Towards cross-platform automated rotation electron diffraction

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Synopsis A DigitalMicrograph script was developed to coordinate TEM goniometer rotation and detector image recording for continuous rotational electron diffraction (cRED) data acquisition. Exploiting fast, automated data collection, it was revealed how experimental settings (selected area mode illumination, parallel nanoprobe mode illumination and different electron dose rate) affect the quality of cRED data.

Abstract A DigitalMicrograph script InsteaDMatic has been developed to facilitate rapid automated continuous rotation electron diffraction (cRED) data acquisition. The script coordinates microscope functions such as stage rotation and camera functions relevant for data collection, and stores the experiment metadata. The script is compatible with both JEOL and Thermo Fisher Scientific microscopes. A proof-of-concept has been performed through employing InsteaDMatic for data collection and structure determination of a ZSM-5 zeolite. The influence of illumination settings and electron dose rate on the quality of diffraction data, unit cell determination and structure solution has been investigated in order to optimize the data acquisition procedure.

Keywords: 3D electron diffraction, DigitalMicrograph script, automated data collection, structure determination, microED

1. Introduction

3D electron diffraction (3D ED) is a well-known technique for structure determination of solids, which is especially advantageous for studies of micro- and nanocrystals. So far, 3D ED data have been used for determination of more than 200 structures (Gemmi et al., 2019), such as zeolites (Jiang et al., 2011; Guo et al., 2015; Simancas et al., 2016; Lee et al., 2018; Bieseki et al., 2018; Zhang et al., 2018), metal-organic frameworks (Denysenko et al., 2011; Feyand et al., 2012; Wang, Rhauderwick et al., 2018; Lenzen et al., 2019), pharmaceuticals (van Genderen et al., 2018; Brázda et
al., 2019), and protein crystals (Nannenga et al., 2014; de la Cruz et al., 2017; Xu et al., 2019) and many others. 3D ED data is traditionally collected by stepwise tilting a crystal around an arbitrary tilt axis within the full tilt range of the microscope goniometer and recording a set of diffraction patterns (Kolb et al., 2007; Nannenga et al., 2014; Zhang et al., 2010). Examples of software which is able to perform crystal tilt, crystal position tracking and diffraction pattern acquisition are described in (Kolb et al., 2007; Wan et al., 2013; Zhang et al., 2010; Wan et al., 2013). A perspective approach to 3D ED, known as MicroED (Nannenga et al., 2014) or cRED, is based on a continuous data collection while the goniometer is rotating. Automation of cRED data collection helps to reduce large number of manual operations and to obtain reproducible results with less human efforts, especially for very large datasets. Up to date, a limited number of software packages are available, designed to interface with both the camera and the microscope and collect multiple diffraction patterns, and those are often commercial and closed source, e.g. iTEM software from Olympus Soft Imaging Solutions GmbH, Münster, Germany (Gemmi et al., 2015). Recently, a script has been developed for an open-source, widely-used in the cryo-electron microscopy community SerialEM software enabling large-scale MicroED data collection on Thermo Fisher Scientific (TFS) microscopes with electron detectors from various manufacturers (de la Cruz et al., 2019). In our lab for cRED data collection we developed Instamatic, a custom software designed for electron crystallography needs, which is able to control both microscope and camera (Smeets et al., 2018), and affords additional features such as crystal tracking through defocusing of diffraction pattern (Cichocka et al., 2018; Wang et al., 2019).

However, to the best of our knowledge, currently there is no widely applicable routine to collect 3D ED data using continuous rotation, and particularly there is no widely accessible and easy-to-install software available. The desired software should neither have steep learning curves nor require its own set calibrations to be performed nor complicated set-ups. Due to diversity of existing TEM platforms the software has to control both the camera and the microscope, dealing with different combinations of those. Here, we propose to employ DigitalMicrograph (DM) (Digital Micrograph GATAN, Pleasanton, CA, U.S.A.) as a mediator controlling hardware interactions between microscope and camera. We developed a dedicated DM script for automated cRED data collection. The script, which we named InsteaDMatic was tested on our Themis Z (TFS) equipped with a Gatan OneView camera and JEM2100F (JEOL) with Gatan Orius SC200D camera. The script is successfully trialling now in five labs worldwide equipped with JEM2100F, JEM3100F and TFS Talos microscopes with different Gatan cameras. The InsteaDMatic follows the same data collection workflow as described previously (Cichocka et al., 2018) but communicates to both the microscope and camera via DM interfaces. The benefit of this design philosophy is easiness of installation and enhanced transferability, since the DM software is an integral part of a vast majority of electron microscopy systems nowadays. To demonstrate the capability of the script, we collected high-quality data on a ZSM-5 zeolite with up to 0.80 Å resolution providing a solid basis for its structure solution. The data quality and the resulting
data statistics have been compared for crystals illuminated in selected area mode or in parallel nanoprobe mode. To highlight the advantages of the approach, parameters such as electron dose rate and monochromator focus have been tailored during the collection of RED data.

2. Experimental

2.1. Sample preparation

Thoroughly ground ZSM-5 aluminosilicate zeolite powder was dispersed in ethanol followed by an ultrasonic bath treatment for 5 minutes. A drop of the suspension was applied to a lacey carbon grid (Cu150P from Okenshoji Co., Ltd). The grid was then dried in air for 10 minutes and the sample holder with the grid was transferred to a TEM.

2.2. Experimental setup

The cRED experiments have been performed on a Themis Z microscope equipped with a Gatan OneView camera (4096 × 4096 pixels, pixel size: 15 µm). The OneView camera is well suited for cRED data acquisition, because it has essentially no readout deadtime when in the movie mode. In-situ data capture mode with 1024 × 1024 pixels resolution (binning × 4) was employed. cRED data were collected using a single-tilt TFS holder (±40°), without using a beam stopper. Since Themis Z is very stable both electrically and mechanically, a crystal tracking procedure described by (Cichocka et al., 2018) is not a prerequisite for keeping the crystal centered in the beam or selected area (SA) aperture. Before data acquisition, a standard TEM alignment routine was performed. All experiments were performed in the parallel illumination mode using a 50 µm C2 condenser aperture. The Z-height of the crystal was adjusted to the eucentric height in order to minimize its movement during tilting. Diffraction patterns were focused to obtain sharp spots in the diffraction mode. The exposure time was set up to be 0.3 s per diffraction frame, the rotation speed was 1.44°/s. cRED dataset with a total rotation range of 80° contains 185 frames, collected in approximately 55 s.

For JEM 2100F equipped with a Gatan Orius SC200D detector (2048 × 2048 pixels, pixel size: 7.4 µm) the exposure time and rotation speed were set up to be 0.5 s/frame and 0.444°/s, resulting in 209 frames within the total rotation range of 46.42° in 104.5 s.

Two different beam settings available on Themis Z were tested, further on referred to as selected area diffraction (SAED) and nanoprobe (NP) modes. In the SAED mode, a 40 µm SA aperture was inserted to limit the area used for diffraction, whereas in the NP mode the field of view was restricted by the beam size. Spot size 5 or 6 was usually used in the SAED mode, and spot size 11 in the NP mode. The electron dose on the specimen was controlled varying the monochromator focus.

2.3. Data processing and structure determination
Diffraction images were collected in .TIFF format and converted to SMV format (.img) using the `process_DM` python script (Smeets, 2019). The collected frames were processed with the `XDS` software (Kabsch, 2010) for the spot-finding, indexing, space-group assignment, data integration, scaling, and refinement. Previously determined lattice parameters and the space group (Olson et al., 1981) were used as an input, REFLECTING_RANGE_E.S.D. parameter in the XDS.INP file was set up to be 0.7 to include very sharp diffraction spots in the indexing procedure. Data statistics indicators provided in the output CORRECT.LP file were used further for data quality comparison. The reflection file for structure solution and refinement was obtained by merging several individual datasets from different crystals using the `XSAMPLE` sub-program. The structure was solved by `Sir2014` (Burla et al., 2015) and `SHELXT` (Sheldrick, 2008) and refined by `SHELXL` using electron atomic structure factors with the help of `Olex2` software (Dolomanov et al., 2009).

3. **InsteadMatic workflow**

`InsteadMatic` follows the data collection workflow described in (Cichocka et al., 2018) using the continuous rotation method for electron diffraction (Arndt & Wonacott, 1977; Nederlof et al., 2013; Nannenga et al., 2014; Gemmi et al., 2015). The same workflow has previously been implemented in Python in the program `Instamatic` (Smeets et al., 2018). However, `Instamatic` requires significant development to interface both the TEM and the camera APIs.

On the camera computer, `InsteadMatic` is run from DM and the GUI is shown in Error! Reference source not found.. Settings for data collection (exposure, binning, etc.) are controlled through the camera panel in DM. When an experiment is started by pressing the “Start” button at the very bottom of the GUI, the script enters a waiting state where it constantly polls the current $\alpha$ tilt value. Once a change larger than a pre-defined threshold (angle activation threshold, typically 0.2°) is detected, data acquisition is initiated. The threshold also serves to eliminate any existing backlash in the $\alpha$ tilt direction. Rotation can then be initiated through any means available, either using the knobs, through the TEM user interface, or software.

It is worth to mention that at present the DM API does not allow fine control over the rotation speed of the goniometer although this function is available on our microscope (Themis Z, TFS) through the TEMScripting interface, as well as other recent TFS/JEOL microscopes. To be able to control the rotation through DM, we implemented a custom Python script in `Instamatic` (Smeets, 2018) to synchronize rotation with data acquisition. The script establishes an interface with the TEM on the microscope computer and accepts connections over the network. A socket interface is then established using the program ‘netcat’ ([https://nmap.org/ncat/](https://nmap.org/ncat/)) on the camera computer through the DM function `LaunchExternalProcess`, which then communicates the requested rotation range and speed over the network to the microscope computer.
Once rotation has been detected, data acquisition is initiated. The DM script hooks into the live view of the OneView camera, and then constantly copies the front-most image to a pre-allocated “image buffer” whose size can be defined in the GUI of the script (“buffer size”) and corresponds to the maximum number of frames that are expected to be collected. Whenever the live view is updated, DM fires an event called `DataValueChangedEvent`, which signals the script to copy the frame. The exposure time and binning are therefore defined through the DM interface, and not through the script. Data collection may be interrupted at any time by pressing the “Stop” button. There is also an automatic check for the completion of data collection, by looking at the change of $\alpha$ tilt after every image cloning operation. When the change is equal to 0, the data collection loop breaks automatically. Finally, the script stores all relevant experimental metadata required for processing to a new directory, such as the rotation range, exposure time, camera length, etc. The image files are stored in the same directory in TIFF format, and can be converted to other desired formats (SMV and MRC) by running the `process_DM.py` script (Smeets, 2019). A flowchart of the workflow is shown in Figure 2. Detailed
instruction of usage can be found from the script. The script is compatible with DM version 2.0 (which introduced the `DataValueChangedEvent`) or newer, and can be used with any Gatan camera that supports a streaming live view.

**Figure 2** Flowchart of InsteaDMatic.
4. Application for structure determination of ZSM-5

A proof-of-concept has been performed through employing InsteaDMatic for data collection and further structure determination of a ZSM-5 aluminosilicate zeolite widely used in industry as a catalyst (Ji et al., 2017; Kunwar et al., 2016). ZSM-5 is relatively stable against beam damage, allowing multiply data to be collected from the same crystal. Consequently, a direct comparison of cRED data quality at different illumination settings becomes possible.

First, we tested InsteaDMatic on Themis Z with a Gatan One View CCD camera. A typical experiment was filmed in order to illustrate the procedure of cRED data acquisition, see Supporting Movie 1. The best Themis Z dataset demonstrated the completeness of more than 75% in the resolution shells ranging from 2.36 Å to 0.8 Å and $R_{\text{meas}}$ of 13.7% (see Table S1) enabling ab initio crystal structure solution from this one individual dataset. Unfortunately, the completeness of most individual datasets does not exceed 50% for the orthorhombic structure, and often only merged data can provide the correct structure (see below). Thus, the OneView camera was found to be well suited for experiments that require continuous read-out of the sensor. To check if the script would work on other cameras we tested it on an Orius SC200D detector installed on JEM 2100F. A “single-crystal” dataset collected over a rotation range of 46.42° reached the completeness of ~30% in the resolution shells from 2.36 Å to 0.8 Å, and exhibited the $R_{\text{meas}}$ of 26.1%. Although the data are of good quality, the microscope has limited tilting capabilities, and thus the low completeness of the data, prohibited a correct crystal structure solution by direct methods (Sir2014 or SHELXT).

Traditionally, collection of electron diffraction data has been performed with diffraction area selection using the selected-area aperture. However, area selection can also be accomplished by adjusting the illumination settings. Almost parallel illumination with sub-micron beam diameter can be obtained either by Köhler illumination (Wu et al., 2004; Meyer et al., 2006; Benner et al., 2011), or by inserting a small 10 µm C2 condenser aperture (Kolb et al., 2007; Dwyer et al., 2007). Nanoprobe gives full control on the beam diameter used and in principle also allows collecting data on a smaller area with respect to SAED (Gemmi et al., 2019). However, it is worth paying attention to the direct comparison of data quality collected on the same sample by cRED in SAED and NP modes, which is currently not available from the literature. Here, an attempt has been made to reveal the difference between these modes using the same area of the sample for collecting diffraction data. In the SAED mode the beam was spread to be roughly 6 µm in diameter at 13 kX magnification and a diffraction field of about 750 nm was selected by inserting the SA. In the NP mode the beam was condensed to illuminate the 750 nm area, and the electron dose rate was kept equal to those in the SAED mode (0.05 e/Å²s) by adjusting the monochromator focus. Two resulting datasets registered on the same isolated crystal are present in Table 1.
Table 1  Data collection parameters and data processing by XDS. Statistics in different resolution shells is given in Tables S2-S3.

|                                | SAED dataset | NP dataset |
|--------------------------------|--------------|------------|
| Spot size                      | 5            | 11         |
| Dose rate, e/Å²s               | 0.05         | 0.05       |
| Diffraction area, nm           | 750          | 750        |
| Tilt range, °                 | 39.64 to -40.00 | -39.71 to 40.00 |
| Oscillation angle, °           | 0.430        | 0.429      |
| Exposure time, s               | 0.30         | 0.30       |
| Acquisition time per frame, s  | 0.30         | 0.30       |
| Camera length, mm              | 580          | 580        |
| Mono focus                     | 100.34       | 78.89      |
| Rotation speed, °/s            | 1.441        | 1.434      |
| Total No. of reflections       | 17224        | 17825      |
| No. of unique reflections      | 2622         | 2692       |
| Completeness, %                | 47.1         | 48.3       |
| Resolution cutoff, Å           | 0.80         | 0.80       |
| $I/\sigma$                     | 4.19         | 4.42       |
| $R_{\text{obs}}$, %            | 20.8         | 21.7       |
| $R_{\text{exp}}$, %            | 23.9         | 24.8       |
| $R_{\text{meas}}$, %           | 22.8         | 23.9       |
| $CC_{1/2}$                     | 98.7         | 98.1       |
| Unit cell parameters           |              |            |
| a/Å                            | 20.38        | 20.56      |
| b/Å                            | 19.58        | 19.60      |
| c/Å                            | 13.21        | 13.18      |

Based on the previous crystallographic reports about the ZSM-5 crystal structure (Olson et al., 1981; van Koningsveld et al., 1987), the lattice parameters $a = 20.07$ Å, $b = 19.92$ Å, $c = 13.42$ Å and the space group $Pnma$ (#62) were used as an input for XDS. Both SAED and NP datasets fit well with the expected orthorhombic structure and the refined unit cell parameters are close to the published values within the accuracy of the electron diffraction method. Figure 3 shows the reconstructed reciprocal lattice of ZSM-5 based on the cRED data collected in the SAED mode, from Table 1.
Among factors affecting the cRED data quality electron dose has an utmost importance. Our experiments have been show that the optimal electron dose rate range for ZSM-5 data acquisition is between 0.05 e/Å²s and 0.1 e/Å²s (Figure 4). In the optimal range, higher the dose the better \( I/\sigma \), however we observed that \( I/\sigma \) for NP datasets is systematically slightly higher than for SAED ones, especially for the low-resolution data. Excessive electron dose (>0.2 e/Å²s) causes read-out biases of the OneView camera, whereas low electron dose rate <0.03 e/Å²s leads to significant deterioration of the signal-noise ratio and, as a consequence, to poor data statistics. Some of the raw SAED/NP diffraction patterns collected at different electron doses are shown in Figure S1.

It should be mentioned that unlike low-d Bragg peaks which can easily be discriminated from a slowly-varying background in diffraction patterns, the high-d peaks are often not very intense and thus cannot be readily separated from the background. Since XDS is relying upon the smallest

**Figure 3** (a-c) Typical 3D reciprocal lattice of ZSM-5 reconstructed and visualized by REDp (Wan *et al.*, 2013). The corresponding crystal image is shown as an inset in (b). (d) The wires/sticks ZSM-5 crystal structure representation.
measured intensities to guide the subtraction of the background, the scaling of the Bragg intensities as a function of resolution shells unavoidably leads to significant deteriorations of weak but still useful high-resolution signal, and consequently, to higher R-values in the 1.00 – 0.80 Å resolution shells. For X-ray diffraction a common practice would be a truncating data at the resolution at which $R_{\text{meas}}$ remains below ~60% and $\langle I/\sigma \rangle$ is ~2 or higher. However, for electron diffraction, we found that including data out to a $CC_{1/2}$ value (Karplus & Diederichs, 2015) of ~70% lead to an improvement of the refined model even though the data at that resolution have $R_{\text{meas}} \sim 200 – 300\%$.

**Figure 4** (a-b) Statistics of the cRED diffraction data collected in SAED and (c-d) NP modes for different electron doses: $I/\sigma$ and $CC_{1/2}$ against the resolution. All SAED data were collected from the same ZSM-5 crystal sequentially, in the ascending order of the electron dose rate. NP data were collected from the second crystal following the same procedure. All lines in the figure are a guide for the eye.
Another important factor for data collection is the stability of the CompuStage, since preserving the same illumination field and the same scattering volume during data collection is a key factor for reliable integration of the reflection intensities. We observed only a few-nm drift of the crystal with dimensions of about 100 × 100 nm inside the SA aperture in the tilting range from -40 to 40°, accompanied by a ~50 nm jump in the beginning of rotation, see Supporting Movie 2. Thus, the CompuStage was found to be perfectly stable for the cRED data acquisition. However, it is worth to mention that InsteaDMatic does not provide an opportunity to track the crystal during the experiment, thus, at higher tilt values the crystal might move out from the SA aperture due to uncompensated Z-height changes. Specific morphology of ZSM-5 crystals having a preferred orientation together with a limited tilting range accessible by the single-tilt holder often lead to the low completeness of the individual dataset. Therefore, it is necessary to merge data from several crystallites for a relevant structure solution. Partially this problem can be overcome by using a high-tilt tomography holder, with accessible tilt range ±80°, but the possible instability of the goniometer at high tilt angles should be always taken into account.

For the structure solution, five individual SAED datasets collected from different crystals were merged, chosen by performing hierarchical cluster analysis based on 15 experiments. Hierarchical cluster analysis helps to find structurally similar data with high correlation coefficients between scaled diffraction intensities and to reach high completeness by merging only few datasets (Wang et al., 2019). Sir2014 (Burla et al., 2015) direct space and SHELXT dual space methods (Sheldrick, 2008) can be both employed for the structure solution. We noted that a minimal signal-to-noise ratio of about 2 (in 1.0 Å resolution limit) is required for revealing the framework of ZSM-5 by means of direct methods, whereas dual space methods are not so sensitive to the I/σ ratio. The atomic positions of all 12 Si- and 24 O-atoms have been successfully found and used as an initial structural model. After the refinement, the model converged with $R_1=0.1998$ and GOF=1.59 (Table 2). All Si and O atoms were refined anisotropically (Figure 5) following by applying the rigid-body restraint (RIGU command (Thorn et al., 2012)). No additional restraints on Si-O bond lengths and O-Si-O angles have been applied. The three-dimensional channel system of ZSM-5 consists of straight channels running parallel to [010] having 10-rings of ca. 5.4 × 5.6 Å diameter and sinusoidal channels running parallel to [100] having 10-ring openings of ca. 5.1 × 5.4 Å, in full agreement with the previous XRD data (Olson et al., 1981; van Koningsveld et al., 1987).

The refined SAED model was compared with the reference ZSM-5 crystal structure (van Koningsveld et al., 1987) obtained from XRD data using COMPSTRU program (Flor et al., 2016). The average displacement between the corresponding Si atoms found to be 0.04(2) Å, whereas between O atoms the value is 0.06(4) Å. All deviations of atomic positions between the reference ZSM-5 structure (van Koningsveld et al., 1987) and those determined from cRED data are listed in Table S6. It is
interesting to compare the accuracy of the atomic positions determination for ZSM-5 with the data collected on a widely used JEM-2100 LaB₆ microscope (Wang, Yang et al., 2018). An average deviation from the same reference structure (van Koningsveld et al., 1987) of about 0.07(4) Å was reported, however, DFIX restraints were applied to Si—O distances (1.61 Å) (Wang, Yang et al., 2018). In our case a better accuracy is achieved with no additional geometry restraints.

Table 2  Selected crystallographic data for merged ZSM-5 datasets. Space group *Pnma* (#62), *Z* = 1, wavelength $\lambda$ = 0.019 Å. Statistics in different resolution shells is given in Tables S4-S5

|                  | SAED     | NP      |
|------------------|----------|---------|
| Datasets merged  | 5        | 6       |
| Averaged unit cell parameters |          |         |
| a/Å              | 20.19    | 20.10   |
| b/Å              | 19.56    | 19.47   |
| c/Å              | 12.95    | 13.30   |
| Total No. of reflections | 61596    | 65672   |
| No. of unique reflections | 5159      | 5299    |
| No. of reflections with $I > 2\sigma(I)$ | 2854      | 3903    |
| Completeness, %  | 95.8     | 98.2    |
| Resolution cutoff, Å | 0.80      | 0.80    |
| $I/\sigma$       | 3.56     | 4.56    |
| $R_{\text{obs}}$, % | 33.0      | 24.0    |
| $R_{\text{exp}}$, % | 33.8      | 27.7    |
| $R_{\text{meas}}$, % | 34.9      | 25.2    |
| $CC_{1/2}$       | 94.8     | 97.4    |
| $R_1$ ($I > 2\sigma(I)$) | 0.1998    | 0.1764  |
| $wR_2$ ($I > 2\sigma(I)$) | 0.2625    | 0.2004  |
| $R_1$ (all data) | 0.5109   | 0.4760  |
| $wR_2$ (all data) | 0.5490   | 0.4928  |
| GOF              | 1.59     | 1.61    |
The nanoprobe mode provides more flexibility in adjustment of the diffraction field diameter, which can be precisely fitted for each individual crystallite, in contrast to the SAED mode, which is limited by pre-defined aperture sizes. It should be also mentioned that in NP, when the illumination system is close to switching or has switched to condensing mode, the beam converges significantly faster as at standard settings causing some difficulties with the focus of diffraction patterns. NP datasets usually demonstrate higher signal-to-noise ratio as compared to the SAED data, whereas other crystallographic indicators are virtually of the same order (Table 1). The structure was solved from six merged NP datasets chosen from 10 datasets by means of hierarchical cluster analysis, and refined anisotropically to $R_1=0.1764$ and GOF=1.61 (Table 2). A comparison with the reference model (van Koningsveld et al., 1987) shows that the average displacements between the corresponding atoms are even lower than those for the SAED model (Table S6). The estimated average displacements between Si atoms is 0.03(1) Å, between O atoms – 0.05(3) Å, arguing for somewhat better data quality of the merged NP dataset.

Summarizing our findings, the selection of diffraction field by illumination provides reliable results comparable with the traditional SAED method and may be highly beneficial for studies of beam sensitive materials since it paves an avenue for tailoring of the electron dose received by a specimen in a controllable manner.

**Figure 5** Refined structure of ZSM-5 viewed along $b$-axis showing anisotropic atomic displacement parameters for Si (yellow) and O (red) atoms.
5. Conclusion

A new custom DigitalMicrograph script named *InsteaDMatic* has been developed to facilitate rapid automated electron diffraction data acquisition. *InsteaDMatic* was successfully examined on both JEOL and Thermo Fisher Scientific microscopes that utilize Gatan DigitalMicrograph for control over the instrument. The script was employed for data collection and structural determination of ZSM-5 zeolite framework. Dose rate between 0.05 e/Å²s and 0.10 e/Å²s found to be optimal for obtaining high quality data with up to 0.80 Å resolution. Positions of the Si and O atoms in ZSM-5 can be found within an accuracy better than 0.06 Å, compared to those obtained by single-crystal XRD data. Both SAED and Nanoprobe beam settings deliver reliable high quality structural data, provided that the beam and CompuStage are stable during the goniometer rotation. Varying the monochromator focus offer an additional degree of freedom for tailoring the electron dose, which is especially relevant in the Nanoprobe mode. We anticipate that the present research will contribute to the development of widely applicable routine for the structure determination of micro- and nanocrystals by cRED.

The *InsteaDMatic* script described in this article available from.

[https://github.com/stefsmeets/InsteaDMatic](https://github.com/stefsmeets/InsteaDMatic)

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## Supporting information

**Table S1**  Comparison of cRED data from Themis Z with One View camera and JEM2100F with Orius SC200D camera.

|                         | Themis Z / One View | JEOL2100F / Orius SC200D |
|-------------------------|---------------------|--------------------------|
| Spot size               | 5                   | 1                        |
| Dose rate, e/Å²/s       | 0.05                | 0.084                    |
| Diffraction area, nm    | 750                 | 1200                     |
| Tilt range, °           | 29.77 to -29.99     | -22.48 to 23.94          |
| Oscillation angle, °    | 0.424               | 0.222                    |
| Exposure time, s        | 0.30                | 0.50                     |
| Acquisition time per frame, s | 0.30          | 0.50                     |
| Camera length, mm       | 580                 | 800                      |
| Rotation speed, °/s     | 1.421               | 0.444                    |
| Rotation axis, °        | -171                | -42.5                    |
| Total No. of reflections| 12058               | 11042                    |
| No. of unique reflections| 4276              | 2086                     |
| Completeness, %         | 77.7                | 34.5                     |
| Resolution cutoff, Å    | 0.80                | 0.80                     |
| I/σ                    | 4.19                | 3.08                     |
| R_{obs}, %              | 11.0                | 23.3                     |
| R_{exp}, %              | 12.4                | 30.0                     |
| R_{meas}, %             | 13.7                | 26.1                     |
| CC_{1/2}               | 98.9                | 98.6                     |
| Unit cell parameters    |                     |                          |
| a/Å                    | 19.93               | 20.68                    |
| b/Å                    | 19.48               | 20.49                    |
| c/Å                    | 13.46               | 13.75                    |
**Table S2**  Statistics of an individual cRED dataset collected in SAED mode on Themis Z in different resolution shells.

| Resolution limit | #obs | uniq | pos comp, % | $R_{\text{obs}}$, % | $R_{\exp}$, % | #comp | $I/\sigma$ | $R_{\text{meas}}$, % | $CC_{1/2}$ |
|------------------|------|------|-------------|------------------|----------------|-------|-----------|------------------|-----------|
| 2.35             | 605  | 107  | 233 45.9    | 12.2             | 13.2           | 601   | 9.01      | 13.5             | 98.3      |
| 1.68             | 1101 | 181  | 382 47.4    | 16.0             | 14.9           | 1097  | 8.24      | 17.6             | 98.0      |
| 1.38             | 1460 | 233  | 496 47.0    | 17.0             | 16.0           | 1452  | 7.41      | 18.7             | 98.5      |
| 1.20             | 1789 | 276  | 574 48.1    | 19.2             | 19.1           | 1785  | 6.27      | 21.2             | 97.9      |
| 1.07             | 2103 | 312  | 650 48.0    | 21.8             | 22.0           | 2098  | 5.47      | 23.7             | 97.9      |
| 0.98             | 2315 | 344  | 719 47.8    | 43.8             | 52.1           | 2300  | 3.09      | 47.6             | 93.9      |
| 0.91             | 2502 | 374  | 786 47.6    | 48.0             | 72.4           | 2489  | 2.51      | 52.2             | 92.5      |
| 0.85             | 2724 | 400  | 832 48.1    | 70.5             | 117.0          | 2714  | 1.71      | 76.5             | 90.1      |
| 0.80             | 2625 | 395  | 890 44.4    | 89.3             | 173.9          | 2614  | 1.21      | 97.0             | 88.7      |
| total            | 17224| 2622 | 5562 47.1   | 20.8             | 23.9           | 17150 | 4.19      | 22.8             | 98.5      |

**Table S3**  Statistics of an individual cRED dataset collected in Nanoprobe mode on Themis Z in different resolution shells.

| Resolution limit | #obs | uniq | pos comp, % | $R_{\text{obs}}$, % | $R_{\exp}$, % | #comp | $I/\sigma$ | $R_{\text{meas}}$, % | $CC_{1/2}$ |
|------------------|------|------|-------------|------------------|----------------|-------|-----------|------------------|-----------|
| 2.36             | 653  | 112  | 234 47.9    | 13.2             | 15.6           | 651   | 9.74      | 14.6             | 98.7      |
| 1.68             | 1149 | 182  | 384 47.4    | 17.5             | 16.8           | 1146  | 8.26      | 19.2             | 97.3      |
| 1.38             | 1500 | 239  | 490 48.8    | 21.4             | 18.3           | 1495  | 7.15      | 23.5             | 95.4      |
| 1.20             | 1845 | 275  | 578 47.6    | 22.4             | 21.0           | 1843  | 6.61      | 24.8             | 98.2      |
| 1.07             | 2092 | 319  | 649 49.2    | 23.0             | 22.9           | 2079  | 5.71      | 25.1             | 98.6      |
| 0.98             | 2382 | 351  | 721 48.7    | 40.9             | 55.0           | 2376  | 3.48      | 44.8             | 94.5      |
| 0.91             | 2600 | 380  | 789 48.2    | 44.8             | 69.2           | 2595  | 3.10      | 49.1             | 90.3      |
| 0.85             | 2782 | 409  | 829 49.3    | 57.6             | 101.3          | 2767  | 2.25      | 62.9             | 90.7      |
| 0.80             | 2822 | 425  | 899 47.3    | 65.2             | 144.0          | 2810  | 1.49      | 71.1             | 93.7      |
| total            | 17825| 2692 | 5573 48.3   | 21.7             | 24.8           | 17762 | 4.42      | 23.9             | 98.1      |
Table S4  
Statistics of SAED dataset merged from 5 crystals in different resolution shells.

| Resolution limit | #obs | #uniq | #pos | comp, % | $R_{\text{obs}}$, % | $R_{\text{exp}}$, % | #comp | $I/\sigma$ | $R_{\text{meas}}$, % | $CC_{1/2}$, % |
|------------------|------|-------|------|---------|---------------------|---------------------|-------|------------|----------------------|-----------------|
| 3.58             | 571  | 61    | 67   | 91.0    | 21.8                | 27.3                | 571   | 7.64       | 23.4                 | 94.2            |
| 2.53             | 1051 | 104   | 110  | 94.5    | 25.4                | 27.1                | 1051  | 7.35       | 26.7                 | 95.5            |
| 2.07             | 1479 | 143   | 150  | 95.3    | 29.5                | 27.7                | 1479  | 6.91       | 31.2                 | 97.4            |
| 1.79             | 1796 | 164   | 175  | 93.7    | 23.7                | 27.7                | 1796  | 6.77       | 25.1                 | 98.2            |
| 1.60             | 1927 | 179   | 189  | 94.7    | 32.6                | 28.6                | 1927  | 6.06       | 34.6                 | 88.9            |
| 1.46             | 2228 | 201   | 208  | 96.6    | 32.7                | 30.5                | 2226  | 5.71       | 34.3                 | 98.4            |
| 1.35             | 2641 | 233   | 241  | 96.7    | 34.1                | 29.9                | 2640  | 5.22       | 36.0                 | 94.3            |
| 1.26             | 2755 | 238   | 244  | 97.5    | 50.8                | 33.7                | 2752  | 4.99       | 53.5                 | 81.3            |
| 1.19             | 2995 | 256   | 267  | 95.9    | 34.2                | 33.0                | 2994  | 5.15       | 36.1                 | 93.8            |
| 1.13             | 3165 | 269   | 277  | 97.1    | 40.7                | 35.2                | 3163  | 4.85       | 42.7                 | 93.4            |
| 1.08             | 3373 | 278   | 289  | 96.2    | 48.2                | 43.5                | 3370  | 4.07       | 50.8                 | 90.9            |
| 0.99             | 3883 | 304   | 318  | 95.6    | 93.8                | 101.8               | 3881  | 2.82       | 98.0                 | 89.1            |
| 0.92             | 4093 | 327   | 334  | 97.9    | 113.9               | 131.4               | 4089  | 2.45       | 119.2                | 86.6            |
| 0.89             | 4500 | 350   | 366  | 95.6    | 134.7               | 160.6               | 4498  | 2.30       | 140.6                | 86.2            |
| 0.84             | 4823 | 371   | 380  | 97.6    | 251.8               | 337.5               | 4819  | 1.74       | 262.5                | 61.4            |
| 0.82             | 4732 | 366   | 381  | 96.1    | 296.4               | 405.5               | 4729  | 1.53       | 309.0                | 55.8            |
| 0.80             | 3357 | 337   | 390  | 86.4    | 255.0               | 394.5               | 3347  | 1.35       | 267.6                | 69.2            |
| total            | 61596| 5159  | 5386 | 95.8    | 33.0                | 33.8                | 61551 | 3.56       | 34.9                 | 94.8            |
Table S5  Statistics of Nanoprobe dataset merged from 6 crystals in different resolution shells.

| Resolution limit | #obs | #uniq | #pos | comp, % | \(R_{\text{obs}}\), % | \(R_{\text{exp}}\), % | #comp | \(I/\sigma\) | \(R_{\text{meas}}\), % | \(CC_{1/2}\) |
|------------------|------|-------|------|---------|----------------|----------------|-------|-------------|----------------|-------------|
| 3.58             | 705  | 67    | 68   | 98.5    | 20.0           | 23.6           | 705   | 9.53        | 21.2           | 98.0        |
| 2.53             | 1302 | 112   | 112  | 100.0   | 17.0           | 24.0           | 1302  | 9.00        | 17.9           | 98.5        |
| 2.07             | 1775 | 146   | 146  | 100.0   | 19.6           | 24.5           | 1775  | 8.59        | 20.6           | 97.7        |
| 1.79             | 2188 | 175   | 175  | 100.0   | 22.3           | 24.8           | 2188  | 7.81        | 23.6           | 97.5        |
| 1.60             | 2507 | 196   | 196  | 100.0   | 21.4           | 25.9           | 2507  | 7.44        | 22.5           | 98.5        |
| 1.46             | 2690 | 203   | 205  | 99.0    | 29.8           | 27.4           | 2690  | 6.98        | 31.2           | 99.6        |
| 1.35             | 3066 | 232   | 233  | 99.6    | 23.4           | 26.5           | 3066  | 6.84        | 24.6           | 84.3        |
| 1.27             | 3422 | 255   | 255  | 100.0   | 28.2           | 29.5           | 3422  | 6.13        | 29.5           | 98.9        |
| 1.19             | 3528 | 258   | 258  | 100.0   | 26.2           | 29.6           | 3528  | 6.38        | 27.4           | 98.8        |
| 1.13             | 3881 | 287   | 288  | 99.7    | 27.7           | 30.7           | 3881  | 5.83        | 28.9           | 96.6        |
| 1.08             | 4016 | 283   | 285  | 99.3    | 38.8           | 39.1           | 4016  | 5.22        | 40.4           | 98.8        |
| 0.99             | 3804 | 324   | 325  | 99.7    | 53.3           | 51.6           | 3804  | 3.73        | 55.9           | 95.3        |
| 0.92             | 4069 | 341   | 341  | 100.0   | 54.4           | 56.9           | 4069  | 3.41        | 57.1           | 92.7        |
| 0.89             | 4268 | 358   | 358  | 100.0   | 67.0           | 65.6           | 4268  | 2.89        | 70.1           | 92.3        |
| 0.84             | 4731 | 395   | 395  | 100.0   | 90.4           | 103.9          | 4731  | 2.27        | 94.7           | 84.3        |
| 0.82             | 4490 | 383   | 388  | 98.7    | 91.6           | 114.8          | 4490  | 2.18        | 95.9           | 95.7        |
| 0.80             | 3234 | 310   | 394  | 78.7    | 91.8           | 112.8          | 3227  | 1.83        | 96.6           | 83.7        |
| total            | 65672| 5299  | 5397 | 98.2    | 24.0           | 27.7           | 65665 | 4.56        | 25.2           | 97.4        |
Table S6  Deviations of atomic positions between the reference ZSM-5 structure (van Koningsveld et al., 1987) and those determined from cRED data collected in SAED/NP modes. Fractional atomic coordinates for the reference ZSM-5 structure are given in Table S7. The origin shift between reference and refined structures can be expressed by a transformation matrix \((P, p)\): \(a, b, c \); \(1/2, 0, 1/2\).

| Atom label | Atomic displacement SAED, Å | Atomic displacement NP, Å |
|------------|-----------------------------|---------------------------|
| Si1        | 0.0467                      | 0.0295                    |
| Si2        | 0.0859                      | 0.0474                    |
| Si3        | 0.0373                      | 0.0243                    |
| Si4        | 0.0294                      | 0.0242                    |
| Si5        | 0.0146                      | 0.0234                    |
| Si6        | 0.0521                      | 0.0153                    |
| Si7        | 0.0230                      | 0.0076                    |
| Si8        | 0.0394                      | 0.0265                    |
| Si9        | 0.0413                      | 0.0429                    |
| Si10       | 0.0552                      | 0.0509                    |
| Si11       | 0.0492                      | 0.0316                    |
| Si12       | 0.0541                      | 0.0144                    |
| O1         | 0.0735                      | 0.0970                    |
| O2         | 0.1003                      | 0.0350                    |
| O3         | 0.0134                      | 0.0247                    |
| O4         | 0.0581                      | 0.0509                    |
| O5         | 0.0307                      | 0.0222                    |
| O6         | 0.0423                      | 0.0289                    |
| O7         | 0.0515                      | 0.0333                    |
| O8         | 0.0430                      | 0.0329                    |
| O9         | 0.0166                      | 0.0464                    |
| O10        | 0.1547                      | 0.0933                    |
| O11        | 0.0413                      | 0.0391                    |
| O12        | 0.1019                      | 0.1142                    |
| O13        | 0.1135                      | 0.1111                    |
| O14        | 0.1061                      | 0.0297                    |
| O15        | 0.0427                      | 0.0570                    |
| O16        | 0.0305                      | 0.0190                    |
| O17        | 0.0564                      | 0.0364                    |
| O18        | 0.0486                      | 0.0754                    |
|   | O19   | O20   | O21   | O22   | O23   | O24   | O25   | O26   |
|---|-------|-------|-------|-------|-------|-------|-------|-------|
|   | 0.0402| 0.0186| 0.0576| 0.0441| 0.0816| 0.0432| 0.0625| 0.1513|
| <Si> average | 0.04(2) | 0.03(1) |
| <O> average   | 0.06(4) | 0.05(3) |
Table S7  Fractional atomic coordinates for the reference ZSM-5 structure (van Koningsveld et al., 1987).

|    | x     | y     | z     |
|----|-------|-------|-------|
| Si1| 0.4224| 0.0565| -0.3360|
| Si2| 0.3072| 0.0277| -0.1893|
| Si3| 0.2791| 0.0613| 0.0312 |
| Si4| 0.1221| 0.0630| 0.0267 |
| Si5| 0.0713| 0.0272| -0.1855|
| Si6| 0.1864| 0.0590| -0.3282|
| Si7| 0.4227| -0.1725| -0.3272|
| Si8| 0.3078| -0.1302| -0.1855|
| Si9| 0.2755| -0.1728| 0.0311 |
| Si10| 0.1206| -0.1731| 0.0298 |
| Si11| 0.0704| -0.1304| -0.1820|
| Si12| 0.1871| -0.1733| -0.3193|
| O1 | 0.3726| 0.0534| -0.2442|
| O2 | 0.3084| 0.0587| -0.0789|
| O3 | 0.2007| 0.0592| 0.0289 |
| O4 | 0.0969| 0.0611| -0.0856|
| O5 | 0.1149| 0.0541| -0.2763|
| O6 | 0.2435| 0.0553| -0.2460|
| O7 | 0.3742| -0.1561| -0.2372|
| O8 | 0.3085| -0.1552| -0.0728|
| O9 | 0.1980| -0.1554| 0.0288 |
| O10| 0.0910| -0.1614| -0.0777|
| O11| 0.1169| -0.1578| -0.2694|
| O12| 0.2448| -0.1594| -0.2422|
| O13| 0.3047| -0.0510| -0.1866|
| O14| 0.0768| -0.0519| -0.1769|
| O15| 0.4161| 0.1276| -0.3896|
| O16| 0.4086| -0.0017| -0.4136|
| O17| 0.4020| -0.1314| -0.4239|
| O18| 0.1886| 0.1298| -0.3836|
| O19| 0.1940| 0.0007| -0.4082|
| O20| 0.1951| -0.1291| -0.4190|
| O21 | 0.0037 | 0.0502 | −0.2080 |
| O22 | −0.0040 | −0.1528 | −0.2078 |
| O23 | 0.4192 | −0.2500 | −0.3540 |
| O24 | 0.1884 | −0.2500 | −0.3538 |
| O25 | 0.2883 | −0.2500 | 0.0579 |
| O26 | 0.1085 | −0.2500 | 0.0611 |
**Figure S1** Typical diffraction patterns of ZSM-5 collected on Themis Z at different electron dose rates. Dotted rings indicate 0.8 Å resolution.
Supporting movie S1
https://stockholmuniversity.box.com/s/96cfu9kspo4voluzynz092o47cpj0722

Supporting movie S2
https://stockholmuniversity.box.com/s/f161l0mb015o97d5qurdsharahpcpfzz