Machine learning for denoising of scattering data and automated crystal alignment process

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Scattering experiments utilizing X-rays and neutrons are widely applied in the field of condensed matter physics to study the band structure of various quantum materials. One of the limitations in these experiments is the scarce beam time, which often results in a difficult management of on-going experiments in terms of what parameters to further investigate such that high-quality results can be achieved. One of the anticipated projects for this proposal aims at providing a machine learning assisted toolbox for the denoising of low-count statistics X-ray data, which would help as a quick and reliable guideline during measurement cycles. A second project is related to the small samples that are frequently only available in the form of single crystals, which need to be exactly aligned in a manual, labor-intensive fashion to yield precise results. Especially in neutron diffraction experiments, a large crystal mass is crucial for the execution of more complex experiments. We will target this problem by implementing a semi-automated single crystal alignment procedure in combination with specially designed and 3D-printed sample plates that allow for a more efficient crystal mounting procedure.

1 Introduction

1.1 X-ray and neutron scattering

The effect of charge ordering in correlated electron systems is a long-studied subject in the field of condensed matter physics. One example of such systems are high-temperature cuprate superconductors such as yttrium barium copper oxide (YBCO) and lanthanum strontium copper oxide (LSCO). Recently, it was shown that the application of uniaxial pressure has potential in the progress of understanding the charge ordering in such cuprates. In YBCO, charge ordering has been studied in various X-ray diffraction experiments upon applying uniaxial pressure [1] whereas in LSCO it was shown that stripe order domains can be disentangled leading to an enhanced charge order signal [2]. On the other hand, the field of neutron scattering is often used in synergy with X-ray diffraction for studying crystal structures of various quantum materials [3]. Neutron scattering experiments are unambiguous and their interpretation is straight forward. This is mainly due to the weak interaction between neutrons and matter, which implies that scattering cross-sections are described by perturbation approaches. With the construction of the European Spallation Source (ESS) in Lund - Sweden, next generation experiments will happen during the upcoming decade. The weak interaction of neutrons with matter also imposes challenging counting statistics since the scattered signal is proportional to the crystal mass. Due to the fact that many field-related experiments require materials that can only be grown as single crystals, experimental advances are very slow. To enable more complex experiments, one needs to either increase the incident particle flux and/or increase the crystal mass. The latter is often not easily achievable as many single crystals are in the mg regime. A common procedure nowadays is to co-align a mosaic of single crystals to increase the overall crystal mass. Such mosaics can include hundreds of single crystals (an example is shown in Figure 1) but their creation process is obviously very labor intensive as it involves taking scattering snapshots, using for example a Laue diffractometer, and then manually adjusting the three possible rotational orientations to achieve a sufficient alignment of the atomic lattice structure to enable precise measurements. This is one of the major problems that is holding back many experiments and because of that scientific progress.

Figure 1: Example of a sample plate that holds many single crystals and thus forms a mosaic. Building such devices can quickly amount to hundreds of manual working hours. Courtesy to Qisi Wang.

1.2 Machine Learning

In recent years the field of artificial intelligence such as machine learning (ML) has opened-up new possibilities in a wide range of topics such as image processing (image super resolution [4] and image restoration [5]), pattern recognition (self-driving cars [6]), natural language processing (DeepL by DeepL SE, Alexa by Amazon, Google web search), stock market forecasting [7] and many more. ML algorithms are constructed in a way to extract specific intrinsic features from a set of data, which allows them to learn underlying classifiers and predictors [8]. ML encompasses fields such as supervised, unsupervised and reinforcement learning and commonly involves the usage of (deep) neural networks. The concept of supervised learning methods is based on learning data-specific features from explicitly labeled data. Regression methods are then used to find the best-performing model for the given data via an optimization routine [8]. Supervised learning is often used in image processing tasks such as image denoising for which different neural network architectures have been introduced in the last couple of years [9–11]. A classical benchmark problem in the field of denoising is that of simulated white Gaussian noise, which is artificially added to natural photographs where then a neural network is trained to remove this noise [12–15]. In this supervised learning scheme, the input data is the noisy image and the label is the noise-free image (ground truth). Therefore, this paradigm is often denoted as "noise2truth". There exist also other denoising-based approaches that don't require the ground truth images but rather two noisy inputs ("noise2noise") [16] or also just a single noisy input ("noise2void") [17]. Recently, researchers have trained a deep neural network to produce high-count angle-resolved photoemission spectroscopy data from artificially generated noisy low-count data where it was assumed that the noise statistic follows a Poisson distribution [18].

One of the major bottlenecks of such supervised networks is the need of a vast amount of labeled data. This is one of the reasons why the branch of reinforcement learning (RL) has gained increasing attention due to its ability of self-learning without the need of having access to labeled data. Among others, RL has been successfully applied in the field of game theory. The paradigm of RL is based on an agent that is situated in an environment in a specific state where it can take actions to obtain a reward as depicted in Figure 2. The main goal is to find an optimal policy that maximizes the total reward. One example is the computer program AlphaZero developed by Google's subsidiary company DeepMind, which recently has achieved superhuman performance in various games such as Pong, Go or chess [19]. But also in other simulated environments similar algorithms have been trained to achieve groundbreaking performance records [20].

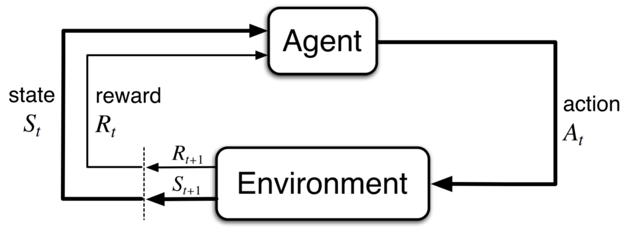

Figure 2: Reinforcement learning and the underlying agent-environment interaction [21].

2 Proposed research activities

Both X-ray and neutron diffraction experiments require excellent counting statistics as well as a consistent and precise alignment of the studied samples in a three-dimensional space. Both of these requirements involve a significant amount of manual work and are often limited by the scarce beam time available. We therefore plan to utilize new algorithmic approaches to increase the efficiency of such data acquisition processes. This proposal focuses on two different main projects both of which are related to computer-assisted scattering experiments. The first project involves the implementation of a deep convolutional neural network (CNN) for denoising low-count statistic X-ray images that is predicting a high-count statistic image from a low-count statistic image in a supervised learning scheme. The trained CNN could serve as an indication tool for future experiments to better organize and manage the limited amount of available beam time. The second project will focus on the implementation of a semi-automated mosaic single crystal alignment procedure via Laue diffraction patterns. Automating the creation of crystal mosaics will open numerous opportunities for new experiments in condensed matter fields such as magnetism, superconductivity, Mott insulators and spin liquids. The last project will build upon the second project by extending the crystal alignment procedure to the regime of reinforcement learning.

2.1 Supervised X-ray denoising

The concept of noise filtering has taken advantage of machine learning (ML) algorithms over the past decade. Especially deep convolutional neural networks (CNN) have been very successful [9–11]. As previously described, the classical benchmark problem consists of adding white Gaussian noise to photographs where then a CNN is trained to remove that artificial noise. Scattering experiments however include different types of noise including Poisson noise, which poses a bigger challenge to filter. The project aims to a) apply supervised machine learning to real data with Poisson noise and b) test the noise2noise methodology on real data. Due to the supervised nature of the projects, the key point of data collection is an important aspect. The noise filtering algorithms will benefit future experiments where some of them offer by design only low counting statistics. For example, diffraction experiments carried out in pulsed magnetic fields or X-ray scattering experiments on organic system that require low doses to avoid beam damage. Yet another application is the small angle neutron scattering where the weak cross section naturally implies poor counting statistic. Often the data analysis includes background subtraction but with an effective noise filter, counting time on the background can be reduced (giving better counting statistics on the foreground).

Noise filters generated by supervised ML techniques generally require millions of pixels to allow for a successful training without overfitting the network. One of such X-ray data sets has already been acquired by Julia Küspert and Ruggero Frison during experiments at the P21 diffractometer, PETRA-III synchrotron at DESY (Hamburg) containing a pixel count of the order of 10^8. Low and high statistic data were recorded systematically. Such frames of low- and high-counting statistic pairs (around 2 respectively 21 seconds exposure time) will be the basis of our training where we will consider three noise problems:

i) Artificial white Gaussian noise

ii) Artificial Poisson noise

ii) Real experimental Poisson noise

These three problems describe a hierarchy of difficulty where noise filtering of real data is the most difficult part, which therefore will be the focus of this work. Initial results of training on real X-ray diffraction data are shown in Figure 3. During this project, we will extend the work to also include small angle neutron scattering data.

For the "noise2noise" training [16] on real data with Poisson statistics, new data needs to be recorded. Here the data structure will be pairs of low-count statistic data and a high-count statistics data frame as reference for later comparison. Beam time proposals will be submitted to the PETRA-III synchrotron to record such data. Obviously, this concept is bound to be less successful as less information is available. Yet, it can have applications for experiments where high statistic is never reached.

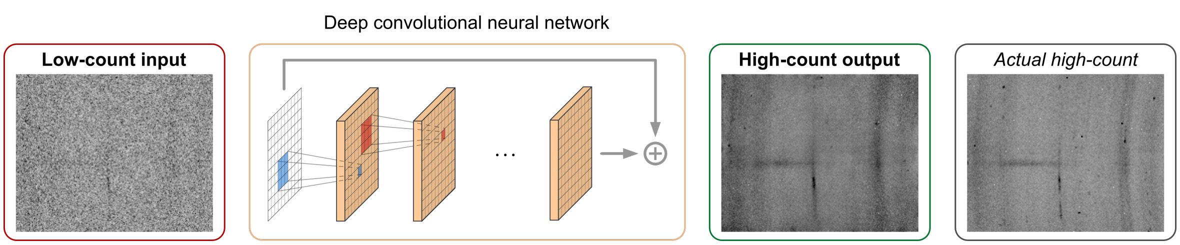

Figure 3: Concept of denoising X-ray diffraction data using a deep convolutional neural network (CNN), which is trained on low-count (LC) and high-count (HC) statistic pairs to remove the noise from unseen noisy input data. The data used here has been acquired at the PETRA-III P21 diffractometer at DESY in Hamburg on the high-temperature cuprate superconductor $La_{2-x}Sr_xCuO_4$ ($x = 0.12$) where the sample was detwinned by uniaxial pressure along the b-axis. The LC corresponds to exposure times of 2 seconds whereas the HC corresponds to exposure times of 21 seconds. The deep CNN (middle panel) is trained on these pairs and should then be able to predict a HC image ("High-count output") from a noisy LC image ("Low-count input") within a fraction of a second. This procedure would help greatly in the decision-making during live experiments.

2.2 Alignment of single crystal mosaics

Here, we propose a semi-automated alignment procedure of crystal mosaics. This method will in addition to reduction of manpower also increase the overall precision of the alignment that is currently mostly done manually. We choose to focus on two common geometries:

i) Two-dimensional materials growing in flake-like formats

ii) Tetragonal crystal structures growing in rod-like format

For two-dimensional flaky materials we envision a procedure that is compatible with existing experimental equipment. The group of Prof. Johan Chang is in possession of a commercial X-ray Laue diffractometer that is already equipped with motors for translational movements. Initially, a batch of around 100 to 200 single crystals will be glued and placed regularly on a metal plate. Laue patterns using non-monochromatic X-rays will then be collected in an automated fashion. In a next step, specific algorithms shall be developed that can automatically analyze and characterize these patterns in terms of translational or rotational operations required for their alignment. A schematic depiction of this process is illustrated in Figure 4.

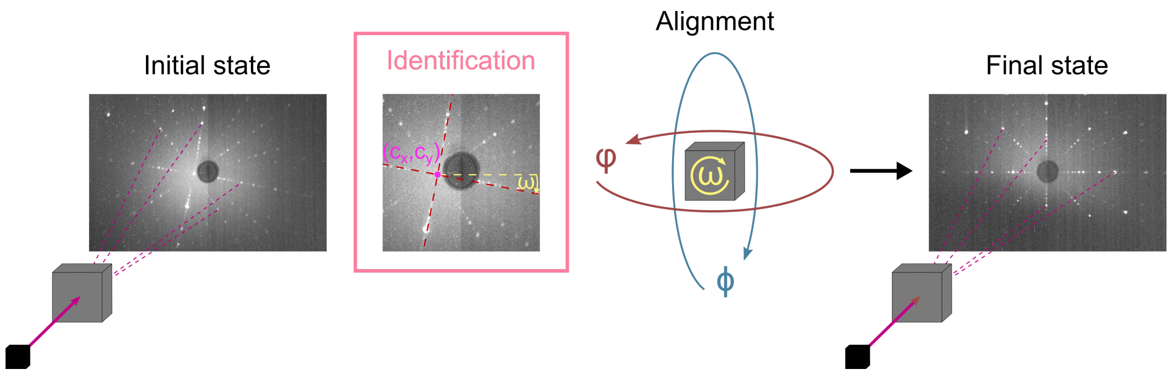

Figure 4: Semi-automated single crystal alignment procedure. A robust algorithm is provided with a Laue diffraction pattern where it will identify the center (c_x, c_y) (related to polar and azimuthal angle (φ, ϕ)) and the in-plane rotation angle ω. This information is then passed to the physical motors, which then will align the sample properly.

Using this analysis as an input, different algorithms shall be implemented for designing a dense packed mosaic arrangement on a sample plate. We envision this design to be manufactured by 3D printing methodology. The final steps involve the placement of the individual single crystals in their 3D-printed matrix. The methodology is then double checked by repeating the Laue automated data collection and analysis. This semi-automated procedure will remarkably accelerate the co-alignment procedure of single crystals. We plan to apply this methodology to different condensed matter topics such as

a) Mott physics represented by Ca_2RuO_4 and $Ca_2Ru_{1-x}Mn_xO_4$

b) Superconductivity represented by YFe_2Ge_2

c) Magnetism represented by $FePS_3$, $FePSe_3$ or $Sr_4Ru_3O_{10}$

d) Spin liquid related compounds such as $KFe_3(OH)_6(SO_4)_2$

The alignment of crystals with rod-like geometry follows a similar procedure but typically involves fewer crystals. We again plan to utilize Laue diffraction patterns to automate the data analysis including CAD drawing programs, which will be used to make specially designed sample holders that can be 3D-printed. This will, in addition to spectroscopy experiments, create new viable options for diffraction techniques within the above mentioned topics, all of which are active and vivid condensed matter sub-fields. Experiments will be carried out at and in collaboration with the Swiss Neutron Spallation Source (SINQ). We also envision the use of international facilities such as ESS in Lund and ILL in Grenoble, which has a built-in four-axis Euler goniometer that is compatible with the local Laue instrument.

2.3 Single crystal alignment using reinforcement learning

We plan to extend the previously described semi-automated single crystal alignment process to the branch of reinforcement learning (RL). Many diffraction and spectroscopy experiments start with the manual alignment of a single crystal. That is to bring a chosen crystal plane into a horizontal scattering plane. Once this is reached, a so-called UB-matrix ensures the script-based access to all points in the reciprocal space. The alignment and setup of the UB-matrix is still manual work. Even though this process in principle straight forward, it almost always takes hours to complete. At synchrotrons and neutron scattering facilities a day of beam time cost the society in the order of 10-20k CHF. Generally, single crystal alignment typically takes 2-4 hours using manual human labor. A suited algorithm would be much more efficient, both time and precision wise. The remaining limitation would be due to the motor movements. Every successful run is thus saving up to 3k CHF. Assuming just one successful execution every week, a giving beam line would annually gain beam time worth of 150k CHF. Worldwide there exist at least 50 of similar instruments that would benefit from such an algorithmic solution, which implies that the annual value of this work could be in the order of 5-10 million CHF. This project thus might have a commercial potential and could add to the entrepreneurial portfolio of the University of Zurich.

This project will be approached in several steps. We will start with a virtual experiment where Laue patterns are simulated. A virtual single crystal will be given a random orientation and the task is to bring a particular crystal plane to the horizontal scattering plane for example. With some education and experience, humans can usually do this job given enough time. Our aim is to produce a RL agent that can execute this task much faster than any human. The algorithm will get Laue patterns and start Euler/arm coordinates as input information. Based on the Laue pattern, the algorithm must produce new coordinates that in turn produce another Laue pattern. Eventually, the algorithm must find the perfect alignment between crystal and scattering plane, which is the analogy to the check-mate status in chess and is so to say the end of the "game" of the single crystal alignment process. By continuous training, the RL agent should become more and more skillful in playing the "game" and surpass human speed.

Once the developed RL agent has been successfully applied on this virtual experiment we will move towards more real experimental conditions. An Euler goniometer or a miniature six-axis robotic arm will be installed at the local Laue instrument. This gives us all degrees of motion required for the problem. Now the algorithm will be tested on real data from real crystals, which presumably will require renewed training due to the properties of real experimental data including various noise and imaging artifacts that might be present. Once succeeding this step, we are heading to test this work at international scattering facilities such as the neutron Laue instrument at the ILL in Grenoble. The entire process will be repeated for monochromatic diffraction instrumentation. First, the algorithm is trained by virtual experiments because within the UZH physics institute we don't have access to any instruments on which training can be done with real experimental conditions. Therefore, we later would seek collaboration with X-ray laboratories or synchrotrons such as the P21 beam line at the PETRA-III synchrotron, operated by Oleh Ivashko - a former UZH PhD graduate.

3 Available resources

We will use a local Laue diffractometer that operates with non-monochromatic X-rays for the basic implementation of the crystal alignment project. Furthermore, some of the proposed projects will require hardware-accelerated computations by taking advantage of state-of-the-art graphics processing units (GPU). As of now the group of Prof. Nicolas Serra is in possession of two high-performance Nvidia Tesla P100 GPUs that could eventually be used for the planned projects. Other options would be to use the UZH ScienceCluster managed by S^3IT or to apply for computation time on an external cluster such as the Swiss National Supercomputing Centre (CSCS).

4 Significance

The problem of scarce beam time is a limiting factor in terms of acquiring enough high-statistic data in scattering experiments where one must decide which of the wide range of potential experimental parameters will yield the anticipated results. With the implementation of a denoising deep convolutional neural network researchers would have access to a tool, which allows them to better plan and steer on-going experiments by being able to obtain a high-statistic output from a low-statistic input in a fraction of a second. A similar goal is targeted with the implementation of an automated single crystal alignment procedure, which would a) increase the efficiency and b) allow for a more sustainable way of operation of over 50 synchrotrons and neutron scattering facilities currently used world-wide. The later project may also open new opportunities, especially in the field of neutron scattering experiments, where the proposed algorithmic approach could enable the automated alignment of thousands of single crystals in a short amount of time and thus enable entirely new experiments.

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