimental difficulties such as the deposition of molecule or atom for tip decoration on surface may change the physiochemical properties of the target samples. Therefore, the functionalizing tip has experimental limitations and cannot be generalized to any molecules and materials of interest.

On the other hand, the computation-based methods attempt to reconstruct high-resolution STM images after the acquisition of STM scans. For example, the localization microscopy methods [16] isolate and pinpoint the spatial fluctuations of topographic features in microscopy images to reconstruct high-resolution density maps. These microscopy localization methods typically require a lot of experimentally-obtained images to eliminate statistical noise, either images of many molecules of the same kind or many images of a single molecule.

C. Single Image Super Resolution

Single Image Super Resolution refers to the task of restoring a high-resolution image from a low-resolution observation of the same scene. Due to its ill-posedness nature, single image super-resolution is a well-known challenging problem. Recently, powerful deep learning algorithms, including Super-Resolution Convolutional Neural Network (SRCNN) [17], SRGAN [11], Deep Recursive Residual Network (DRRN) [18], Enhanced Deep Residual Network (EDRN) [19], Deep Back Projection Network (DBRN) [20], and many others, have been developed for Single Image Super Resolution and have achieved state-of-the-art performance in many applications. Yang et al. [21] provides an excellent survey of the available deep learning architectures for Single Image Super-Resolution.

In this work, different from the existing experiment- or computation-based methods for STM image super-resolution, inspired by the effectiveness of deep learning methods for single image super-resolution, we attempt to use the experiment-generated high-resolution STM images on a wide variety of molecules to enhance low-resolution STM images. While the purpose of this paper is not to compare different deep learning single image super-resolution methods on STM image super-resolution, we adopt a deep learning architecture similar to SRGAN to achieve STM image super-resolution.

III. Methods

A. STM Data Collection

In the physics lab, target molecule samples are prepared and the raw images are scanned on the STM platform. The STM lab software is used to generate images of the sample. The STM processes include sputtering, annealing, depositing the molecules on the surface, and tip forming; these processes are shown in the flowchart of Fig. 1.

The preparation of the sample begins with sputtering. Sputtering uses charged ions to atomically clean the substrate (in this case, a gold plate). Sputtering causes deformations in the gold substrate, thus annealing is used to smooth the surface of the substrate. Then the molecules are deposited on gold substrate. The tip then approaches the sample until it detects tunneling current. The tip is then used to scan images of the sample surfaces. During scanning, the tip can crash and deform. If this occurs, tip plunging and pulsing are used to repair the STM tip. The scanning generates images from the tunneling current between the tip and the sample. These images sometimes have low quality and low resolution; thus, we use image super-resolution methods to break the STM's experimental limit.

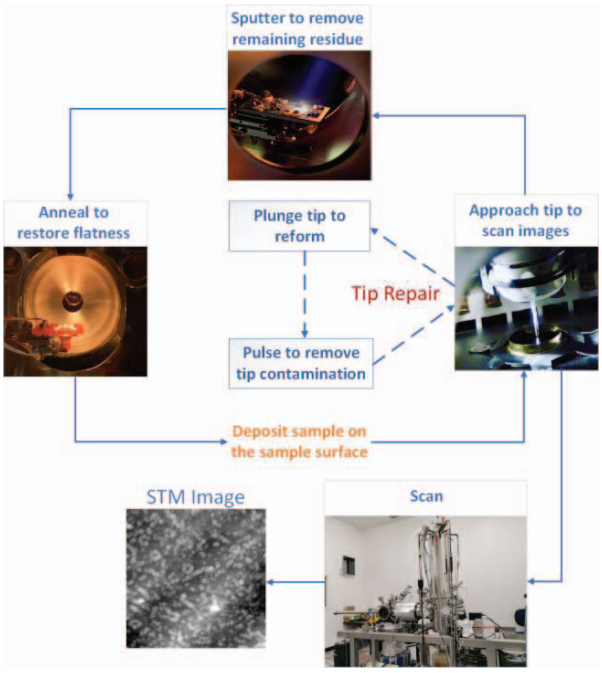

Fig. 1: Procedure to Generate Target Molecule Scan

B. *Target Molecule Identification*

In an STM experiment, a scan is carried out on a certain area of the sample surface that may contain the target molecules. Each raw STM scan contains from none to a few small target molecules (Fig. 6). Here, we develop a target molecule identification program by training a Yolov5 [10] program to automatically identify the target molecules.

C. *STM Super Resolution (STM-SR)*

The STM-SR program adopts the similar mechanism as that of SRGAN. As a Generative Adversarial Network (GAN) architecture [22], STM-SR consists a generator and a discriminator, which are illustrated in Fig. 2.

In the STM-SR generator, the low-resolution STIM image is passed as input through an initial convolutional layer of 9x9 kernels and 64 feature maps followed by a Parametric ReLU layer. The next layers utilize 16 residual blocks, each containing a convolutional layer of 3x3 kernels and 64 feature maps followed by a batch normalization layer, a Parametric ReLU activation function, another convolutional layer with batch normalization, and a final elementwise sum method by adding up the feedforward output with the skip connection output [25]. Then the upsampling block, consisting of a convolutional layer, a upsampling layer, and a leaky ReLU activation is used to resize toward the size of the target high-resolution STM image. The rest of the generator model is constructed by 2 upsampling blocks, a convolutional layer, and a Sigmoid activation function to generate the high-resolution STM image. In our implementation, the input low-resolution STM images are 64x64 monotone images and the output is 256x256 high-resolution images.

The STM-SR discriminator architecture makes use of an initial convolutional layer followed by a Leaky ReLU activation function. Then, after 7 repetitive discriminator blocks, each including a convolutional layer, a batch normalizing layer, and a Leaky ReLU activation function, a single one-dimensional vector is converted by a flatten layer. Afterwards, a series of fully-connected dense layers followed by a Sigmoid activation function are used to carry out the classification action. The discriminator helps the generator to effectively learn the features of high-resolution STM images.

Similar to the SRGAN, the overall loss of the STM-SR is the weighted sum of the content loss measured by Visual Geometry Group (VGG) [26] loss and the adversarial loss. This loss allows the STM-SR model to focus on the improvement of the quality of the STM image instead of pixel-by-pixel comparison.

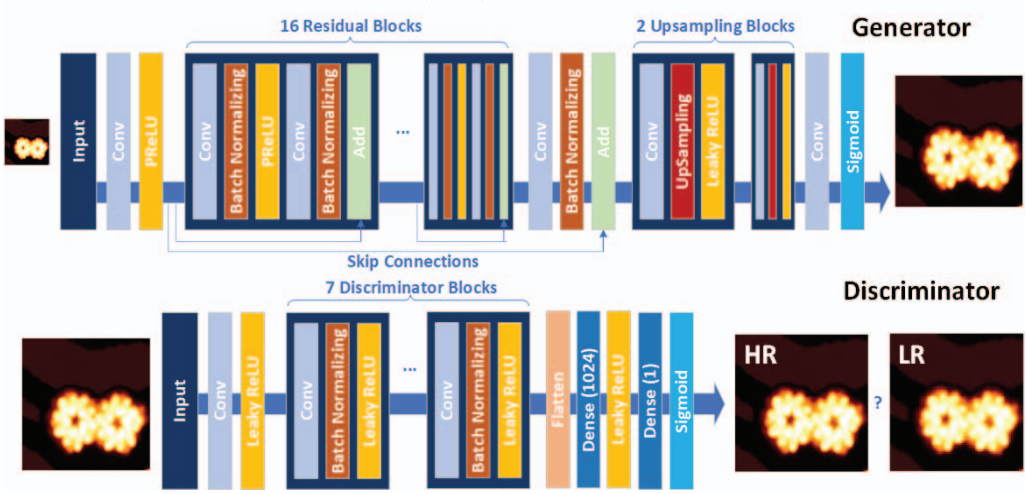

Fig. 2: Architectures for the Generator and Discriminator in STM-SR

D. *Localized Scanning Tunneling Microscopy*

Localized-STM is a new method of processing STM images inspired by [16]. STMs record the tunneling current in specific intervals; each reading is then recorded as a pixel on the STM image. Thus, we can use the nature of tunneling current to further increase resolution by using Localization-STM. The

rationale of Localization-STM is that the tunneling current $I(d)$ is inversely proportional to e^d such that

$$I(d) = k \cdot e_0 V \cdot e^{-2\frac{\sqrt{2m\Phi}}{h}d},$$

where k is the constant, e_0 is the charge of an electron, m is the mass of an electron, Φ is the work function, h is Planck's constant, and d is the tip-sample distance.

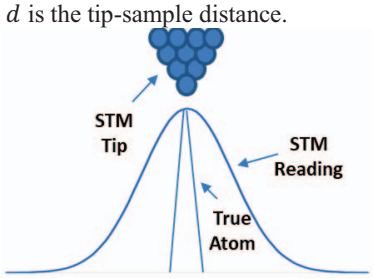

Fig. 3: STM tip over a true atom.

The tunneling current increases as tip-sample distance d decreases. When an atom is scanned by the STM, it forms a Gaussian curve. In reality the atom is much smaller than the width of the Gaussian as shown in Fig. 3. However, the atom lies under the peak of the Gaussian. So the local maxima of the Gaussian will align with the true atom's maxima. Not only does this method pinpoint where the true atom is, it also reduces the impact of experimental noise on the locations of the atoms. Fig. 4 depicts the flowchart of the Localization-STM algorithm. Fig. 4 depicts the flowchart of the Localization-STM algorithm. The first part of Localization STM is to generate 100 different super resolution images from a single STM scan. These 100 images are solutions to the super resolution problem and are each equally representative of the atomic topography below. Thus, each of these images will have slightly different local maxima. Compiling these maxima together, we get an image with the centers of each atom. Finally, we apply a Gaussian on each local maximum to account for the true size of each atom

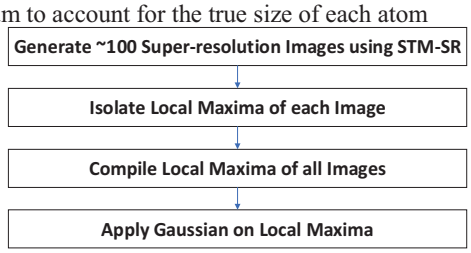

Fig. 4: Flowchart of Localization-STM

IV. Results

A. Evaluation of Target Molecule Detection

The target molecule detection program based on Yolov5 is trained on 183 raw images obtained from STM experiments, with totally 720 manually labeled target molecules. Each target molecule is labeled by drawing a bounding box around it using the image labeling and annotation tool DarkLabel [24]. This bounding box is defined by its center coordinates, height, and width. These labeled STM images are split into the training and validation set. 80% of the images are grouped into the training set, and 20% of the images are grouped into the validation set. The YoloV5 training model is run for 100 epochs. Once the model is trained, it is applied to the remaining images scanned. The molecules are identified and then automatically cropped for super resolution processing.

Target molecule identification using Yolov5 is rather effective. After training is done, a test set of 32 images with 102 instances of target molecules is used to determine the precision and recall of the identified target molecules. Precision indicates the proportion of target molecules correctly identified to the total number of identifications. Recall indicates the proportion of target molecules correctly identified to the total number of target molecules available. Our trained Target Molecule Detection program reaches recall at 0.863 and precision at 0.844 in identifying the supramolecule. Below, Fig. 5 shows the progression of precision and recall on the test set during the training process.

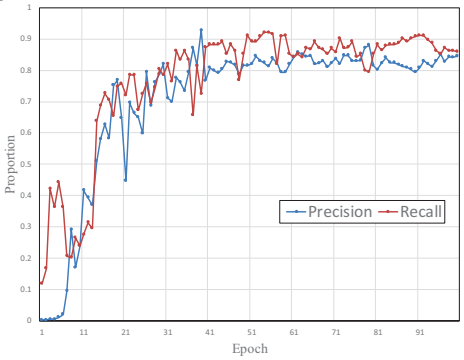

Fig. 5: Progression of Precision and Recall on Test Set duing Training of Target Molecule Detection using Yolov5

Fig. 6(a) displays a large raw STM scan of sample containing instances of supramolecule as the target molecule. Fig. 6(b) shows two instances of supramolecule identified by the trained target molecule identification using Yolov5. The cropped instances of the supramolecule are shown in Fig. 6(c).

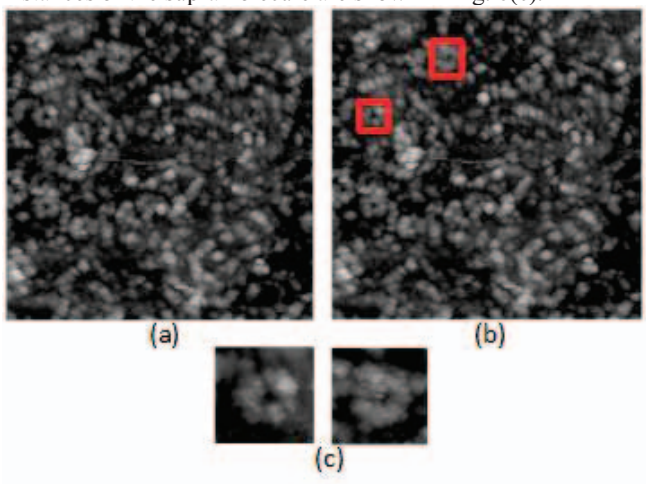

Fig 6. Detecting and Cropping Low-resolution Supramolecule Images from a large STM Scan

B. Performance of STM-SR

We collect 119 high-resolution STM images of a wide variety of molecules to train the STM SRGAN. These STM images are obtained by high-resolution STM experiments with functionalizing tips available from the public domain. All of

these high-resolution STM images are scaled to 256x256. Data augmentation methods, such as rotation, and vertical and horizontal reflections, are applied to increase the size of training samples. Additional 17 experiment-generated, high-resolution STM images, which are on molecules different from those in the training set, are collected and used as the test set to evaluate the performance of STM-SR.

Peak Signal to Noise Ratio (PSNR) is used to measure how similar the reconstructed super-resolution image O is to the actual high-resolution STM image P with sizes of $m \times n$. The mean squared error (MSE) between O and P is defined as

$$MSE = \frac{1}{mn}\sum_{i=1}^{m}\sum_{j=1}^{n}[O(i,j) - P(i,j)]^2.$$

Then, PSNR is defined as

$$PSNR = 20 \cdot \log_{10}(MAX_P) - 10 \cdot \log_{10}(MSE),$$

where MAX_P denotes the maximum signal value that exists in the original high-resolution STM image P.

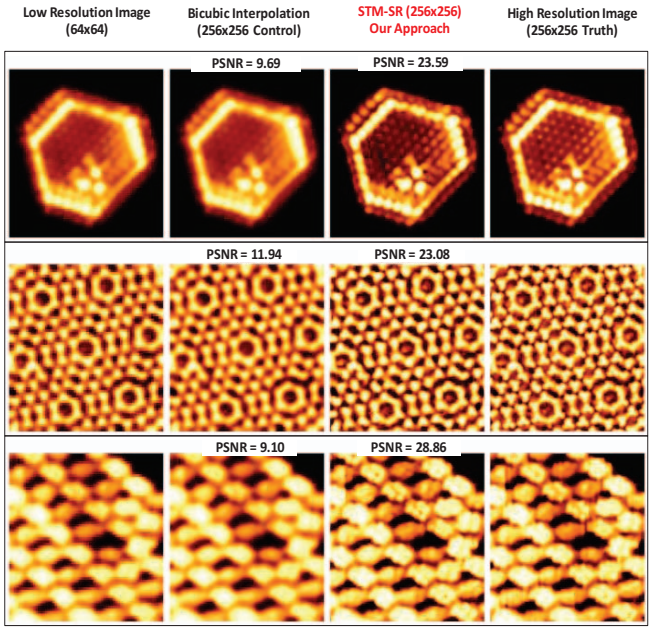

Fig. 7: Comparison of results on three STM samples in the test set

We use the bicubic interpolation method [23] as the baseline to compare with STM-SR on converting low-resolution STM images (64x64) to high-resolution ones (256x256). Fig. 7 shows the super-resolution results of bicubic and STM-SR on three STM images in the test set as examples. One can find that STM-SR yields a significantly higher PSNR than bicubic interpolation, while visually reconstructing the high-resolution images. On the overall test set, the mean PSNR of bicubic interpolation is 10.88 with standard deviation of 0.87. In comparison, the mean PSNR of STM-SR is 23.41 with standard deviation of 0.98.

C. *STM Super-resolution on the Supramolecule*

Here, we use the STM image of a supramolecule [27-29] as a case study to demonstrate the effectiveness of our STM image super-resolution method. A supramolecule is a molecule that is composed of two or more smaller subunits that are bonded together. These subunits can be the same type of molecule of different types of molecules. They can be arranged in a variety of ways to form a wide range of structures by a variety of non-covalent interactions such as hydrogen bonding, electrostatic interactions, van der Waals forces, and hydrophobic interactions. Supramolecules have a lot of important applications in materials science, biochemistry, and pharmacology. However, obtaining high-resolution topography images of a supramolecule is technically challenging in STM experiments, because supramolecules are usually deposited in liquid phase and impurities from the liquid make STM images quality compromised.

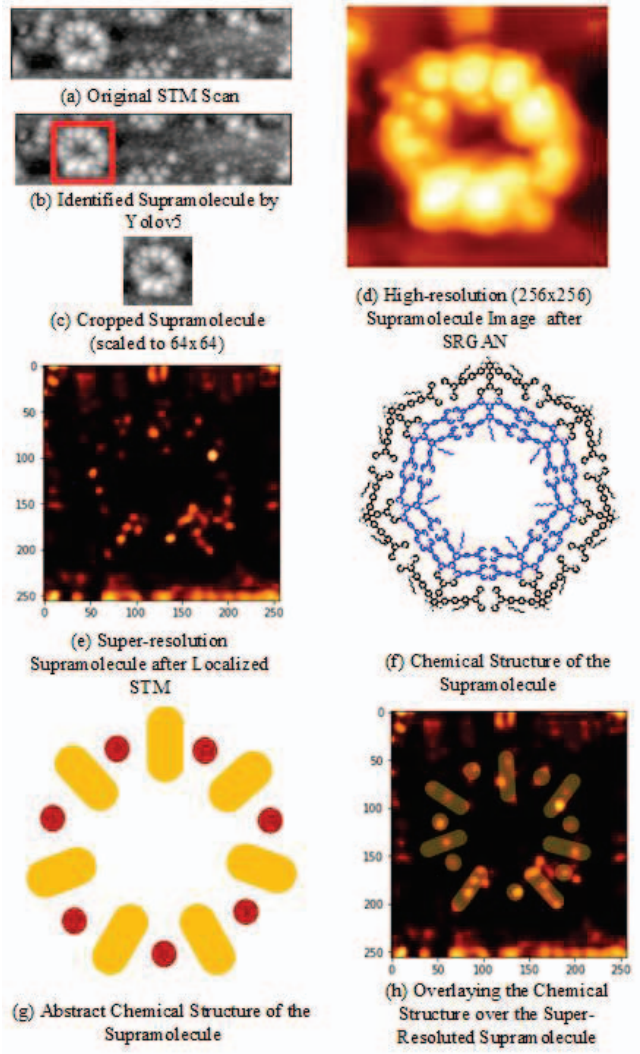

Fig. 8: Super-resolution of the Supramolecule

Fig. 8 shows the procedure of applying our super-resolution techniques to the supramolecule STM image. Fig. 8(a) is the original STM scan, which contains a supramolecule. Fig. 8(b) segments the supramolecule by our trained object detection program. The cropped supramolecule image is scaled to 64x64, as shown in Fig. 8(c). Fig. 8(d) depicts a high-resolution supramolecule image in 256x256 after STM-SR. The

localization STM algorithm requires a lot of high-resolution STM images to achieve good statistics. We generate 100 high-resolution STM images by feeding the trained STM-SR with 100 low-resolution images smeared by Gaussian noise as input. The super-resolution supramolecule after localization-STM is shown in Fig. 8(e), where each metallic atom shows as a lighted dot. Fig. 8(f) is the theoretical chemical structure of the supramolecule and Fig. 8(g) is its abstraction. We superimpose the abstract chemical structure on the super-resolution supramolecule STM image, which is shown in Fig. 8(h). One can find the super-resolution supramolecule image obtained by our STM super-resolution approach has a good agreement with the theoretical chemical structure, indicating the preliminary success of our method.

V. Conclusion

In this study, we developed and applied super-resolution techniques based on machine learning methods to break the experimental and physical resolution limits of the STM. Collecting raw images from STM experiments, we train a Yolov5 model to identify target molecules from the STM scans. This target molecule detection program can confidently identify target molecules from the large STM scans with high precision and high recall. Using the target molecule detection model, we crop the target molecules from the large STM scans. We train an STM-SR model adopting the SRGAN architecture to convert the low-resolution STM images into high-resolution ones. STM-SR generates high quality images that are superior to those generated by conventional bicubic interpolation methods. We develop a Localization-STM method to process multiple images of a supramolecule sample generated by STM-SR, isolate the true atomic signals, and retrieve a high-resolution image that conforms to its theoretical chemical structure. Localization-STM is capable of surpassing the physical resolution limits of the STM. In our study of the supramolecule, the image produced by Localization-STM denotes every single metal atom in the molecule.

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