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SISYPHOS: An automatic procedure for the serial refinement of single-crystal diffraction data in Olex2

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ABSTRACT

A program for serial handling of crystallographic data is presented within Olex2. Especially for small molecule electron and x-ray diffraction, the handling of several datasets of the same structure can be tedious and prone to errors, which can affect comparability. The program SISYPHOS allows for the individual refinement of a starting model against several recorded datasets (in ".hkl" format) with adaptation to changes in the unit cell, wavelength, among other parameters. The program was tested for resonant diffraction (also known as anomalous dispersion), investigations on radiation damage, the benchmarking of different configurations for quantum crystallographic modeling, electron diffraction data, and for testing several datasets from the same measurement using various settings to identify the most suitable dataset.

INTRODUCTION

With the steady improvement of x-ray diffraction devices, small molecule crystallography is no longer heavily dependent on measurement time but instead has an abundance of data. Especially with the latest innovations of rotating and metal-jet anode devices,[1] using photon-counting detectors,[2,3] as well as the recent introduction of the first commercial electron diffractometers,[4,5] the time for data collection is significantly reduced. At the same time, the improved datasets reveal more of the imperfections of the average crystal structure model. These include disorder, anharmonicity, absorption effects, and others. Often, several datasets of the same compound are collected—either at different wavelengths[6] or from different crystals to be able to find the best crystallographic model of the compound at hand.

When complete datasets are collected in minutes rather than formerly days or even weeks, an automatic procedure becomes feasible and maybe even necessary. Especially for chemical crystallography in synchrotron facilities, it is common to record multiple datasets—either for obtaining a complete dataset despite angular restrictions, radiation damage, varying wavelengths (e.g., resonant diffraction), or at different temperatures and pressure conditions.

This, however, leads to the problem that less time is spent on the modeling and refinement per structure model, increasing "avoidable" errors in published structures. As recently described in a review by Raymond and Girolami,[7] who categorize the different origins of "pathological crystal structures." According to the authors, there are causes intrinsic to the method of crystallography, such that the time spent per crystal structure has an impact. Thomson has already reported that the number of publications with multiple datasets of the same structure has increased significantly since the turn of the millennium, and has developed CX-ASAP, a high-throughput tool for the serial refinement and analysis of crystallography data collected under different conditions.[8] However, the program relies on the ShelXL[9] refinement engine and is therefore limited to its functionality regarding non-spherical atomic form factors,[10,11] the modeling of anharmonic motion,[12,13] and anomalous dispersion.[14]

In this work, the software module SISYPHOS, as part of the crystallographic software suite Olex2,[15] is introduced. SISYPHOS aims to make the refinement of different datasets of the same or very similar crystal structures easily accessible and convenient to a wide range of crystallographers. An overview of the fields in which

the software can be applied can be found in Fig. 1. The use cases include crystal checking, multitemperature and multi-pressure series, quantum crystallographic benchmarking, analysis of radiation damage, and resonant diffraction experiments. The general principle is to refine the same crystallographic starting model either against several datasets of the same structure or with a series of different settings against the same dataset, or both. Direct communication with the established quantum crystallographic software NoSpherA2[16] enables serial use of quantum crystallography for a variety of scenarios. SISYPHOS collects resulting data and model parameters, alongside refinement indicators, and provides the user with a formatted .csv file for easy comparison of the different models. Each generated model is stored in a separate location for further investigation. Fully written in Python 3,[17] SISYPHOS can be adjusted for user-specific applications. Especially with the advent of high-level, commercial electron diffractometers, the program offers a pathway for a smoother and optimized structure elucidation process.

IMPLEMENTATION

Architecture and workflow

In the following, the typical workflow (see Fig. 2) within SISYPHOS is explained. The program searches for all uniquely named *.hkl* files in the chosen data directory recursively and creates a new directory in the parent output directory for each structure, copying any identically named *.ins* files for later reference, and the template model *.ins* along with the *.hkl* file into it. Differences in unit cells, wavelength, temperature, etc., are read from the respective *.ins* file. The program automatically ignores *.hkl* files within its output directory and increments output folder names with integer numbers to enable multiple runs on the same dataset. SISYPHOS proceeds to refine each structure in its new directory, leaving all old models unchanged. It checks automatically if extinction correction is required and compares the extinction factor to its standard uncertainty, omitting the extinction correction if the value is smaller than 3σ. During the refinement, the different modes are applied individually. For anomalous dispersion refinement,[18] the corresponding parameters for all atoms of one or many element types as selected during the setup are included in the least squares refinement of olex2.refine. If NoSpherA2 is used, the settings for the quantum chemical calculation, currently only supporting ORCA,[19–22] can either be set from the NoSpherA2 GUI or a benchmark file ("Benchmark Mode").

Once the refinement is complete, the information extraction process begins. This involves built-in Olex2 functions as well as parsing of the resulting *.cif*. SISYPHOS to extract all bond lengths and uncertainty from the model, using the variance–covariance matrix. Where defined, uncertainties of derived values are collected as well. All results are first appended to a .txt output file, which is subsequently converted into a .csv file for convenient data handling and visualization.

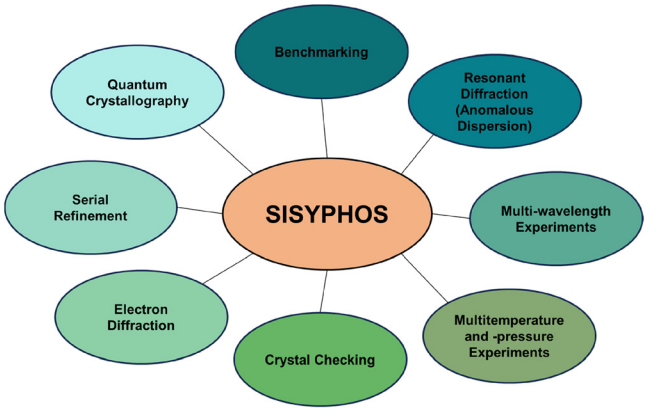

FIG. 1. Overview of the different areas of application for SISYPHOS.

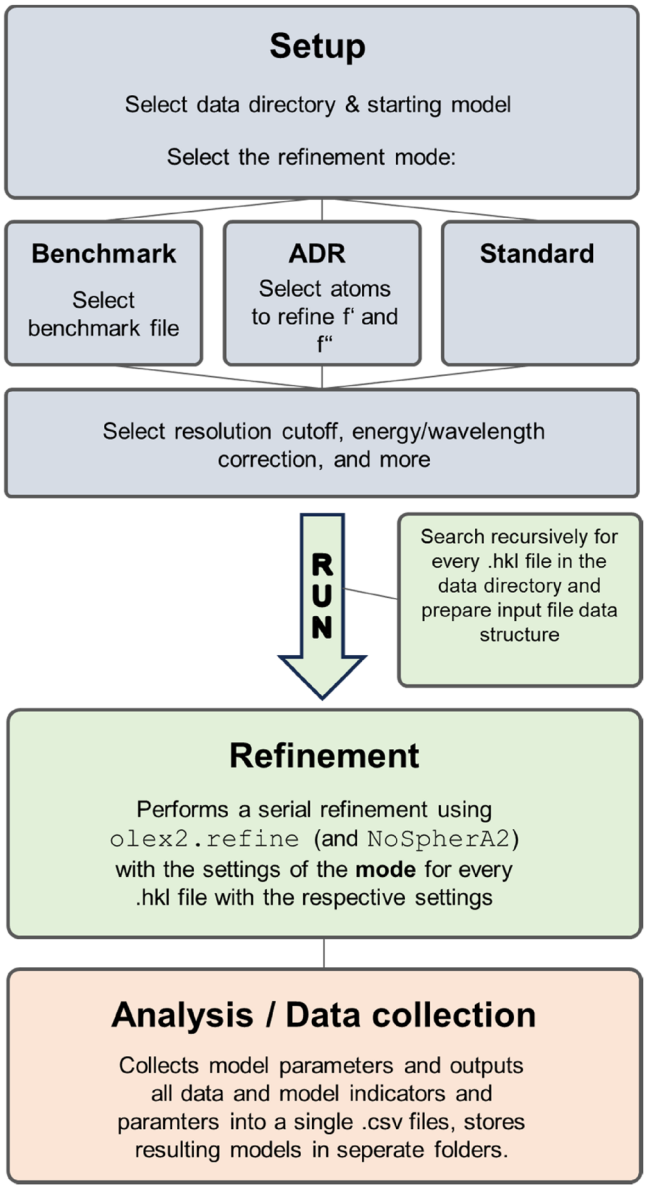

FIG. 2. Schematic workflow with options in SISYPHOS.

User interface

Figure 3 shows the graphical user interface of SISYPHOS as implemented in Olex2. The blue button labeled "DIR" allows the user to select the main directory for the data. The routine will search through all subfolders for uniquely named *.hkl* files (including *.hklf4* and *.hklf5* files). On the right side of the directory textfield, the *.ins* button allows the user to select the starting model against which each dataset is refined. By default, it will open the current structure folder. The settings in the following rows depend on the selected mode, which are changeable using the tickboxes in the row labeled "Mode options." By default, the user can apply corrections to the incoming x-ray energy and restrict the outer resolution limit for all datasets, which is helpful for comparison purposes, for example, where datasets show different maximum resolution, but should be refined using the same cutoff.

In "Use NoSpherA2" mode, a Hirshfeld Atom Refinement[10,23] will be performed on each dataset, using the settings as shown in the GUI that is imported from the NoSpherA2 software. In combination with the benchmark mode, the user can select a text file containing a dictionary of each combination of settings for the HAR procedure they want to perform on the structure. An example of the layout of this benchmark file is given in the program folder. Finally, the user can perform a refinement of anomalous dispersion parameters by enabling the mode "DISP refinement." In an additional line of the GUI, the element (or elements, white space separated) can be entered. Additionally, if there is more than one position for the selected element, the values for anomalous dispersion can be constrained to be equal or individual, by the checkbox "Individual DISP." For comparison, the structures can be refined

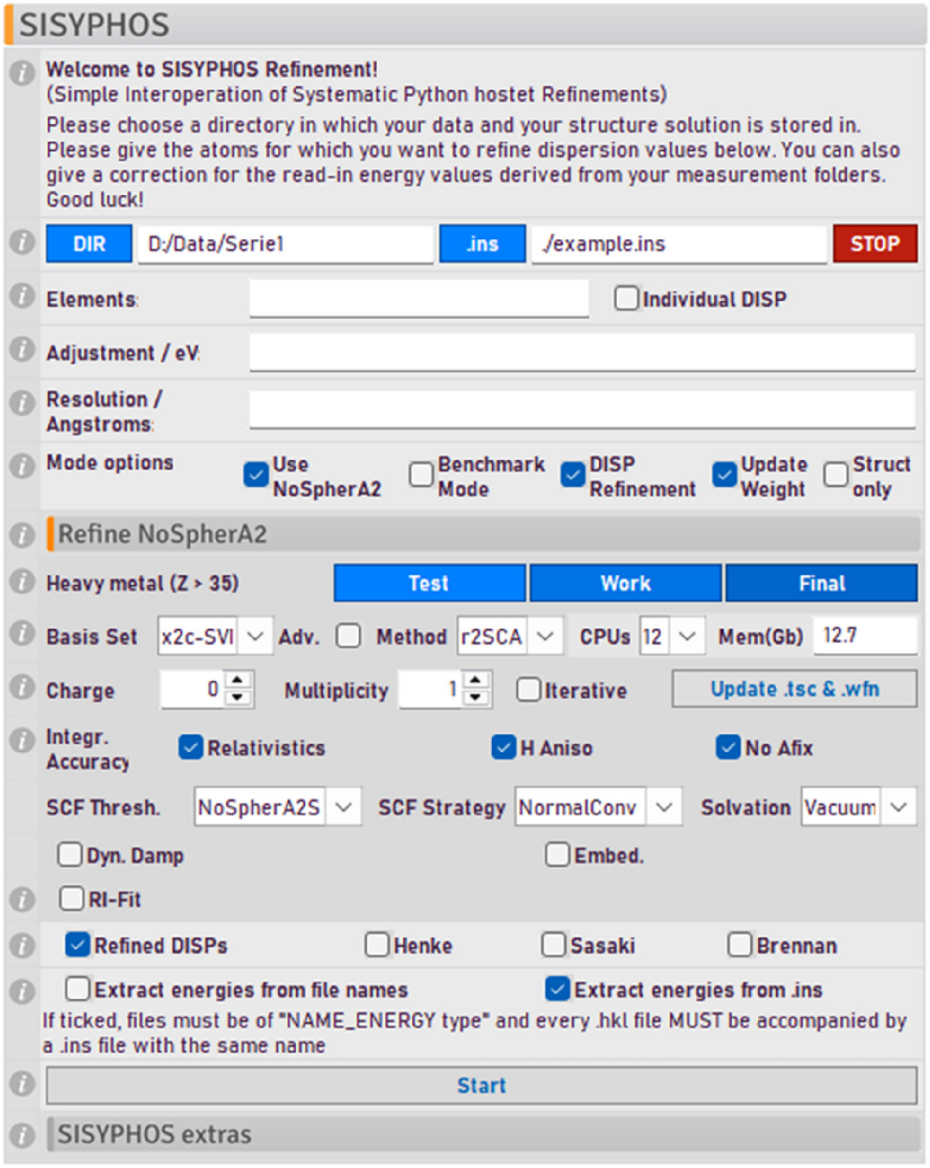

FIG. 3. Graphical user interface of SISYPHOS in Olex2 showing settings for a refinement of anomalous dispersion parameters while employing a Hirshfeld Atom Refinement within NoSpherA2 using ORCA.

using the tabulated values according to Henke *et al.*,[24] Sasaki,[25] or Brennan and Cowan.[26]

Additionally, the user is able to only include CrysalisPro[27] style "struct" folders for the search of datasets, and, therefore, exclude datasets within the main experiment folder, or whether the weighting scheme used by the refinement should be updated ("Update Weight"). Finally, the user can tell the program where to find the correct energy. By default, the x-ray energy from the template *.ins* file is used, but alternatively, an *.ins* file corresponding to each dataset can be used to ensure the correct energy matches the model. Additionally, for synchrotron applications, it is possible to read the energy from the dataset name if it adheres to the naming scheme of "NAME_ENERG.hkl," where NAME can be anything and ENERG is the energy in integer eV. The gray "Start" button at the bottom will start the serial refinement, which can be stopped using the red "STOP" button.

APPLICATIONS AND DISCUSSION

Crystal screening

A possible scenario for crystal screening on different x-ray diffractometers would be to measure some visually selected crystals on various devices. The aim is to find the best combination of diffractometer and wavelength for each crystal. To emulate this scenario, we measured a total of three different sucrose crystals at various x-ray sources: a Rigaku Oxford Diffraction SuperNova equipped with an Ag Kα source, a Rigaku Oxford Diffraction SuperNova equipped with Cu Kα and Cu Kβ sources, and a Rigaku Oxford Diffraction Synergy DW with Cu Kα and Mo Kα rotating anode sources.

Figure 4 compares the R1, wR2, Rint, I/σ(I), cell volume, and unique reflections for a total of eleven different measurements of two different sucrose single crystals at the in-house diffractometers of the University of Regensburg.

Multitemperature experiments

To demonstrate the application of SISYPHOS for serial measurements, we investigated the well-known YLID[6,28] structure at temperatures ranging from 250 to 110 K in 20 K increments as a small test case. Figure 5 displays selected parameters of the crystal structures as generated by CrysAlisPro automatic data reduction routine. As expected, quality indicators exhibit a negative correlation with temperature, showing lower residuals at lower temperatures due to reduced atomic motion.

For the same reason, the signal-to-noise ratio increases with lower temperatures. All three cell dimensions increase with rising temperature, also resulting in more unique reflections based on the larger cell volume at a fixed wavelength. The steady R_{int} statistics indicate a low dependency of the agreement of systematically equivalent reflections at different temperatures.

For this dataset, all data processing and refinement steps, except for the initial model, were automated using CrysAlisPro (data processing, reduction, and multi-scan absorption correction) and SISYPHOS (model refinement and information extraction). This way, the required manual software interaction time for creating this data series was drastically reduced, allowing the processing to be performed unsupervised. The last refinement shift in the output .csv file also gives an overview of model convergence.

Anomalous dispersion refinements

Resonant single-crystal diffraction is a powerful technique that allows probing spatially resolved x-ray absorption spectra of crystalline materials. The chemical applications as well as the crystallographic implications can be found in a variety of publications by the Betley and Chen groups, who pioneered this field for the single-crystal community using data from the APS synchrotron facility.[31–33] We recently reported on the implementation of ADR in Olex2 and the effect of using tabulated values for AD in the vicinity of absorption edges,[18] as well as structural improvements by ADR for heavy element structures.[34,35]

SISYPHOS can be used to efficiently evaluate resonant diffraction data, e.g., from a serial synchrotron experiment. For example, see Refs. 18 and 34, where the program was used to efficiently analyze a large amount of synchrotron datasets measured at the Rossendorf Beamline at the ESRF.[36]

Quantum crystallographic benchmarking

Quantum Crystallography is at the forefront of analyzing and understanding diffraction data.[30] The independent atom model has been the default model for structural refinement over the last 50 years.[37] With the advance of new x-ray generation and detection technologies, its inadequacies become more and more obvious. With NoSpherA2, there is already a powerful tool to make the Hirshfeld Atom Refinement available for daily use, and, by being able to employ the state-of-the-art quantum chemical program ORCA,[19–22] it can be performed faster than ever. SISYPHOS fully supports the use of NoSpherA2, to enable efficient serial quantum crystallographic analyses.

Using the example of the Olex2 shipped structure "SuperSugar," the application of SISYPHOS for quantum crystallographic benchmarking is evaluated. For this, a single structure is chosen, alongside the "Benchmark file," which contains the desired settings for the quantum chemical calculation conducted by NoSpherA2 and the ORCA software suite. For this assessment, the Hartree–Fock (HF) method was compared to density functionals of various levels of accuracy on Jacobs ladder: PBE, PBE0, B3LYP, and r2SCAN.[38–40] They were applied during an HAR procedure within NoSpherA2, employing ORCA5 for the underlying quantum chemical calculation using either only the split-valence or the triple-zeta Ahlrichs basis sets def2-SVP or def2-TZVP, respectively.[41] For comparison, SISYPHOS also generates the results from the independent atom model (IAM). This allows for convenient benchmarking of the shown (see Sec. S3 for more details) settings options for performing a HAR.

Figure 6 shows selected model indicators for each of the IAM and HAR models of the "SuperSugar" sucrose dataset.

As expected, every HAR model surpassed the quality indicators of the IAM. The differences between the HAR models show a clear preference for the triple-zeta basis sets compared to the split-valence ones. For functionals, the meta-GGA r2SCAN performed best in terms of R1, wR2, and negative residual density peaks, while, surprisingly, HF, together with B3LYP, had the lowest positive residual electron density.

A more thorough testing of the various settings for HAR is explored in another paper within this special issue.[42]

In summary, SISYPHOS was used to successively refine several thousand models by varying the level of theory underlying the

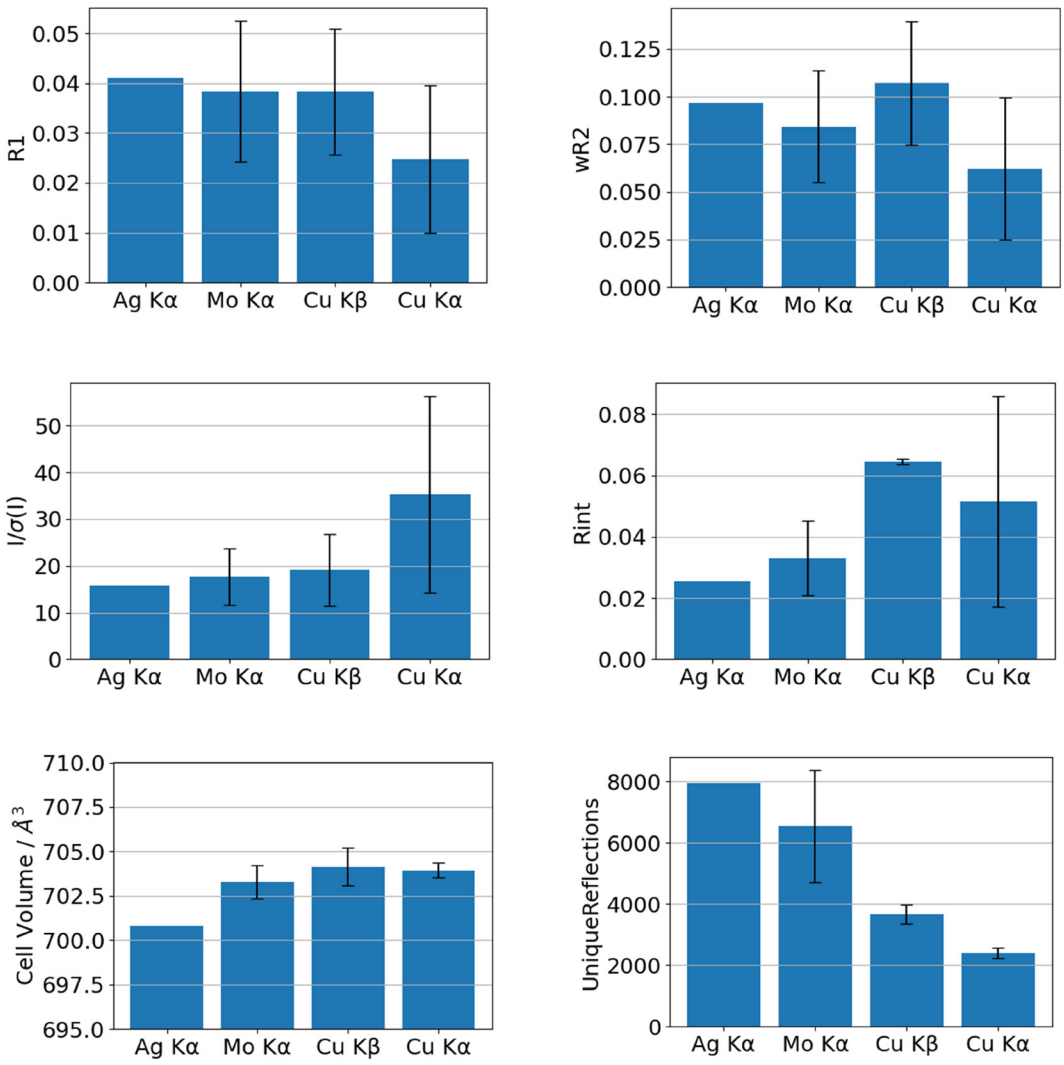

FIG. 4. Bar diagrams of eleven datasets and crystallographic model indicators as average values for each wavelength, and, where applicable, one standard deviation of two sucrose single crystals measured on different devices of the central analytics x-ray department of the University of Regensburg. Device configuration and further information for each wavelength can be found in Sec. S4 of the supplementary material.

quantum chemical calculations. This approach allowed them to obtain information about the performance of the various NoSpherA2 settings, as illustrated in Fig. 6. The authors discussed trends regarding the different parameters and noted that even DFT methods that do not belong to the highest class of functionals, such as the aforementioned R2SCAN, can produce decent refinement outcomes. The refinement results were then evaluated using classical crystallographic indicators such as R1 and wR2, as well as the position of hydrogen atoms relative to neutron reference structures. A SISYPHOS run provides the necessary parameters for all of this, enabling the results to be extracted using lightweight Python scripts. As executing several thousand refinements successively in a reasonable time frame on a single computer is hardly achievable, a SISYPHOS run can also be parallelized using subprocesses and a headless version of Olex2.Therefore, an initial generated set of method files from predefined parameter sets can be distributed on independent Olex2 headless executions across multiple CPU cores. This procedure can also be transferred to the cluster capacities offered by high-performance computing facilities. By distributing the method files across an array of jobs via a batch script, automated, high-throughput exploration of settings can be enabled with SISYPHOS.

Mixed-position structures

Our final example is the application of SISYPHOS to a crystallization study. The group VI hexacarbonyls $M(CO)_6$ (M = Cr, Mo, W) can be co-crystallized from a stochiometric solution mixture of two or all three of them. To statistically evaluate the reproducibility of this crystallization method, eight crystals of a 1:1 stoichiometric mixture of $Cr(CO)_6$ and $W(CO)_6$ were measured using a Mo Kα rotating anode and modeled using SISYPHOS with an EXYZ and EADP constrained central metal

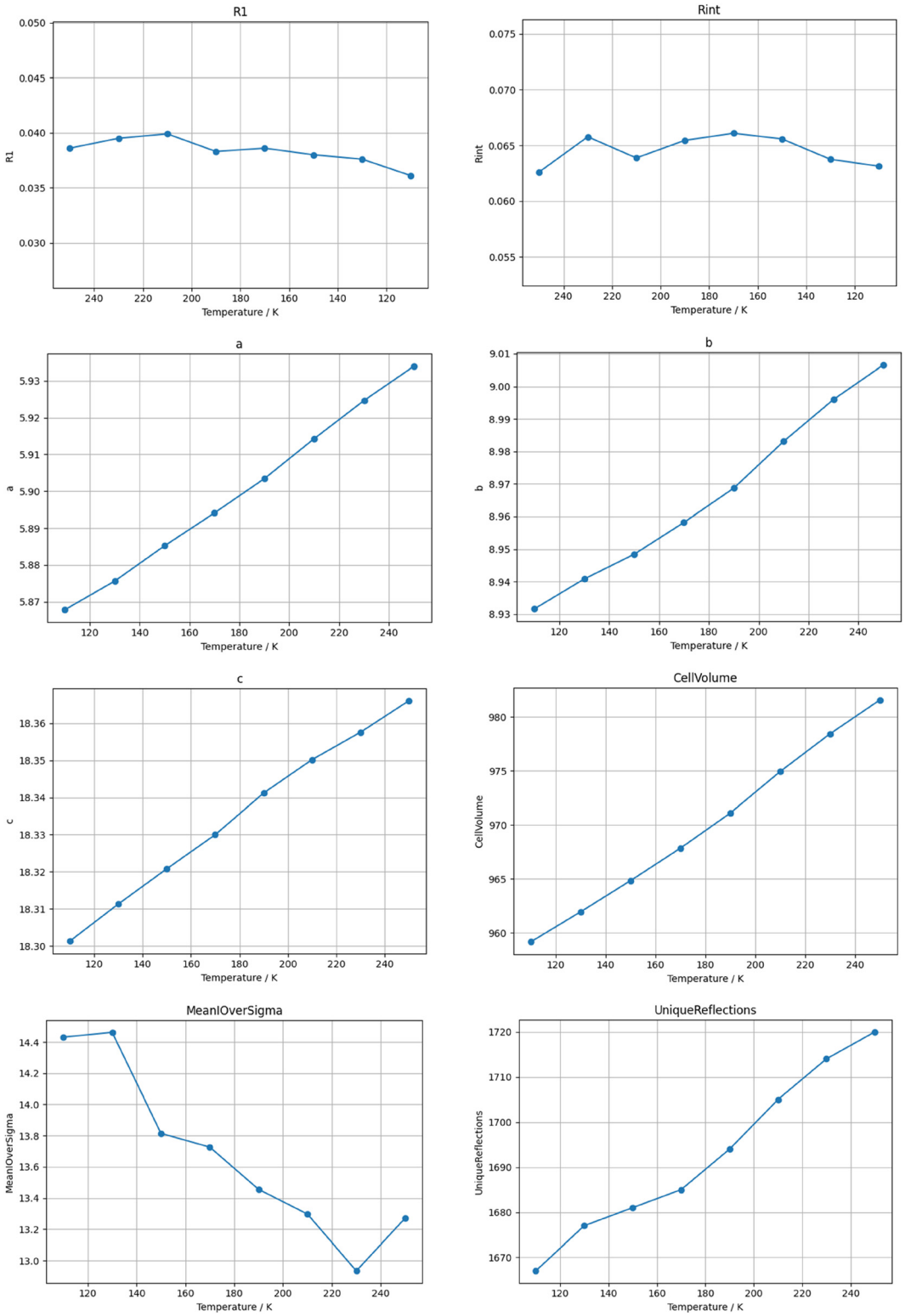

FIG. 5. Selected data for the multi-temperature run of YLID from the SISYPHOS output.csv file. Plots were created by a simple Python script employing the Pandas[29] and Matplotlib[30] modules.

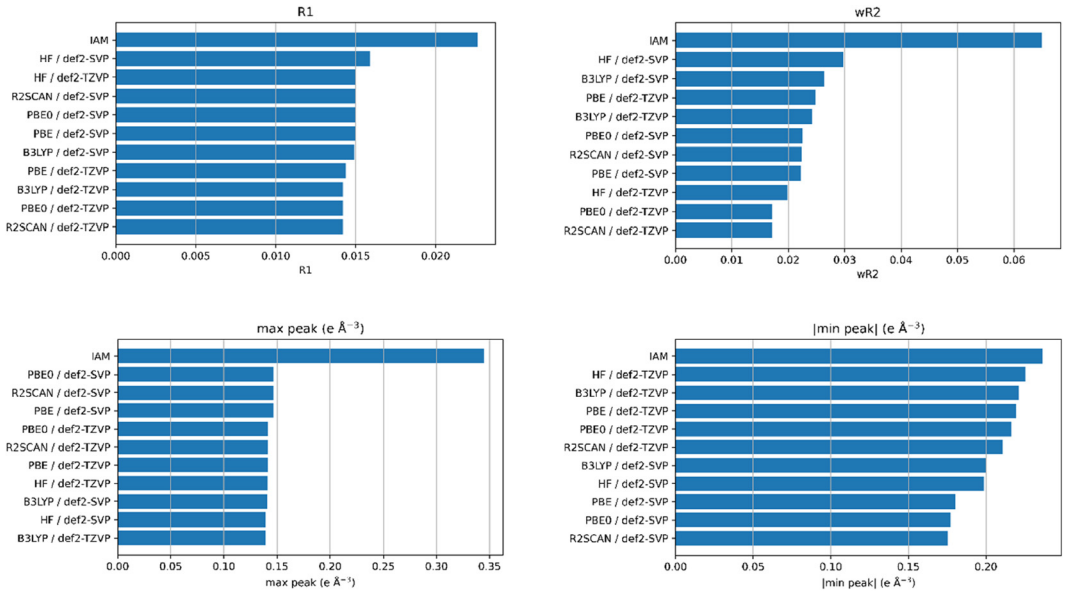

FIG. 6. Quality indicators of the IAM and HAR models created by SISYPHOS employing ORCA and NoSpherA2 on the Olex2 internal "SuperSugar" dataset.

position and freely refined occupancy of the metal position, constrained to add up to 1. Table I shows the results of these datasets, making it easy to spot outliers (such as Nr. 6) and calculate statistical indicators for larger datasets (such as mean, median, accuracy).

CONCLUSION

In this work, we introduced the Olex2 plugin SISYPHOS and demonstrated the applications it was designed to address. SISYPHOS can be used to refine a structural model against several datasets of the same structure or refine different modeling approaches for the same dataset, collecting the model and data information in the .csv format. The applications for crystal screening, benchmarking quantum crystallography, anomalous dispersion refinements, multitemperature studies, as well as crystallization studies were described.

TABLE I. Agreement statistic R1 of measured and modeled structure factors and the refined occupancies [Occu (Cr/W)] for Cr and W at the central metal position. Nr. 9 and Nr. 8 are data from the same crystal (with 10 and 7, respectively), using different data processing strategies.

Nr	R1	Occu(Cr)	Occu(W)
1	0.0135	0.495	0.505
2	0.0195	0.496	0.504
3	0.0124	0.467	0.533
4	0.0192	0.466	0.534
5	0.0231	0.454	0.546
6	0.0697	0.448	0.552
7	0.0128	0.472	0.528
8	0.0127	0.472	0.528
9	0.0161	0.504	0.496
10	0.0166	0.504	0.496
Avg.	0.0216	0.4778	0.5222
SD	0.0164	0.0194	0.0194

SUPPLEMENTARY MATERIAL

See the supplementary material for crystallographic data tables corresponding to the sections on crystal screening, multitemperature experiments, quantum crystallographic benchmarking, and mixed metal position along with additional information regarding the experimental conditions. SISYPHOS is available as an extension module within Olex2, or via GitHub at the address https://github.com/FlorianMeurer/plugin-SISYPHOS. All output files and crystallographic models used in this work are available via zenodo.org with DOI 10.5281/zenodo.17151558.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Florian Meurer: Conceptualization (equal); Data curation (equal); Project administration (equal); Software (equal); Validation (equal); Visualization (equal); Writing – original draft (equal). **Florian Kleemiss:** Software (supporting); Writing – review & editing (equal).

Birgit Hischa: Data curation (equal); Investigation (supporting). **Daniel Brüx:** Software (supporting); Writing – review & editing (equal). **Ann-Cathrin Koch:** Data curation (equal); Formal analysis (supporting); Investigation (supporting). **Michael Bodensteiner:** Conceptualization (equal); Project administration (equal); Supervision (equal); Writing – review & editing (equal).

DATA AVAILABILITY

The data that support the findings of this study are openly available in Zenodo at 10.5281/zenodo.17151558.

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