SIMULATIONS OF HIGH-ANGLE ANNULAR DARK FIELD IMAGES OF CRYSTALS

Project report
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ABSTRACT

Multislice HAADF-STEM image simulations of SrTiO$_3$ are performed at 300 K. The procedure of these simulations and the used techniques are briefly explained and reasoned. The results are presented and discussed in a concise way and in an attached paper a comparison to experimental images is made. The paper proofs that the electron optical setup developed in Dresden is indeed capable of producing atomic-sized EVBs, a precondition for measuring EMCD with atomic resolution.
Results of this project work have been published in *Scientific Reports*:


**ACKNOWLEDGMENTS**

My deep gratitude goes to my supervisor Ján Rusz for the opportunity to work on this project and his great patience and support. Furthermore I would like thank the other authors, especially Dr. Darius Pohl, for the opportunity to work with them on this project. For computational resources I would like to thank the Swedish super computing centers SNIC and NSC.
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<table>
<thead>
<tr>
<th>Acronym</th>
<th>Description</th>
</tr>
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<tbody>
<tr>
<td>HAADF</td>
<td>high-angle annular dark field</td>
</tr>
<tr>
<td>HRTEM</td>
<td>high resolution transmission electron microscope/microscopy</td>
</tr>
<tr>
<td>EMCD</td>
<td>energy-loss magnetic circular dichroism</td>
</tr>
<tr>
<td>FWHM</td>
<td>full width at half maximum</td>
</tr>
<tr>
<td>HOLZ</td>
<td>higher order Laue-zone</td>
</tr>
<tr>
<td>RSMS</td>
<td>real space multislice</td>
</tr>
<tr>
<td>STEM</td>
<td>scanning transmission electron microscope</td>
</tr>
<tr>
<td>XMCD</td>
<td>X-ray magnetic circular dichroism</td>
</tr>
<tr>
<td>ADF</td>
<td>annular dark-field</td>
</tr>
<tr>
<td>BFP</td>
<td>back-focal plane</td>
</tr>
<tr>
<td>CCD</td>
<td>charge-coupled device</td>
</tr>
<tr>
<td>EVB</td>
<td>electron vortex beam</td>
</tr>
<tr>
<td>OAM</td>
<td>orbital angular momentum</td>
</tr>
<tr>
<td>STO</td>
<td>Strontium-Titanate (SrTiO$_3$)</td>
</tr>
<tr>
<td>TDS</td>
<td>thermal diffuse scattering</td>
</tr>
<tr>
<td>TEM</td>
<td>transmission electron microscope/microscopy</td>
</tr>
<tr>
<td>VLM</td>
<td>visual light microscopy</td>
</tr>
<tr>
<td>BF</td>
<td>bright-field</td>
</tr>
<tr>
<td>DF</td>
<td>dark-field</td>
</tr>
<tr>
<td>DP</td>
<td>diffraction pattern</td>
</tr>
<tr>
<td>FT</td>
<td>fourier transform</td>
</tr>
<tr>
<td>MD</td>
<td>molecular dynamics</td>
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<tr>
<td>PM</td>
<td>photomultiplier</td>
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Part I

REPORT

The main report containing background information and detailed information on the performed calculations.
In 2016 Ján Rusz, who was at that time supervising this report’s author’s Bachelor thesis, contacted the author about a possible side project about HAADF-STEM image simulations for a colleague of his from Dresden, Dr. Darius Pohl. They were developing at that time a new electron optical setup that allows for the generation of electron vortex beams (EVBs) of atomic size. Such atomic sized EVB beams are a prerequisite for atomic resolution energy-loss magnetic circular dichroism (EMCD) measurements. EMCD is a novel technique to measure the magnetic properties of a material exploiting the in a transmission electron microscope/microscopy (TEM) and is thereby the counterpart of the widely adopted X-ray magnetic circular dichroism (XMCD) technique[10]. It was first carried out experimentally by Schattschneider et al.[11]. The interested reader is pointed towards the introductory paragraphs of the attached paper, which contain a more thorough description and further references about EVBs and EMCD.

In order to compare to his experiment Dr. Darius Pohl requested HAADF-STEM calculations of a STO specimen for a range of thicknesses up to 60 nm. A convergent 300 keV-EVB of orbital angular momentum (OAM) \( l = 1 \) and convergence angle 22.6 mrad was to be used and the experimental inner and outer collection angle were 49.5 mrad and 200 mrad, respectively. Furthermore the source size broadening was to be considered for a suitable range of values between 0 pm and 200 pm.

The outcomes of this project have been included in an article in Scientific Reports and they are to be presented in the following. This report is divided into three parts: the first part wraps up this report’s author’s contribution to the paper and the second part contains the appendix with additional information about developed codes and settings of the simulations. In the first part chapter 2 gives an introduction to the theoretical concepts and the background of HAADF-STEM imaging and image simulations. Chapter 3 describes in detail the calculations carried out and chapter 4 the results. A conclusion is drawn in chapter 5.
BACKGROUND AND THEORY

This chapter is dedicated to a brief overview of the necessary theory required to understand the performed simulations. The conceptual idea of a TEM, the multislice algorithm and EVBs are explained in section 2.1. Thereafter section 2.2 deals with the HAADF imaging technique. Section 2.3 completes the discussion of the theoretical background by giving a brief description of classical molecular dynamics.

2.1 TRANSMISSION ELECTRON MICROSCOPY

This chapter summarizes briefly the important aspects of chapters 1-10 of the book by Williams and Carter[15]: Transmission electron microscopy is a versatile imaging and material characterization technique where electrons with energies of commonly 60-400 keV are used as probes in a similar way as photons probe samples in visual light microscopy (VLM). A TEM allows thereby for high spatial resolution, down to atomic resolution, which is a direct consequence of the small de-Broglie wavelength \( \lambda \) of electrons: The expression for the diffraction limited resolution according to the Rayleigh-criterion reads for parallel illumination as in VLM

\[
    r_{\text{diff}} = \frac{\lambda}{\beta}
\]

where \( \beta \) is the maximum collection angle. Thus increasing the acceleration voltage allows to lower the diffraction limited resolution, which is principle of high resolution transmission electron microscope/microscopy (HRTEM). Due to the in comparison with optical lenses poor quality of electron lenses the resolution of static TEMs is actually limited by lens imperfections such as spherical and chromatic aberration, and astigmatism. An introductory discussion of those effects can be found in chapter 6 of the book by Williams and Carter[15].

In a TEM electrons get accelerated up to a specific kinetic energy and this beam of electrons is guided to illuminate a specimen by the use of electron lenses. If the specimen is thin enough, i.e. it is transparent to electrons, the electrons pass through the specimen as the "T" for transmission in the abbreviation TEM emphasizes. A detector, such as a scintillator-photomultiplier (PM) tube or a charge-coupled device (CCD) camera, is located behind the sample in the beam line of the microscope. It detects the scattered electrons and an diffraction pattern (DP) or image is formed by measuring millions of electrons.

One distinguishes two types of illuminating beams: parallel and convergent electron beams. A parallel electron beam consists of electrons
that travel parallel to the optical axis of the TEM, as the name suggests. A convergent beam on the other hand is a beam that is focused on a certain point of or close to the specimen. The electrons constituting such a beam travel thus on trajectories that are angled towards the optical axis. The maximum angle between the beam of electrons and the optical axis is called the convergence angle $\alpha$. Figure 2.1 depicts a convergent beam schematically.

Inside the specimen a multitude of scattering (interaction) processes between the beam electrons and the specimen’s atomic cores and their shell electrons happens. However, the total cross section of most of these processes is very low and the majority of the beam electrons pass the specimen without being deflected from the optical axis. These electrons constitute the so-called direct or non-scattered beam. Different scattering processes have different angular distributions of their cross section and the selection of electrons scattered to specific ranges of angles allows thus to limit which scattering processes are contributing to the experiment. Experimentally this is achieved by apertures of various diameters, which block unwanted electrons.

Two important “outputs” of a TEM-experiment are the DP and the image. The DP is formed in the back-focal plane (BFP) of the objective lens in a TEM. It shows the angular electron beam intensity variations behind the specimen. The image, by contrast, is formed in the Gaussian image plane of the objective lens whereas the image shows the spatial electron beam intensity variations behind the specimen.

TEMs generally allow to capture bright-field (BF) or dark-field (DF) images. BF images are formed by selecting the direct beam for imaging, which consists mainly of those electrons that are forward scattered under small angles with respect to the beam direction. Contrary DF images are formed by selecting scattered/diffracted beams for imaging. One dis-
tunguishes two important types of DF images: annular dark-field (ADF) and HAADF images. ADF detectors exclude all but electrons that are scattered towards a range of small angles thereby excluding the direct beam. The name stems from the annular shape. HAADF detectors allow for the collection of only those electrons that are scattered to large angles.

One widely used type of a TEM is the scanning transmission electron microscope (STEM). It allows for the electron beam to be "scanned" over the surface of the sample, i.e. it is not statically centered on the area of interest of the specimen. The imaging process in a STEM is thus different from the imaging process in static TEM: In static TEM the electron beam is used to illuminate an area of the specimen and the objective lens and subsequent electron lenses form the image or diffraction pattern. The resolution is thus limited by the quality of the optical (imaging) system. Parallel and convergent beams can be used in a static TEM.

The imaging process in STEM, by contrast, does not require subsequent imaging lenses: The beam is scanned over the area of interest of the sample and at each beam position the total intensity of electrons reaching the detector is measured. Then the image reveals itself on a computer screen in a pixel-by-pixel manner as the scan proceeds. The resolution of this process is limited by the probe (beam) size: STEM requires a convergent beam because only this type of beam allows for the formation of sufficiently small probes. The expression for the diffraction limited resolution becomes in the case of STEM

\[ r_{DL} = \frac{\lambda}{\alpha}, \]

where \( \alpha \) is the convergence angle of the electron beam.

An important tool to predict, confirm and interpret TEM results are computer simulations. One family of methods are the so-called multislice methods.

2.1.1 Multislice method

The measurement process of the electrons in the TEM detector corresponds to the collapse of the wave function \( \psi \) of the electron within the Copenhagen interpretation of Quantum Mechanics. The DP and the image correspond thus to the probability distribution \( |\psi|^2 \) of finding an electron after passing through the specimen. Elastic scattering of an electron on a crystal lattice is quantum mechanically equivalent to the diffraction of its associated wave function \( \psi \), in close analogy to the diffraction of light by a grating, or more precisely an array of gratings. The electron wave function behind the specimen is commonly referred to as the exit wave function.

Thus one needs to solve the Schrödinger equation in order simulate a TEM image or DP. Inside a TEM beam directed in \( \hat{z} \)-direction the wave
2.1 Transmission Electron Microscopy

Transmission electron microscopy

\[ \psi(\mathbf{r}) \text{ of the beam electrons satisfies the fast electron Schrödinger equation} \]

\[ \frac{\partial \psi(\mathbf{r})}{\partial z} = \left[ \frac{i \lambda}{4\pi} \left( \nabla_{xy}^2 + \sigma V(\mathbf{r}) \right) \right] \psi(\mathbf{r}), \quad (2.1) \]

where the interaction parameter \( \sigma = 2me/\hbar^2 \) and the relativistic electron mass \( m = \gamma m_0 = m_0 \frac{R+T}{R} \) is used. \( R = 511 \text{ keV} \) is thereby the electron’s rest mass and \( T \) its kinetic energy. The potential \( V(\mathbf{r}) \) encodes in equation (2.1) the influence of the scattering potential of the atomic sites of the specimen on the electron wave function. Given a scattering potential and an initial wave function \( \psi_0 \) at the surface of the specimen in \( z \)-direction, the solution to this equation can be numerically obtained to high accuracy by the so-called multislice method, which is based on the idea to propagate the wave function of the beam electrons through the specimen in a slice-by-slice manner.

Mathematically the potential \( V(\mathbf{r}) \) is thereby sliced into thin layers of thickness \( \epsilon_j \), \( 1 \leq j \leq N_z \), in which the potential is a taken to be a constant average (cf. figure 2.2) according to

\[ V_p(\mathbf{r}_j) = \frac{1}{\epsilon_j} \int_{\sum_{i=1}^{j-1} \epsilon_i}^{\sum_{i=1}^{j} \epsilon_i} V(\mathbf{r}) \, dz, \quad (2.2) \]

where \( \mathbf{r}_j \) is the set of points situated in layer \( j \) with \( z \)-coordinate \( \sum_{i=1}^{j-1} \epsilon_i \leq z_j \leq \sum_{i=1}^{j} \epsilon_i \). The solution to equation (2.1) at "depth" \( z = \sum_{j=1}^{n} \epsilon_j \) may then be expressed as:

\[ \psi(\mathbf{r}_n) = \prod_{j=1}^{n} \exp \left[ \frac{i \lambda \epsilon_j}{4\pi} \left( \nabla_{xy}^2 + \sigma V_p(\mathbf{r}_j) \right) \right] \psi(\mathbf{r}_0), \quad (2.3) \]

\( \mathbf{r}_0 \) are the points in the \( xy \)-plane at the surface of the specimen where \( z = 0 \) and the electron still possess its initial wave function \( \psi_0 = \psi(\mathbf{r}_0) \) produced by the optics of the TEM. Equation 2.3 can be computed in an iterative manner using the so-called real space multislice (RSMS)-method developed by Cai et al. [1].
One important aspect of multislice calculations at finite temperatures $T$ is the so-called frozen phonon approximation, which allows the final image or DP to be calculated as an average over a suitable set of multislice images or DPs of distorted crystal structures. In reality one often uses the Einstein-model of thermal vibrations and randomly displaces the atoms according to that. Another way is to simulate the atomic displacements using molecular dynamics (MD), which is done in this project. Wang describes in detail the reasoning behind the frozen phonon approximation [14]: Because the time between consecutive electrons in even very intense beams is very large compared to the time scale for atomic vibrations, each electron passes so to say through a different configuration of scattering atoms which is uncorrelated to the configuration which the previous electron "experienced". Thus one may consider the image or DP as a time average of many electron propagations through the specimen.

### 2.1.2 Electron vortex beams

A special choice of an incident beam is a so-called electron vortex beam (EVB) which carries an orbital angular momentum $l$. EVBs have been produced experimentally for the first time by Uchida and Tonomura[13]. The wave function of a free EVB reads

$$\psi_{\text{EVB}} \propto \exp(il\varphi) \exp(i2\pi k_z z),$$

where $l$ is the orbital momentum quantum number and $\varphi = \arctan(k_y/k_x)$ is the phase in $\hat{k}_x-\hat{k}_y$-direction. As one can see from equation (2.4) and the definition of $\varphi$ the wave front is in a sense spiraling around the $k_z$-direction. The phase of the EVB wave function exhibits a phase singularity at the origin, where $k_x = k_y = 0$, and $\psi_{\text{EVB}}$ needs thus to vanish there. This gives the EVB wave function a donut-like shape in $k_x$-$k_y$ plane.

### 2.2 HIGH-ANGLE ANNULAR DARK FIELD IMAGING

In HAADF imaging one limits the electrons contributing to the image to those that are incoherently scattered to large angles. This imaging method is based on so-called $Z$-contrast, which refers to the circumstance that atoms which have a large nuclear charge $Z$ scatter electrons stronger than atoms that possess a nucleus with a lower charge. Before explaining the origin of this $Z$-contrast it is helpful to define what is exactly meant by contrast: Image contrast is defined as the intensity difference between neighboring parts of a picture. A high contrast makes it easier to distinguish both parts. In order to be able to distinguish features of a specimen in a TEM image one needs enough contrast between the these features. But how precisely does HAADF-STEM imaging work? A good description of $Z$-contrast imaging given by Pennycook[7].

Most electrons are (quasi-)elastically scattered in TEM. Elastic scattering can generally be divided into two types: coherent and incoherent scat-
2.2 High-angle annular dark field imaging

Both originate from the elastic scattering of electrons on the attractive Coulomb potential of the atomic cores of the specimen and this process is thus the most important scattering process with respect to contrast in TEM\cite{9}.

The basic process of elastic scattering common to both incoherent and coherent scattering is so-called Rutherford scattering: the scattering cross section for elastic scattering of a charged particle on a screened, attractive Coulomb potential is the famous Rutherford differential scattering cross section which reads

$$\sigma_R(\theta) = \frac{Z^2 \lambda_R^4}{64 \pi^4 a_0^2} \frac{d\Omega}{\sin^2(\frac{\theta}{2}) + \frac{\theta_0^2}{4}}$$

(2.5)

after being corrected for relativistic effects through the relativistic wavelength $\lambda_R$. $\theta_0$ is a screening parameter and $a_0$ is the Bohr radius. Strictly speaking the Rutherford cross section should not be used for electrons since they possess a spin moment which interacts magnetically with the orbital angular momentum of the scattering electron. This situation is described by the so-called Mott cross section. The Rutherford cross section is nevertheless suitable for an introductory understanding of the HAADF imaging method.

Coherent scattering arises then from the constructive and destructive interference of the electron wave after interaction with many atomic sites in a crystalline specimen and gives rise to a distinct spot pattern, the diffraction pattern. This mode of elastic scattering is called Bragg scattering or simply diffraction. The periodicity of the crystal lattice plays a key role in this process since only a periodic array of scatterers can produce a diffraction pattern because the scattered electron wave function is coherent, i.e. there is a defined phase dependence between two scattered waves.

The uncorrelated thermal vibrations of crystalline materials at finite temperature destroy their perfect periodicity and the diffraction pattern becomes blurred by so-called thermal diffuse scattering (TDS). The Bragg spot intensities become reduced by the so-called Debye-Waller factor \cite{4}

$$\exp\left(-\frac{1}{3} \langle u^2 \rangle G^2\right)$$

(2.6)

where $\langle u^2 \rangle$ is the mean square displacement and $G$ is the magnitude of the reciprocal lattice vector of the respective Bragg spot. The mean square displacement is a temperature dependent property of the studied system and increases generally with temperature $T$. Thus the Bragg spot intensity is lowered due to an increase in temperature and the lowering is the stronger the greater the magnitude of the reciprocal lattice vector of the respective Bragg spot is. TDS additionally blurs the DP, making it harder to distinguish Bragg spots in the DP. Temperature reduces therefore the angular size of the diffraction pattern and suppresses higher order Laue-
Figure 2.3: Schematic drawing how $Z$-contrast develops in HAADF-STEM imaging. Regions of the specimen where the atomic charge is lower in comparison with another region of higher charge appear in a HAADF-STEM image darker due to in comparison lower scattering towards high angles. This figure is inspired by figure 22.4 in Williams and Carter [15].

The beam size is very important for the resolution of HAADF-STEM images as mentioned in section 2.1.

2.3 Classical Molecular Dynamics

Classical MD is a versatile simulation technique for the dynamics of atomic systems consisting of hundreds up to millions of atoms: given initial...
conditions \((q_1, \ldots, q_N)\) and \((p_1, \ldots, p_N)\) the phase space trajectory is computed by integrating the Hamilton’s equations of motion

\[
\dot{p}_j = -\frac{\partial H}{\partial q_j}, \quad j = 1, \ldots N
\]  
\[
\dot{q}_j = \frac{\partial H}{\partial p_j}
\]  

in time. Classical MD is thus a technique based on classical physics which is a suitable approximation for high temperatures \(T\) and large atomic masses.

Various thermostats allow to enforce certain thermodynamical conditions. The \(NVT\)-thermostat is used for example when one wants to study the systems in question within the \(NVT\)-ensemble, i.e. constant particle number \(N\), volume \(V\) and temperature \(T\). In order to achieve a constant temperature throughout the simulation so-called Nosé-Hoover thermostats add an additional friction term to the equations of motion \((2.7)\) and \((2.8)\), which allows to add or remove kinetic energy to the system as needed to keep the overall temperature constant\([3, 6]\). The LAMMPS software package implements actually a further refined thermostating scheme published by Shinoda et al.\([5, 12]\).
The general procedure of the HAADF-STEM image simulation carried out in this project is as follows: First classical MD is used to simulate the thermal equilibrium vibrations of the STO crystal structure. "Snapshots" of the positions of all the atoms in the crystal structure are thereby taken at certain time steps of the MD simulation. HAADF-STEM images are computed from these snapshots at certain thicknesses by means of RSMS calculations. In the following each of these steps is described in more detail.

The classical MD simulations are performed using the LAMMPS software package [5]. The inter-atomic forces are thereby modeled by a combination of Born, Morse and Coulomb potentials that was developed by Goh et al. [2]. A NVT-thermostat at 300K is used to keep the simulation in thermal equilibrium at constant particle number $N$, constant temperature $T$, and constant Volume $V$. A time step of 1 fs is used and snapshots are taken every 1000 time steps. The first 12 snapshots are discarded because the simulation is not yet stabilized at those times and only every fourth of the following snapshots is considered for further calculations in order to minimize correlation effects. Appendix A contains the content of the LAMMPS input file which gives more details about the exact settings of the MD simulation.

Recalling section 2.2 HAADF-STEM images are experimentally acquired by scanning the beam over the specimen and selecting only electrons that have been scattered to high angles for imaging. In an image simulation one puts a grid over the area of interest which is in this case one unit cell of STO. Each of the grid points corresponds thus to one beam position and the finer one chooses this grid the finer the resulting image will be but the whole simulation will as well become computationally more demanding. In this project the grid was chosen to have a size of $25 \times 25$ over one whole unit cell. Using rotational and translational symmetries one can reduce these 625 beam positions to 1461. At each of these beam positions one then needs to compute the electron wave function and exclude those parts that are blocked by the HAADF detector. In this project a lower collection angle of 50 mrad and an upper collection angle of 200 mrad are used as they are experimentally accessible values in Dresden.

As described in section 2.1.1 the RSMS method is a way of calculating the electron’s (exit) wave function after passing through an arbitrary configuration of scattering atoms. The reciprocal (k-) space exit wave function can be easily obtained by a fourier transform (FT) and its intensity corresponds to the DP. If the initial beam direction coincides with

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1 A computer code that determines the necessary beam positions has been developed for this project. Please see appendix B.1
the z-direction the intensity distribution in the diffraction pattern signifies which fraction of the total beam intensity is scattered towards which angle since each point in the diffraction pattern has a defined $k_x$ and $k_y$ coordinate from which the scattering angle can be deduced.

Thus it becomes an easy task to exclude the electrons blocked by the HAADF detector in reciprocal space, where one just discards those parts of the diffraction pattern (the electron beam) that do not fall within the limits set by the upper and lower collection angle. The remaining intensity gets then integrated to obtain a quantity that shall be called HAADF electron intensity in the following.

In order to get a thermal average of the HAADF electron intensity within the frozen phonon approximation one averages the HAADF electron intensity at for the respective beam position over all considered snapshots. The HAADF-STEM image is then formed by performing these computations for all considered beam positions and depicting them as an image, where the pixel position is the beam position and the color (gray scale) encodes the value of the electron intensity. A last step in calculating realistic images is to add a Gaussian blur which accounts for source size broadening, i.e. that the cone of the convergent electron beam is not perfectly pointy but flattened. Since the experimental source size is unknown the full width at half maximum (FWHM) of the Gaussian blur is varied between 0 Å and 1 Å.

The exit wave function is computed using an in-house developed code that implements the RSMS method. The initial beam is chosen to be an EVB with angular momentum $l = 1$, convergence angle $\alpha = 22.6$ mrad, and it is directed along the [001]-direction in STO. The acceleration voltage is set to 300 keV.
Figure 4.1 gives an overview of half of the images that are calculated for this project. The other half considered source size broadenings of more than 100 pm and it was quickly found that the experimental source size broadening is well below 100 pm. Only a fraction of these images are actually included in supplementary figure S2 of the final paper.

All of the HAADF-STEM images in figure 4.1 are atomic resolution images since the electron intensities associated with different atomic sites are well separated and one can be differentiated from each other. Furthermore the Z contrast between different atomic columns of STO becomes apparent: the brighter spots correspond to the Strontium atoms which scatter more strongly than the Titanium atoms which are situated at the sites with darker intensity. The Oxygen atoms deliver not enough Z-contrast to be visible in these images due to their low atomic charge.

The signature of the EVB can be seen in the donut shaped intensity spots at the atomic sites. Since an EVB has a zero intensity right in the center of the spiraling wave front the scattered intensity of the beam is reduced when the beam is centered exactly on an atomic column because more electrons pass further away from the atomic core and get thus scattered less. The maximum scattered intensity is observed for beam positions slightly off the atomic column as long as the source size broadening is not too large. The effect of source size broadening is to increase the intensity of the central minimum at every atomic site and the blurring and broadening of the whole (donut) shaped spot. The central minimum disappears above a source size broadening of about 50 pm and the considered values for the broadening are never large enough to not allow for atomic resolution imaging.

The thickness has an influence on the background intensity between atomic sites. The intensity in those areas increases with increasing thickness.
Figure 4.1(a): Simulated HAADF images of STO.
Figure 4.1: Simulated HAADF images of STO using an electron vortex beam with \( \text{OAM } l = 1 \) at 300 K. The rows show the image produced for different specimen thicknesses between 0 nm and 60 nm in steps of about 2 nm. The columns correspond to a source size broadening between 0 pm and 100 pm. The intensities are normalized such that the maximum intensity in each image is 1 and thus depicted in white color.
CONCLUSION

HAADF-STEM images of STO are simulated at 300 K using the multislice algorithm in the frozen phonon approximation. These images do not show unexpected results or hints of new physics but they are a valuable resource in the analysis of the experimental HAADF-STEM images in the sense that the comparison of images at different thicknesses and source size broadenings give the experimental side arguments for the atomic size nature of their technique. After all this is a well rounded project and there are satisfactorily no open ends. From an experimental point of view the next step is to show that the proposed setup is indeed capable of measuring EMCD as mentioned in the paper.

Personally I am very grateful and happy to be part of this project. I enjoy performing those simulations and it is very motivating to compare them with an actual experiment. The exchange with colleagues from Germany was an enriching experience, too. This is in some sense physics at its best: theoretical modeling and experimental observation go hand in hand and create new knowledge.


Part II

APPENDIX

Additional material that is referenced in the main text.
A.1 CLASSICAL MD

LAMMPS simulation input file

log file.log

# real units:
# mass = grams/mole
# distance = Angstroms
# time = femtoseconds
# energy = Kcal/mole (thermochemical calorie = 4.184 J)
# velocity = Angstroms/femtosecond
# force = Kcal/mole-Angstrom
# torque = Kcal/mole
# temperature = Kelvin
# pressure = atmospheres
# dynamic viscosity = Poise
# charge = multiple of electron charge (1.0 is a proton)
# dipole = charge*Angstroms
# electric field = volts/Angstrom
# density = gram/cm^dim

# strontium titanate system

atom_style charge
units real
dimension 3
boundary p p p

# lattice
lattice custom 3.905 &
a1 1.0 0.0 0.0 &
a2 0.0 1.0 0.0 &
a3 0.0 0.0 1.0 &
basis 0.0 0.0 0.0 &
basis 0.5 0.5 0.5 &
basis 0.5 0.5 0.0 &
basis 0.5 0.0 0.5 &
basis 0.0 0.5 0.5

region simbox block 0 12 0 12 0 160
create_box 3 simbox
create_atoms 1 box basis 1 1 basis 2 2 &
    basis 3 3 basis 4 3 basis 5 3

group sr type 1

group ti type 2

group o type 3

mass 1 87.62
mass 2 47.867
mass 3 15.999

# from the paper of Goh
set group sr charge 2.0
set group ti charge 2.2
set group o charge -1.4

pair_style hybrid/overlay &
    born 11.0 &
    coul/long 11.0 &
    morse 11.0 &

pair_coeff * * coul/long
pair_coeff 1 1 born 0.26 0.26 3.692 0.0 0.0
pair_coeff 1 2 born 0.23 0.23 3.231 0.0 0.0
pair_coeff 1 3 born 0.31 0.31 3.696 0.0 0.0
pair_coeff 2 2 born 0.20 0.20 2.770 0.0 0.0
pair_coeff 2 3 born 0.28 0.28 3.235 0.0 0.0
pair_coeff 3 3 born 0.36 0.36 3.700 627.0 0.0
pair_coeff 2 3 morse 26.0 2.00 1.60

kspace_style ewald 0.0001
pair_modify tail yes shift no

# initialize
timestep 1.0

velocity all create 300.0 299792458 mom yes &
    rot yes dist gaussian

neighbor 5.5 bin
neigh_modify every 1 delay 0 check yes

# output
variable N equal step
variable Epot equal pe
variable Etotal equal etotal
variable T equal temp
variable Press equal press
variable V equal vol
variable kine equal ke
variable Lx equal lx/12
variable Ly equal ly/12
variable Lz equal lz/160

fix extra all print 1 &
"${N} ${T} ${V} ${Press} ${kine} &
${Epot} ${Etotal}" &
file strontium_titanate.out

fix cell all print 1 &
"${N} ${T} ${Lx} ${Ly} ${Lz}" &
file strontium_titanate.cell

dump snapshot all custom 1000 &
strontium_titanate.snaps id q type x y z

# production run
fix NVT all nvt temp 300.0 300.0 100.0
run 201000
B.1 Optimizing the Number of Required HAADF Beam Positions

FORTRAN code.

```fortran
program symopt

! Calculates the required number of beam positions for the computation of HAADF vortex beams for a cubic crystal and a given grid

! Vortex beam -> no mirror symmetry
! Cubic system -> exploit rotational and translational symmetry

implicit none

integer :: i,j,n=0

! Calculation grid within one unit cell
integer, parameter :: gridx = 25
integer, parameter :: gridy = 25

! Grid to perform symmetry operations on
integer, dimension(gridx,gridy) :: grid=0

! Output array for beam positions
integer, parameter :: pairleng = (gridy + (( (gridx + gridy) - 2) * ((gridx + gridy) - 1)) / 2)
integer, dimension(2, pairleng) :: output=0

print *, pairleng

! Variable to store the number of required beam positions
n = 0

! Cycle over one quarter of the grid and perform on grid point symmetry operations in order to minimize the number of needed beam positions
do i=1,gridx
  do j=1,gridy
    select case ( grid(i,j) )
      case (0)
call mark_beam_pos(i,j,n,grid,gridx,gridy, 
    output,pairleng)

    case (1,2)
        cycle
    end select
end do
end do

! Output results
print *, n
open(unit=10, file='grid.out', action='write')
open(unit=11, file='beam_pos.out', action='write')

! Output visualization of the necessary beam positions
do i=1,gridx
    write(10,*) grid(i,:)
end do

! Output beam positions
do i=1,pairleng
    write(11,*) output(:,i)
    1000 format(i3)
end do

close(10)
close(11)

end program symopt

subroutine mark_beam_pos(i,j,n,grid,gridx,gridy,output, 
    pairleng)

    integer :: i,j,k,l,n
    integer :: rux,ruy,llx,lly,rlx,rly
    integer :: gridx,gridy,pairleng
    integer, dimension(gridx,gridy) :: grid
    integer, dimension( 2, pairleng) :: output

    grid(i,j) = 1
    n = n + 1

    ! Use modified Cantor pairing function to assign each
    ! grid position
    output(1, (j + ( (i+j-2) * (i+j-1) ) / 2)) = i
    output(2, (j + ( (i+j-2) * (i+j-1) ) / 2)) = j

    ! Perform 4-fold rotational symmetry operation
    ! Rotation by pi/2
    llx = gridx - j + 1
    lly = i
    if ( grid(llx,lly) == 0) grid(llx,lly) = 2
    ! Rotation by pi
b.1 optimizing the number of required HAADF beam positions

rlx = gridx - i + 1
rly = gridy - j + 1
if ( grid(rlx,rly) == 0) grid(rlx,rly) = 2
! Rotation by 3\pi/2
rux = j
ruy = gridy - i + 1
if ( grid(rux,ruy) == 0) grid(rux,ruy) = 2

! Perform translational symmetry operation on all grid points
! Translate point grid(i,j)
do k = i, gridx, (gridx - 1)
  do l = j, gridy, (gridy - 1)
    if ( grid(k,l) == 0) grid(k,l) = 2
  end do
end do
! Translate point grid(llx,lly)
do k = llx, 0, -(gridx - 1)
  do l = lly, gridy, (gridy - 1)
    if ( grid(k,l) == 0) grid(k,l) = 2
  end do
end do
! Translate point grid(rlx,rly)
do k = rlx, 0, -(gridx - 1)
  do l = rly, 0, -(gridy - 1)
    if ( grid(k,l) == 0) grid(k,l) = 2
  end do
end do
! Translate point grid(rux,ruy)
do k = rux, gridx, (gridx - 1)
  do l = ruy, 0, -(gridy - 1)
    if ( grid(k,l) == 0) grid(k,l) = 2
  end do
end do

end subroutine mark_beam_pos