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Flexibility and utility of pre-processing methods in converting STXM setups for ptychography

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Ptychography is an advanced diffraction based imaging technique that can achieve resolution of 5 nm and below. It is done by scanning a sample through a beam of focused x-rays using discrete yet overlapping scan steps. Scattering data is collected on a CCD camera, and the phase of the scattered light is reconstructed with sophisticated iterative algorithms. Because the experimental setup is similar, ptychography setups can be created by retrofitting existing STXM beam lines with new hardware. The other challenge comes in the reconstruction of the collected scattering images. Scattering data must be adjusted and packaged with experimental parameters to calibrate the reconstruction software. The necessary pre-processing of data prior to reconstruction is unique to each beamline setup, and even the optical alignments used on that particular day. Pre-processing software must be developed to be flexible and efficient in order to allow experimenters appropriate control and freedom in the analysis of their hard-won data. This paper will describe the implementation of pre-processing software which successfully connects data collection steps to reconstruction steps, letting the user accomplish accurate and reliable ptychography.

INTRODUCTION

Ptychography and STXM

Nanomaterials have become ubiquitous in modern technology. Their properties are much different than the same materials in bulk, and is important to characterize these new forms. We must employ novel approaches to study these properties and the increasingly important processes they participate in. One of the most advanced and exciting of these characterization techniques is x-ray ptychography, a diffraction based imaging method. From the Greek root *ptycho-*, meaning "to fold," ptychography essentially works by folding lots of information about a sample into package of data. This package is later unfolded with software methods into a beautiful, high resolution image of a very small thing.

Ptychography experiments generally require several steps [Figure 1]. First, a sample is placed on a moving stage in the path of a coherent x-ray beam. The sample is moved through the beam in a raster scan pattern with overlapping scan steps. At each scan position, an image is collected of the light scattered into the far field by the sample. This stack of raw data is saved with associated metadata. The raw data is pre-processed to correct for some experimental imperfections. Lastly, the stack of processed data is passed to the reconstruction software, where each scan is examined in context of its neighbors for reconstruction. This occurs through an iterative process that involves moving between real space (image) and reciprocal space (far field scattering pattern) using Fourier and Inverse Fourier transforms. Neighboring images with overlap are considered concurrently.[1]. This examination of the scattering patterns of overlapping neighbors allows the real image to converge from the scattering data.

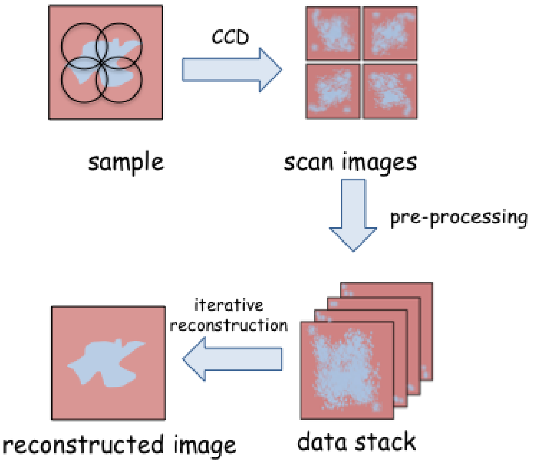

FIG. 1. This flowchart describes the steps involved to get from just light off of a sample to a reconstructed image. To begin with, the sample is scanned in discrete yet overlapping steps and a CCD (charge coupled device) camera collects an image at each point. These images are processed into a manageable data set. This data set is then used by the reconstruction software, which uses iterative algorithms and the redundancy of the overlapping scans to determine the image of the sample

This technique is attractive in part because it can be run similarly to the more conventional technique of scanning transmission x-ray microscopy (STXM). Converting a STXM setup for ptychography requires a few important changes. In STXM, the transmitted light is collected on a diode at close range which only measures the intensity of x-rays that comes through the sample, not the far field scattering pattern [2]. The typical current resolution achievable in STXM is 25-30 nm, which is the

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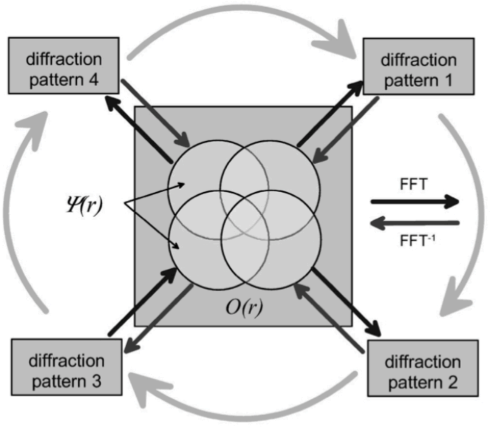

FIG. 2. Schematic describing the process of iterative reconstruction. There are four scattering scans shown here that each overlap. From each diffraction pattern (which is the reciprocal space representation of the object) alone, many different shapes of sample are possible. As the redundant bits in the overlaps are calculated, the possibilities of the sample shape in real space start to converge to a single one. These redundancies are found through many cycles of Fourier and Inverse Fourier transform. Figure taken from J. M. Rodenburg 2007 [3]

smallest spot size on the sample of incident x-ray beam that can be achieved with current optics. Because the only information collected is transmitted intensity, there can be no attempt to access information about scattering. Ptychography uses different information about the sample. Instead of measuring transmitted light, it measures scattered light in the far field. This measurement alone is not quite enough reconstruct the image, because the detector still only measures the intensity of scattered light. The phase remains hidden. However, using iterative methods, the phase can be extracted from the overlapping scan data and allow convergence on the real space image. Software designed for ptychography uses these iterative methods to take advantage of redundancy in the overlapping data and re-claim the lost phase information. With these advancements in reconstruction, ptychography has become a viable way to image samples at a resolution of 5 nm and below. However, because of the unique character of each x-ray source, these methods must be carefully tailored to each experimental setup in order to obtain useful data. This paper will outline the steps taken to implement such methods at the Stanford Synchrotron Radiation Lightsource (SSRL).

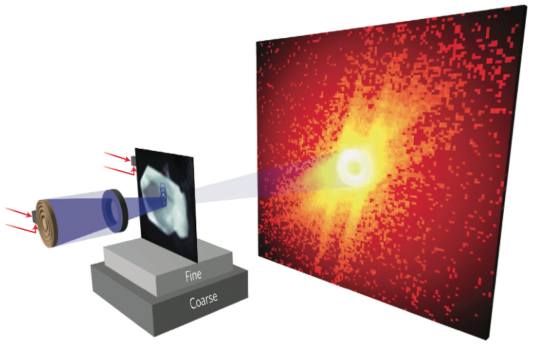

FIG. 3. This figure shows the setup used for ptychography. There is a zone plate at the beginning which focuses the x-ray beam down to a spot, and the sample is represented as a vertical plane on the stage, which has fine and coarse motion capability. Shown on the sample plane are small circles which represent the overlapping scan spots. After the light passes through the sample it is collected on a CCD, with a ring of unscattered light on the image representing the probe, or incident x-ray beam. Figure taken from D. Shapiro 2014 [4]

INSTRUMENTATION AND RECONSTRUCTION

Like all x-ray imaging techniques, ptychography requires a good source of x-rays. All measurements were made with a synchrotron x-ray source, specifically Beamline 13-1 in SSRL located at SLAC National Accelerator Laboratory. This beamline accesses radiation in the soft x-ray part of the spectrum. It is used for materials science and condensed matter physics applications, and has an energy range of 500 eV - 1200 eV. The current experimental setup is being converted from a traditional STXM configuration into one that can more fluently handle ptychographic experiments. This involves the addition of an improved interferometer controlled piezo stage for the sample which allows more precise control of the sample position. This is an important requirement for ptychography, because the precision in the motor must be at least below the resolution in the image being reconstructed. Otherwise there is too much uncertainty to be able to converge on an image in reconstruction. Other optical improvements will include the addition of a shutter to prevent smearing as well as adjustment of the alignment to ensure that the sample is completely illuminated with a coherent beam of x-rays. Lastly, and perhaps most importantly, will be the addition of a CCD camera in the far field of the sample stage to enable collection of diffraction data instead of just transmitted intensity [Figure 3].

The software used to reconstruct the images is called Sharp Camera and was developed for ptychography users at the Lawrence Berkeley National Laboratory's Advanced Light Source (ALS). It is run with a CXI file containing image data and associated metadata. Sharp Camera is a very useful software that solves the non-

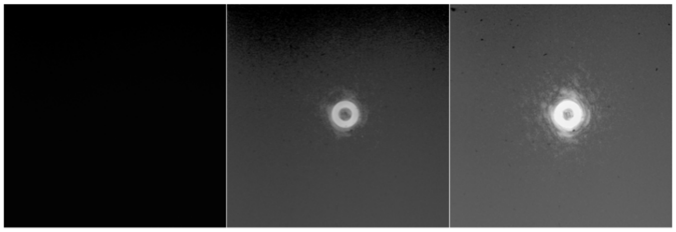

FIG. 4. This figure shows three raw data files, represented on the log scale to better show the dim scatter data. On the left is the ambient detection of the CCD, which gets subtracted out. The middle image was exposed for a short time (15 ms) and is underexposed. The image on the right was exposed to the x-ray beam for a longer time (150 ms) and is overexposed. The rightmost two images are averaged together to get a final data file with a higher dynamic range

linear loss of phase problem with the use of a innovative iterative algorithm called RAAR (Relaxed Average Alternating Reflections) [1]. This algorithm is able to use the overlaps in the scan to determine what parts of the scattering pattern are shared between neighboring scans [Figure 2]. By convolving the images and iterating back and forth through real and reciprocal space, the shared scattering patterns emerge. A shared scattering pattern in a region allows one to back calculate the shape of the scatterer, and it is the combined shapes of all these scatterers that form the reconstructed image.

In many discussions of ptychography, the focus is either experimental setup or reconstruction algorithms. The missing piece that we have developed is a system of pre-processing to link these two ends of the pipeline together. All pre-processing scripts are tested on existing ptychographic data collected at ALS on beam line 11.0.2. The sample is a $\mathrm{LiFePO_4}$ battery nanoparticles at a pressure of 10^{-3} mbar. Imaging was done to determine the mechanism of lithiation and delithiation, a process that defines the charge and discharge abilities of the battery. Though the necessary pre-processing methods for SSRL are different, both facilities use a comparable experimental setup Because the camera is consistent, the pre-processing scripts to create the CXI written for this test data should be applicable to future data with only the modification of experimental parameters. This pre-processing script is written in python.

DEVELOPMENT OF PRE PROCESSING SOFTWARE

Before the raw image data becomes a reconstructed ptychography image, it must be adjusted and packaged correctly to allow for accurate reconstruction. The steps in this process should be modular in order to give the user flexibility and choice in their analysis methods. Modular steps implemented here include cropping, centering, downsampling, exposure averaging, dark subtraction, probe reconstruction, and calculation of the STXM image. All of these corrections are done in ptyhon on raw data and then everything is written together in a specific file type, the CXI file.

The creation of a CXI (coherent x-ray imaging) file is the crucial jumping off point to begin reconstruction. It is the package that enfolds all the information collected in the experiment. The processed image data, all the associated experimental parameters, and different calculated images like the STXM image and the guess at the probe go into a CXI file after pre-processing is finished. Developed by Filipe Maia at Uppsala University [5], this file contains all the relevant information to calibrate the Sharp Camera software and get an accurate reconstruction of the sample. This file type is essentially a nested list, with python objects stored at different levels

Geometric Transformations

The data collected in each experiment comes in the form of a large stack of images. Within this stack there are three separate light conditions on the sample; dark frames, underexposed and overexposed[Figure 4]. Overexposed images have a lot of saturated pixels, but also show faint scattered light at the edge. Underexposed images have cannot distinguish signal from noise at the edges of the detector because the signal is so faint, but can how the brighter parts of the image without hitting saturation. Averaging different exposure conditions optimize the final image by giving it a higher dynamic range.The image data are averaged using the method discussed by David Shapiro in 2014 [4], where s is a threshold value for saturated pixels [Equation 1].

$$I_{merged} = \frac{I_{short} + s(I_{long})}{t_{short} + s(t_{long})} \tag{1}$$

The dark frames are a result of the ambient detection of CCD cameras. There is always a spurious signal even when the detector is in total darkness. For this reason, dark frames are collected at each exposure time and averaged. These average frames are subtracted out to reduce noise in the data.

To do calculations on this group of images, it must be represented as a three dimensional numpy array. Dimensions correspond to the number of scans taken, and the number of pixels per scan in both the x and y directions. Each single image generally looks like a dark field with low intensity scattering, and a bright central annulus which we refer to as the "probe." This annulus is the detection of the un-scattered x-ray light, and represents the footprint of the incident beam. To get good reconstruction, the center of this annulus must be in the center of the image. This is done by using the numpy.roll function to preserve avoid a sharp edge. The images can also cropped and zero-padded to reduce sharpness of the edge. This edge blurring is very useful when using Fourier methods later in the analysis. Lastly the files are downsampled using the rectangular bivariate spline method over a mesh. By downsampling to a square with image dimensions as some power of two, we reduce noise that confuses the reconstruction in addition to making the processing much faster and more memory efficient.

Probe Reconstruction

One crucial process is the reconstruction of the probe, which is the high-intensity annulus in the center of the image and represents the un-scattered beam. This is an unavoidable part of data collection, but contributes to a lot of less-useful high intensity pixels which can muddy the reconstruction. The probe must be reconstructed iteratively just like the scattering data from the sample because its shape is different with each data collection. This reconstruction is begun in the pre-processing. Since the probe is the highest intensity part of the images, a Fourier transform of the image is done in pre-processing to represent the best guess for the probe in the un-reconstructed data [Figure 5]. This is stored in the file separately to be iteratively reconstructed and subtracted in the later part of the processing. Though not strictly necessary, this step goes a long way to avoid certain pathologies that can come from scanning the sample in a grid pattern. Grid scanning helps to simplify the processing because it allows for a pretty simple matrix representing the physical translation of the sample, but there can be information lost by not using a continuous scan[6]. Robustness of the algorithm improves when the probe is reconstructed, and the data converges more quickly to a single image. [4]

STXM Image

Because of the similarities in the experimental setup, ptychographic data can also be used to calculate the STXM image for the same sample. This is done by summing the entire CCD image collected at a certain point. Because the CCD collects the scattered light at far field, the sum of that image is equivalent to the total transmitted intensity collected in the near field. Using the same raster scan pattern, we use the summed image as the intensity at that position. The STXM image is simply built pixel by pixel from the ordered scans that have already been collected. This data is very useful in debugging ptychography pre-processing software. It does not require any metadata, and the scatter patterns are each collapsed into a single value on the image. Calculating the STXM image also provides a good reality check to use in comparison with the often opaque output from Sharp Camera. Figure 6 shows the differences between ptychography and STXM images quite well. The STXM has all the same features but is much blurrier and doesn't resolve image objects well.

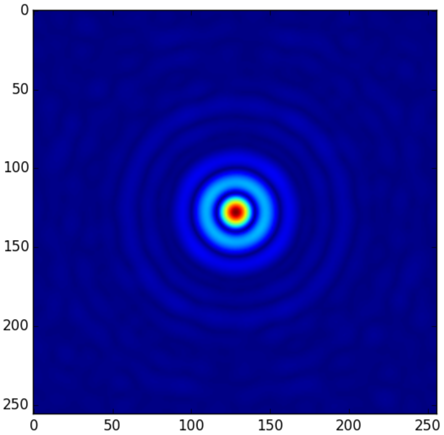

FIG. 5. This is the image of the probe estimate, represented on a scale where blue equals low intensity and red equal high intensity. The estimate works well as a first guess for iteration because it is a high intensity annulus in the center, and minimal operations were done to achieve such a thing

FUTURE DIRECTIONS

With this imaging method, the achievable resolution is 5 nm and smaller. The theoretical limit with a strongly scattering sample is only constrained by the wavelength and the numerical aperture of the detector. With weaker scatterers, the limit is defined by the scattering vector of the sample. None of these can be overcome by hardware configuration or pre-processing software, but through optimizing these two things we can get very close. In writing the pre-processing software, special consideration has been taken to make things flexible and intuitive to the non-expert python user. Since the target audience of this code is scientists who want to use ptychography in their own experiments, it is essential to limit the setup and run time to a length manageable during the run of an experi-

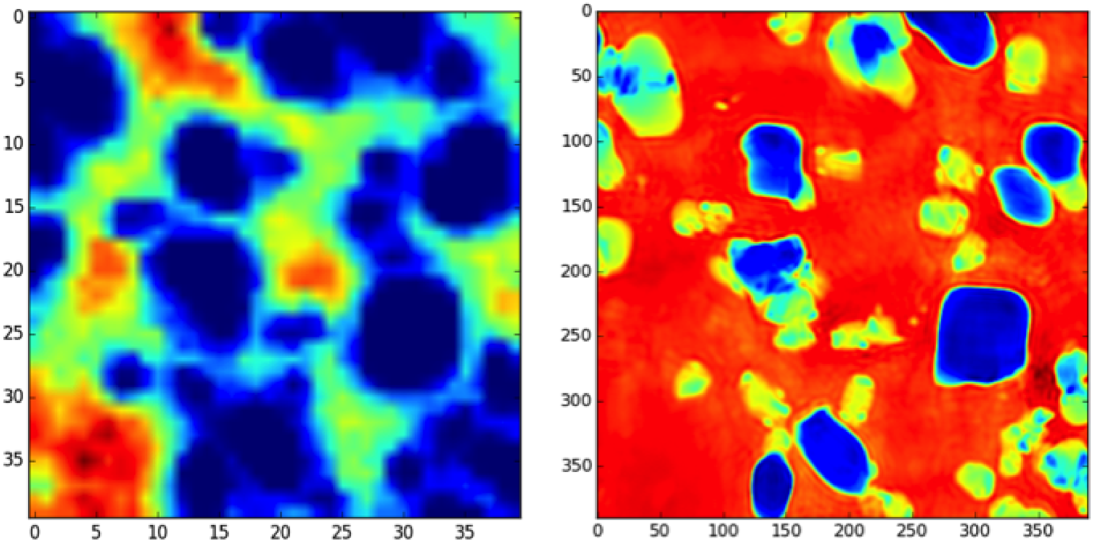

FIG. 6. These two images are of the same region, imaged using both STXM and Ptychography. The image on the left, created with STXM, is not as resolved and there are instances when to distinct objects are indistinguishable. The image on the right, taken using ptychography, is much better resolved at 5 nm and clearly shows the objects as distinct.

ment. As development continues, we hope to build a GUI and more interactive features to allow the user more freedom. As software improves, users will be able to easily test the results of different processing features and choose the best for their specific experimental setup on beam line 13-1. This is possible with the current setup, but the process could be more streamlined and memory efficient. The next steps will be to include infrastructure that can access Graphical Processing Units, as well as splitting the data into smaller chunks and processing in parallel on multiple cores. As the method progresses, it will be important to give the user freedom to experiment with different pre-processing steps in order to achieve the most accurate and efficient reconstruction. Ptychography as a technique can also be extended to richer imaging regimes, such as three dimensional imaging, time resolved imaging, and absorption edge imaging. With the addition of a third dimension of motion on the sample, rotation, a three dimensional picture can be resolved. Ptychography can also be done at different energies to acquire pictures of the same sample across the absorption edges of different elements. When merged, these different energy images can resolve chemical structure very finely. Finally, time dependent processes can be visualized when the area is scanned is fast enough. This allows for very interesting research on chemical processes like battery charge and discharge.

CONCLUSIONS

Ptychography as an imaging technique allows for excellent resolution of some of the most important molecules in current technology. However, the experimental setup must be slightly more complex to accomplish this, both in the hardware and in the software. The resolution of the reconstructed image is limited by how much of the scatter pattern can be detected. To aid in this hardware and software are both being optimized to allow for collection of reconstructable data. To link these ends of the experimental pipeline, pre-processing steps were successfully implemented. The new pre-processing software is flexible and allows the user to perform a number of important image calculations to optimize the robustness of the reconstruction algorithm further down the pipeline. The pre-processing software also packages the data along with the experimental parameters necessary to calibrate the reconstruction software. Pre-processing methods both facilitate and optimize reconstruction, and are a critical part of the process of reconstruction. In reconstruction, processed images are compared with their overlapping neighbors and redundancies are identified. These redundancies allow the software to recover the phase of the scattered light and thus converge on the shape of the scattering object. This shape is exactly what we have been seeking: a high resolution image of a very small thing. Ptychography will allow for the study of some of the most important technological developments of our time, and it all comes together in pre-processing.

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