A Physics-Informed Gaussian Mixture Neural Network to Extract Atomic Signals from Scanning Tunneling Microscope Images

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***Abstract*—Scanning Tunneling Microscopes (STMs) have led to many scientific breakthroughs in nanoscience. The STMs cannot achieve their full imaging potential due to signal overlaps and interference. This project seeks to treat the signal interference using a Gaussian Mixture Neural Network (GMNN) with physics-based constraints incorporated. This method is able to localize the individual atomic signals within an STM image, providing a quantitative analysis of nano-material properties. The GMNN is applied to a case study of Graphene Nano-Ribbon (GNR). The underlying atomic structures of GNR are successfully extracted from blurry STM images. Additionally, the extracted atomic signals of GNR from STM images provide a unique way to measure the pairwise atom distances, which enables us to gain important insight into the irregularity of experimental GNR structures. This will allow us to study the correlation between GNR structure irregularity and its superconductivity and superfluidity in the future.**

Keywords—Scanning Tunneling Microscope, Graphene Nano-Ribbon, Gaussian Mixture Neural Network

I. Introduction

Since its invention in 1981, the Scanning Tunneling Microscope (STM) is known to be one of the most powerful microscopes in existence. STMs are capable of imaging surfaces of materials at an atomic level, contributing to many scientific breakthroughs and advancements in nanophysics, semiconductor science, and biochemistry [1]. However, the STM has not reached its full potential yet. Applying advanced machine learning methods can further improve the resolution of STM images [2, 3], which allow us to gain new insights into the atomic world.

When the STM scans atomic topography, it guides a metal wire tip above the sample, recording the scanning tunneling current. The signal interference occurs due to the nature of the geometry of the scanning process and the tunneling current phenomenon. Each atomic signal take the form of a Gaussian wider than the actual atomic signal, resultng in individual signal overlaps [4]. This overlap creates a Gaussian mixture problem where the individual signals are unable to be detected.

This project seeks to enhance STM images using machine learning approach to isolate overlapping atomic signals so that we can perform precision analysis of structures, bonds, and size. In this study, target molecules are scanned and prepared as raw images by the STM. Then, a physics-informed Gaussian Mixture Neural Network (GMNN) is developed and trained to localize the individual atomic signals within the STM image. GMNN recognizes individual Gaussians, representing atomic signals, within the sample image, and isolates them. GMNN enables the sample molecule structures to be viewed in a clearer image, allowing precise measurements and observations to be made.

These methods are applied to a case study observing the Graphene Nano-Ribbons (GNR). After successfully applying GMNN, identifying the underlying hexagonal carbon structure within GNR images, we are able to measure pairwise carbon atom distances and derive their distribution, which can be used to characterize the structure irregularity in GNR samples. This novel approach will have the potential to lead to future breakthroughs and advancements to be made beyond the current capabilities of the STM.

II. Background

A. *Scanning Tunneling Microscope (STM)*

The STM is a microscope of atomic resolution with maximum lateral resolution of 0.1nm and maximum depth resolution of 0.01nm. STMs work by scanning a sharp metal wire tip over a surface of material samples to generate images [5]. Using the piezoelectric effect, the STM can achieve the angstrom level of control for scanning. When the STM tip approaches the surface at a sub-nanometer distance, the voltage bias across the tip and scanning surface allows electrons to form a tunneling current, travelling across the vacuum in between due to quantum tunneling effect [6]. The tunneling current changes as the tip encounters sample features of different heights [7]. The tunneling current is monitored and coordinated with the position of the tip resolving the conformations of individual atoms.

B. Gaussian Mixture Model (GMM)

A GMM is a probabilistic model that assumes data is generated from a mixture of multiple Gaussian distributions, each with its own mean and variance. GMM is commonly used under an unsupervised learning technique. The parameters of the GMM are typically estimated using the Expectation-Maximization algorithm, which iteratively refines the estimates of the means, variances, and mixing coefficients of the Gaussian distributions. GMM is particularly useful in situations where data sets exhibit multimodal distributions [8].

C. Graphene Nanoribbon (GNR)

GNR is a 2D allotrope of carbon exhibiting unique electrical, optical, mechanical, and quantum properties. In a GNR layer, carbon atoms are arranged in a hexagonal, honeycomb lattice, as shown in Figure 1, with armchair or zig-zag edges. GNRs have many remarkable properties and have been suggested for a wide range of applications, such as nano-filter, semi-conductor, and catalyst [9]. In practice, these properties are impacted by GNR structural irregularity [10].

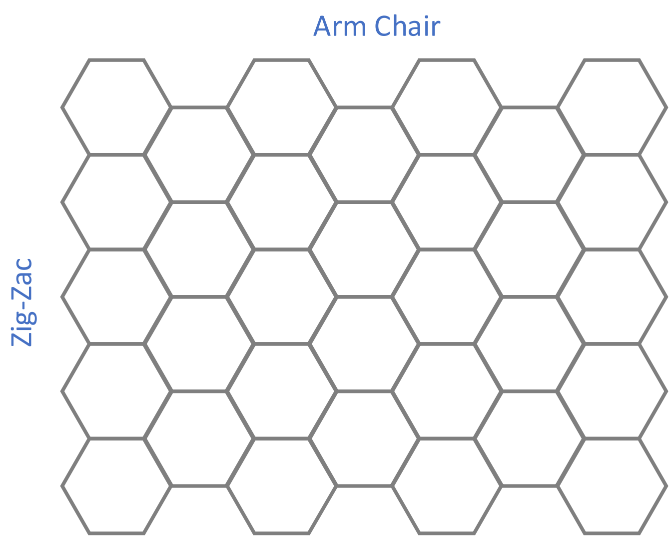

Figure 1: Structure of GNR with Hexagonal, Honeycomb Lattice

III. Methods

A. The Physics Behind

STM measures the tunneling current by scanning a sharp metal tip over a surface of material samples. When the tip is sufficiently close to the surface, the voltage bias enables electrons to tunnel through the vacuum in between the tip and the scanned surface to form tunneling current. According to equation (1), as the tip encounters the sample surface from different distances, the tunneling current changes, according to the tunneling current equation which is inversely proportional to e^d.

$$I(d) = k \cdot e_0 V \cdot e^{-2\frac{\sqrt{2m\Phi}}{h}d}, \tag{1}$$

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, and d is the tip-sample distance.

Thus, the tunneling current density increases as tip-sample distance d decreases. When an atom is scanned by the STM, a Gaussian tunneling current density is formed. In reality, the atom is much smaller than the width of the Gaussian. Therefore, when multiple atoms are present, their tunneling current densities can easily mix together, resulting in a Gaussian mixture and thus a blurry, low-resolution STM image [11]. Since the Gaussians represent the underlying atom structures, narrowing the Gaussians leads to a reveal of the precise atomic signals and therefore a clear, high-resolution STM image. Fig. 2 shows the process of an STM scan, where the mixture of individual atomic signals results in blurry STM images.

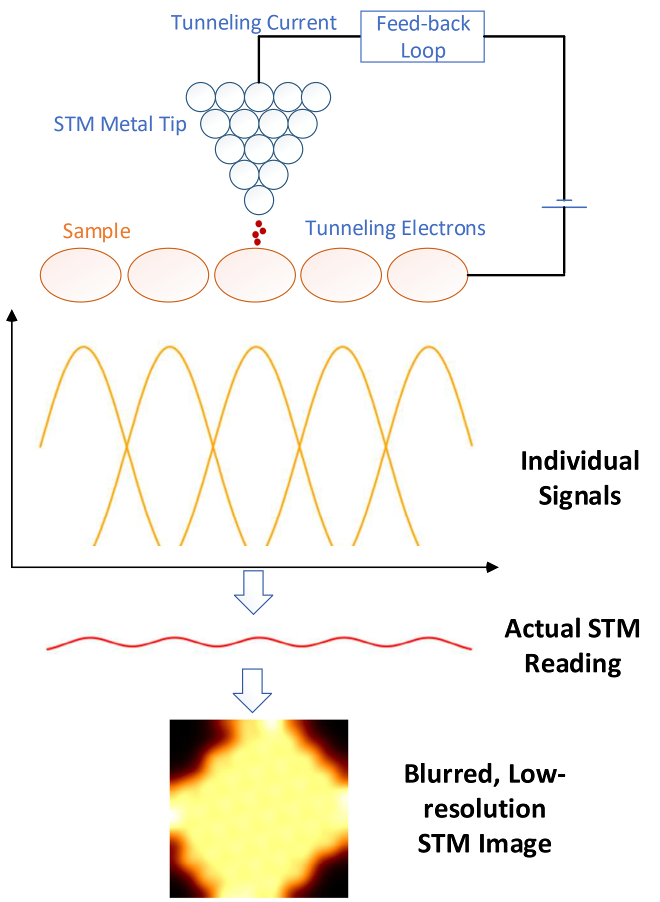

Fig. 2. STM Scanning of the Sample Surface. Mixture of the Individual Atomic Signals leads to Blurred, Low-resolution STM Images.

B. Architechture of Gaussian Mixture Nerual Network

We develop a GMNN model to identify and isolate the individual Gaussian signals from the STM image. The neural network in GMNN takes random noise as an input. It then runs through a fully connected layer with 1,000 hidden nodes followed by a retified linear unit (ReLU) activation function layer. The output layer is the parameters of 1,000 2D Gaussians. Each Gaussian is recorded as a center, two variances (x and y direction), a covariance (rotation), and a magnitude. Considering that an image is a mixture of many 2D Gaussian distributions, the predicted Gaussians by GMNN are then applied to create a generated image to approximate the target STM image.

C. *Loss Function*

This new image generated by GMNN is compared to the target STM image by measuring the Mean Squared Error (MSE) of the pairwise pixels to determine the reconstruction loss. GMNN works in a form of supervised learning and seeks to minimize the loss function. The training stops when the loss function can no longer be reduced.

D. *Physics Constraints*

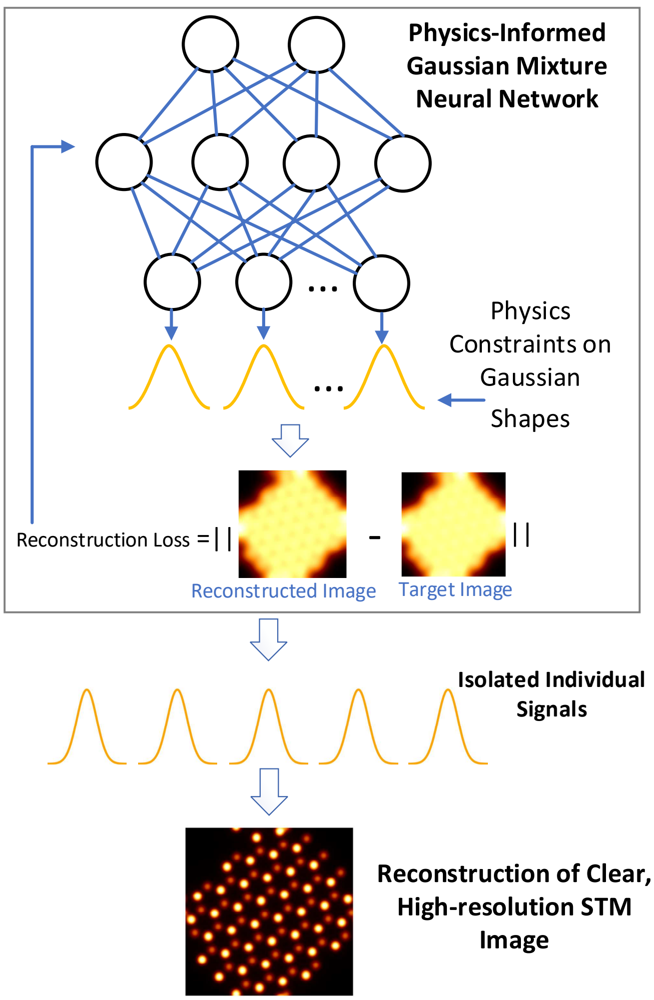

Fig. 3 Architecture of Physics-Informed GMNN and Reconstruction of Clear, High-resolution STM image from Isolated Individual Gaussian Signals

While the above GMNN model is able to generate Gaussians of any shapes, we are only interested in the Gaussian signals representing the actual atomic signals within the STM image. Therefore, we incorporate the physics constraints into the GMNN to limit the shape of the generated Gaussians to approximate the size of an atom. We also filter the generated Gaussians with small magnitudes, which typically representing the background noise. Using these physics constraints, our GMNN model becomes a physics-informed GMNN made specifically for identifying the atomic signals within the STM images.

The architecture of the physics-informed GMNN is illustrated in Fig. 3. By reducing the variances of the extracted individual Gaussian signals, a clear, high-resolution STM image can be reconstructed.

IV. Results

A. *Case Study on GNR*

To validate the effectiveness of GMNN on STM images, a case study is performed on STM images for GNR. GNR has a well-known chemical structure, which is ideal for verify the correctness of GMNN results.

We select two GNR images scanned by the STM at Old Dominion University (ODU) Atom Manipulation Lab, where one is at the resolution of 1nm and the other at 3nm. These two images are rather blurry due to the overlaps of the underlying atom signals. Fig. 4 shows the results of applying GMNN to these two GNR images. By narrowing the variances of the extracted Gaussians, we can show each individual signal separated from one another. One can find that the reconstructed STM images from the Gaussian signals extracted by GMNN exhibit much clearer structures, where the location of each honeycomb composed of six carbon atoms can be clearly identified in a hexagonal geometry. It is observed that we can reconstruct the GNR chemical structure precisely, which resemble the theoretical GNR structure illustrated in Fig. 1. The accurate localization of atomic signals by GMNN allows us to measure the important nano-properties of the experimental GNR samples that cannot be done before.

B. *Measurement of GNR Irregularity*

By localizing the atomic signals precisely, we can measure the distances between the centers of each individual signal, representing a honeycomb structure composed of six carbon atoms. Fig. 5 shows the distributions of the pairwise honeycomb distances on the two STM images. One can find that there exist irregularities within these two experimental GNR samples. These irregularities can be measured by the standard deviations of the first peak of the pairwise honeycomb distance distribution, representing the distance between the nearest honeycombs. The means of the nearest honeycomb are at 4.26 Angstrom. The standard deviation of the first peak is 0.439 in the first GNR image and 0.366 in the second one, indicating that the first GNR sample is more irregular than the second one. These irregularity measurements can help provide crucial insight into the physical properties of GNR samples, such as superconductivity and superfluidity.

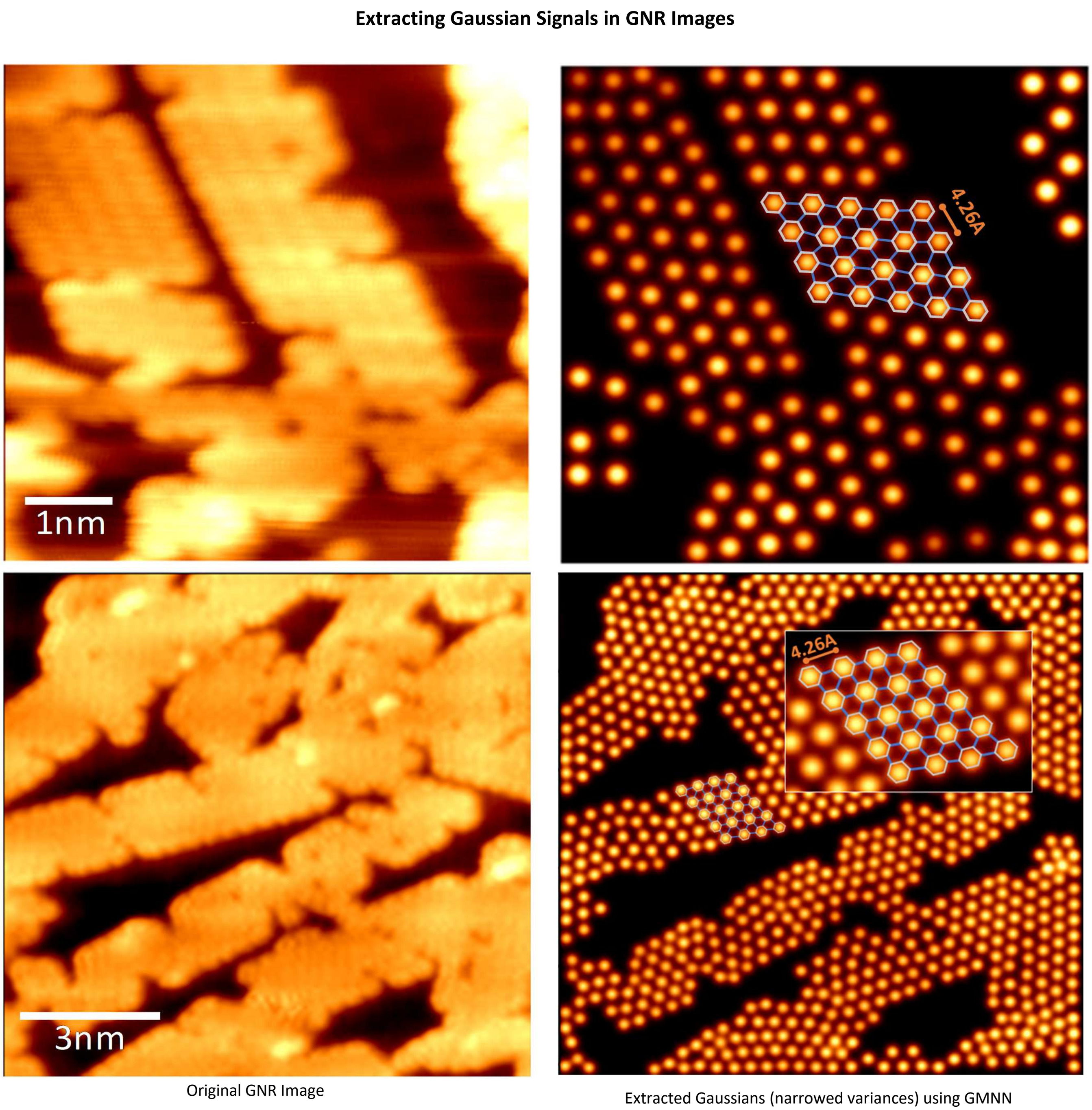

Fig. 4 Images in the left column are the original STM images of the GNR samples. Images in the right column are the results from GMNN. Clearer structures are found in the GMNN generated GNR images, matching the theoretical hexagonal GNR structures.

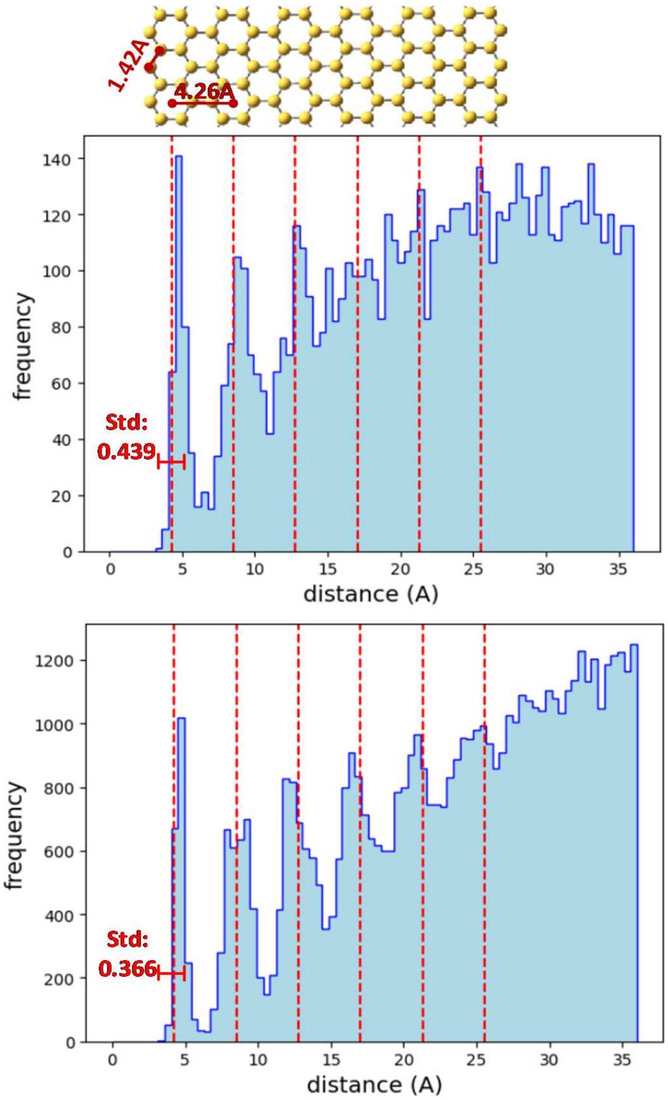

Fig. 5. Histogram of pairwise honeycomb distances. The standard deviation (std) of the first peak indicates the irregularity of the GNR sample.

V. Discussion

Considering an STM image representing a 2D probability distribution function, the Gaussian mixture model (GMM) [12], a classical soft clustering algorithm, can also be used to determine the Gaussians underlying the STM image. In order to do so, GMM has to specify a fixed number of Gaussians. Also, GMM is difficult to restrict the shape of the generated Gaussians. In comparison, GMNN has the advantage of incorporating physics constraints into the machine learning model to focus on extracting Gaussians with respect to the underlying atomic signals. Moreover, GMNN can adaptively determine the number of Gaussians by filtering those with small magnitudes, without the need of specifying the number of Gaussians beforehand.

VI. Conclusion

In this study, we develop a physics-informed machine learning model, GMNN, to isolate the underlying atomic signals within STM images. The method has been successfully validated on a case study of GNR where the localized signals match the theoretical chemical structure. The localized signals not only provide us clearer STM images, but also allow us to measure the nano properties such as irregularity, which cannot be done before.

VII. Future Work

The next steps of this work are to give further analysis of the STM images, thanks to the new nano property measurements enabled by GMNN. The association between nano-properties and macro-properties of the nanomaterials can be quantitatively studied. These will help us further understand how to design materials with desired properties in everyday use.

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