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CrystalPlan: an Experiment Planning Tool for Crystallography

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Beam time at large user program based x-ray and neutron scattering facilities is in high demand and always at a premium. CrystalPlan, a highly efficient experiment planning software has been developed to maximize the use of available beamtime per sample per experiment. This program can calculate and optimize the data coverage of a crystal in reciprocal space in a single-crystal diffraction time-of-flight experiment. CrystalPlan can help a user build an experiment plan that will acquire the most unique data possible, with sufficient coverage but limited redundancy, therefore increasing scientific productivity. A user friendly GUI (Graphical User Interface) including a 3D viewer, an automated coverage optimizer, and an option to reorient the crystal for the measurement of selected *hkl*s on specific detector positions are among its useful features. A sample use case of the program with the TOPAZ beamline at SNS will be presented.

1. Introduction

The TOPAZ beamline at the Spallation Neutron Source (SNS) at Oak Ridge National Laboratory (ORNL), currently finishing commissioning, is a neutron time-of-flight Laue diffractometer with high wavelength (time) resolution. It is designed to acquire single-crystal diffraction data of small-to-moderate sized unit cells with high throughput. The final design calls for an array of 48 detectors covering three quarters of 2π steradians. However as of this writing, only 14 of the 48 detectors are installed, giving only a quarter of 2π steradian coverage. To have sufficient data for a reliable structure solution and refinement, an experimenter will typically want to measure $> 85\%$ of the unique reflections within a certain range of d-spacings. However, TOPAZ's wide wavelength band combined with its complex geometry makes it difficult to come up with an effective set of sample orientations that will cover all the required reflections without unnecessary redundancy.

Existing diffraction experiment planning software programs (for example, CrysAlis Pro from Oxford Diffraction; COSMO from Bruker AXS) were designed for X-ray experiments where the crystal is rotated stepwise through the Bragg diffraction condition using a single incident wavelength, resulting in a large number of sample orientations. These programs are not suitable for TOPAZ, since as a time-of-flight instrument (time-resolved Laue), it is collecting data from a wide bandwidth of incident neutrons, thus contracting the Ewald sphere through the reflection condition while keeping the crystal stationary, as described in (Schultz, 1994; Wilson, 2000). Additionally, this means that relatively few sample orientations are needed to complete a measurement: whereas an X-ray measurement might have hundreds or thousands of individual frames, a TOPAZ run may have as few as 10 to cover the same volume of reciprocal space.

The goal of CrystalPlan was to make an easy-to-use software tool to aid users in planning their experiments. By quickly making an experimental plan that will be tailored to the needs of the scientist (enough reflections measured for a structure analysis), but without spending time collecting redundant data, CrystalPlan will greatly increase the scientific productivity and throughput of TOPAZ and other similar beamlines. Users will be able to measure samples highly efficiently, allowing them to explore parameters such as temperature and pressure more fully.

2. Architecture

CrystalPlan is designed with these capabilities:

- Load a geometry file for a specific instrument (using a generic, flexible file format).
- Calculate the orientation of a crystal at different goniometer settings.
- Calculate the coverage of these orientations in reciprocal space, in volume and for single reflections.
- Take into account crystal Bravais lattice symmetries and the higher redundancy of higher space groups.
- Optimize the experiment plan automatically.
- Compare predicted and actual measurement of reflections.

2.1. Instrument Configuration

CrystalPlan currently supports area detectors with flat faces and arbitrary rectangular dimensions; this reflects the detector types in use at SNS (Anger cameras and 8-pack ^{3}He tube detectors), but the software could easily be extended for more complicated detector shapes as needed. The detector configuration is loaded from a text file containing the location, size, orientation and number of pixels of each detector. Other settings to be adjusted for an instrument include the detectable range of neutron/x-ray wavelength.

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2.2. Sample Orientation

CrystalPlan can accomodate a variety of sample orientation goniometers. A base Goniometer class (in the object-oriented programming sense) is subclassed for specific types of goniometers, allowing a unified interface. This base class acts as a *universal goniometer*, which uses the conventional Eulerian goniometer angles of ϕ, χ, ω as defined in (Busing & Levy, 1967). Each class inheriting from the base Goniometer can convert the desired ϕ, χ, ω angles to the internal motor positions it requires. The class hierarchy allows for goniometer limitations: for example, TOPAZ is currently equipped with a goniometer where χ is fixed at 45 degrees (at ω to the incident beam). CrystalPlan handles this by limiting the sample orientation angles accessible to the user to ϕ and ω.

The sample unit cell and orientation is typically determined by the data collection/reduction software. In our case, from an initial data set, the data analysis program ISAW (Integrated Scattering Analysis Workbench (Mikkelson *et al.*, 2005)) provides us with the unit cell of the sample (via the B matrix) and the sample mounting orientation (U matrix). The combined UB matrix is then saved to a text file along with the lattice parameters. CrystalPlan can load this UB matrix file and uses it for reciprocal coverage calculations and optimization.

2.3. Reciprocal Space Representation

The reciprocal space to be measured is divided into a cubic 3D matrix of **Q**-space (with **Q** defined as $\frac{2\pi}{d}$). The user specifies the minimum d-spacing (d_{min}, in Å) that he or she is interested in, defining the limit $Q_{max} = \frac{2\pi}{d_{min}}$ of the modeled volume. There is no lower limit on **Q**.

The resolution of the 3D matrix is also specified (in $Å^{-1}$), which determines the number of points (voxels or volumetric pixels) to be modeled. Available machine memory limits the resolution that can be achieved in practice.

Each point of the reciprocal space 3D matrix holds an integer representing the number of times that voxel of **Q**-space has been measured – this is the total coverage matrix. To speed up the calculation of the total, a coverage matrix for each sample orientation is calculated and saved in memory and the total coverage matrix can be computed by simply adding multiple 3D matrices corresponding to each sample orientation.

2.4. Calculating Coverage of One Sample Orientation

For each sample orientation, a rotation matrix combining the sample's mounting orientation (the U matrix loaded from ISAW) and the goniometer rotation matrices (Φ, χ, Ω) is computed. The coverage in reciprocal space is computed in this way: for each pixel in the face of a detector, the direction of the scattered beam is known. Then, two incident beam vectors are considered, corresponding to the λ_{min} and λ_{max} wavelength limits of the detectors and/or source. The scattered beam direction and the two possible incident beam vectors are used to find two $\vec{q}$ vectors; these point in the same direction. At the high wavelength limit we find $\vec{q}_{min}$, and the low wavelength limit corresponds to $\vec{q}_{max}$. Since the instrument has a wavelength bandwidth, we fill in the 3D **Q**-space matrix by filling all points from $\vec{q}_{min}$ to the $\vec{q}_{max}$, for all pixels in the face of the detector. A typical wavelength band for TOPAZ is from 0.5 to 3.6 Å. This readily defines a volume of coverage, as shown schematically in two dimensions in Figure 1.

To reduce memory usage yet allow for quickly trying different detector configurations, the 3D matrix of each sample orientation is saved as a 32- or 64-bit integer, with each bit representing a particular detector. A simple binary mask can then be applied to quickly compute the coverage if some detectors are disabled, for example.

2.5. Crystal Parameters and Single Reflection Coverage

Crystal lattice parameters (lengths a, b, c and angles α, β, γ) are typed in or loaded from the ISAW UB matrix file. CrystalPlan then generates a list of *hkl* reflections with d-spacings larger than the d_{min} specified. For each sample orientation being simulated and for each reflection, a function calculates at what wavelength λ and in which direction it scatters. If λ is within range of the limits of the instrument, the beam direction is projected onto each detector face to determine where, on each detector surface, it will be measured.

Each *hkl* reflection is a separate object that holds all predicted recordings, including which detector measured it, at what (x,y) position on the detector face, at what wavelength, and for which sample orientation. These data are used to graphically display coverage and calculate statistics such as redundancy.

2.6. Crystal Symmetry

The crystal's point group symmetry can be entered along with its lattice parameters. This information is then used to calculate the increased measurement redundancy for higher symmetries (Giacovazzo *et al.*, 1992). A list of n 3×3 multiplication matrices is generated based on the point group; all *hkl* vectors are multiplied by these matrices to find the n equivalent *hkl* reflections; for example, for orthorhombic (mmm) symmetry, $n = 8$; therefore 8 multiplication matrices are generated that multiply *hkl* to give $\pm h$, $\pm k$, $\pm l$.

Each reflection is tracked separately, but a primary *hkl* is identified that points to a list of equivalent *hkl*'s when the crystal symmetry is known. The interactive GUI displays either all *hkl*, ignoring crystal symmetry; or combines them to display only the primary *hkl*'s, and calculates the redundancy by summing all measurements on all equivalent *hkl*'s.

In volume coverage mode, a 4D array is generated where the first 3 dimensions correspond to **Q**-space, and the 4^{th} dimension is a list of n entries. Each entry in the 4^{th} dimension points to a voxel that is an equivalent due to crystal symmetry. This allows CrystalPlan to quickly compute a volume coverage map that takes into account crystal symmetry and the resulting redundancy: at each voxel, all n equivalent voxels are checked to see if any of them were measured.

2.7. Automatic Coverage Optimizer

CrystalPlan allows the user to manually enter lists of sample orientations to simulate and graphically display the coverage in reciprocal space; however, finding an optimal plan by trial

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and error is in general time consuming and only feasible for an experienced crytallographer. To make the process more efficient, CrystalPlan includes an Automatic Coverage Optimizer, which takes as its inputs the desired number of sample orientations and uses a genetic algorithm to find a solution that satisfies a user-specified coverage criterion (for example, the user may ask for 85% of Bragg reflections to be measured, or for 90% of **Q**-volume).

Genetic algorithms have been covered in the literature before, see e.g. (Goldberg, 1989). In this application, the genes consist of goniometer angles. For instance, if a goniometer has freedom in 3 angles and the user requests 10 different sample orientations, then each individual has 30 genes. The initial population is created randomly and is made to evolve using mutations and crossover. Individuals whose goniometer angles turn out to be impossible to reach or beyond set limits are eliminated. For each individual set of sample orientations, the coverage statistics are computed (taking crystal symmetry into account if desired), and this is used as the fitness of the given individual in the normal genetic algorithm process. The best individuals are saved after each cycle. The simulation has succeeded once the desired value is reached or exceeded or the maximum number of cycles was calculated.

The Automatic Coverage Optimizer can find an acceptable solution (if any is possible) in times ranging from a few seconds to several minutes, depending on the difficulty of the problem and the computing hardware. Given that at a neutron facility, a measurement of a single orientation may take a few hours, this feature can result in significant beam-time savings.

3. Implementation

3.1. Language and Libraries

The program was designed to be cross-platform, and to run equally well on Linux, Mac OS and Windows, although Linux is our primary target operating system. Another design goal was to use open-source software and toolkits as much as possible.

To this end, CrystalPlan was written in Python (van Rossum, 1995), a powerful, open, cross-platform scripting language. Python was chosen for its ease of cross-platform deployment, and for its attractive language features. The numpy and scipy libraries (two open-source, commonly installed Python libraries) were used for many of the calculations (Oliphant, 2007; Jones *et al.*, 2001–). The most time-critical calculations were also written with inline C code, which increases the computation speed by up to 400× as compared with the pure Python version (which can still be used, in case of incompatibilities).

3.2. Code Layout

The calculation code of CrystalPlan is placed in a model layer and kept completely separate from eventual GUI elements, using a Model/View-Controller architecture (Gamma *et al.*, 1995). This makes it possible to run calculations using Python scripts, if desired. Unit tests for each part of the calculations are used to ensure consistent results on different platforms and systems.

3.3. Graphical User Interface

3.3.1. Design. An attractive and easy-to-use GUI was considered an important part of CrystalPlan, since the program is meant to be used interactively as a guide to the experimenter whether new or experienced. This requires intuitive GUI interfaces and workflows. The user interface was written using wxPython (Talbot, 2000), a mature cross-platform GUI toolkit, as well as the Enthought Traits GUI. Some of the 2D plots use matplotlib (Hunter, 2007), and the 3D visualization elements use the Mayavi2 toolkit for Python (Ramachandran, 2003), itself based on VTK (Schroeder, 2003). All of these libraries are open-source.

3.3.2. Main Program Workflow. A screenshot of the main window GUI is shown in Figure 2. The main window consists of a series of tabs, with workflow proceeding intuitively from left to right:

- **Q**-Space tab: define the volume of interest (minimum d-spacing) and resolution of reciprocal space.
- Detectors tab: load instrument detector geometry files, and enable or disable individual detectors.
- Goniometer tab: to choose the goniometer and degrees of orientation freedom of the sample.
- Sample tab: enter or load the crystal's lattice parameters and UB matrix.
- Try an Orientation tab: interactively rotate a sample and observe the changes in coverage in real-time.
- Add Orientations tab: type in a list of sample orientation angles to add to the experiment plan.
- Experiment Plan tab: a list of the previously calculated sample orientations. Here you can manage the orientations you will use in the experiment: add, delete, or temporarily disable them to see the effect on coverage. Once complete, the plan can be sent to the SNS/TOPAZ data acquisition computers in order to begin a run (Zolnierczuk & Riedel, 2010).

3.3.3. Reciprocal Space Coverage Visualization. Figure 3a) shows a screenshot of the reciprocal space coverage 3D interface. The 3D view can represent **Q**-space in two ways: volume coverage, or single-reflection coverage. The view shown can be rotated, translated or zoomed using the mouse or keyboard.

The volume coverage shows a solid isosurface representing the volume measured at least once. Redundant regions can also be shown using semi-transparent isosurfaces; or the coverage can be inverted, showing non-measured volumes only; or slices between user-selectable q_{min} and q_{max}.

The reflections view (Figure 4a) displays each *hkl* as a small sphere; the color of which indicates the number of times the *hkl* is predicted to be measured in the current experiment plan.

Both views can take into account crystal symmetry in their display using the techniques described previously in Section 2.6. The volume coverage (Figures 3b) fills in the sphere of coverage; whereas the single reflection view (Figure 4b) leaves only the primary reflections (one eight of the total, in this example).

Finally, both views display coverage statistics in the lower

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right corner, showing the percentage of volume (or of the number of reflections) measured, as well as the proportion of redundancy (voxels or reflections measured 2 or more times).

3.3.4. Single Reflection View. Figure 5 shows a screenshot of the GUI allowing one to observe in detail the predicted measurements of a particular reflection, by clicking in the 3D view or by typing in the *hkl* values. The position on the detector and wavelength of measurement are displayed. An additional window (not shown) allows the user to place the reflection on a particular position on a detector by automatically finding the necessary sample orientation angles. This is used, for example, to center important reflections or diffraction directions in the most sensitive part of a detector.

4. Sample Use Case

As part of the TOPAZ beamline commissioning process, a natrolite ($Na_2Al_2Si_3O_{10} \cdot 2\,H_2O$) single crystal was measured on a fixed 45° χ goniometer at room temperature and ambient atmosphere, with a wavelength range from $\sim$ 0.5 Å to 3.5 Å. An initial short event file (each neutron detection event is saved with its time-of-flight and position) was processed using ISAW to determine the UB matrix and cell parameters of the crystal.

The resulting UB matrix was loaded into CrystalPlan, where the Automatic Coverage Optimizer was used to generate an experiment plan with 8 sample orientations (Figure 6a) that covered 97% of the reflections, taking into account the crystal Laue symmetry (orthorhombic or mmm) (Figure 6b). The eight orientations were run unattended overnight.

Reflections were integrated using ISAW, and the resulting file was loaded into CrystalPlan for comparison. The predicted and actual measurement positions on each detector face were compared for the 554 integrated reflections found. Good agreement was found; the root-mean-square difference between the predicted and actual measurement wavelength was only 0.016 Å. The offsets in the reflection position were plotted, which helped show that the main contributor to this error was a slight misalignment of the goniometer.

CrystalPlan was also successfully used in a similar way during experiments run on the TOPAZ instrument by Yuri Janssen from State University of New York, Stony Brook, and Henrik Clausen from University of Aarhus, Denmark.

5. Conclusions and Future Development

We have described CrystalPlan, a software program designed to quickly produce efficient experiment plans for wide bandwidth time-of-flight Laue diffractometers. This program will help increase scientific productivity at SNS beamlines such as TOPAZ. With some relatively minor modifications, the program could also be adapted for use at reactor sources. Monochromatic reactor sources could be handled by adding support sweeps consisting of many motor positions; Laue diffractometers would requiring detecting and removing reflections with chromatic overlap.

One planned future development for CrystalPlan include extending the software to help predict coverage in inelastic neutron scattering experiments. Another desired feature is the introduction of feedback loops to help determine the right time to end data acquisition at a particular sample orientation. CrystalPlan would interface with data analysis software such as ISAW and use recently developed capabilities for real-time data analysis to monitor the signal to noise ratio on particular reflections. When the signal intensity becomes sufficient, the sample will automatically be moved to the next sample orientation, further adjusting data acquisition time for each specific sample. Combined with advanced sample changers coming on-line at TOPAZ, this will allow for extremely efficient, unattended and automated data acquisition.

Acknowledgements

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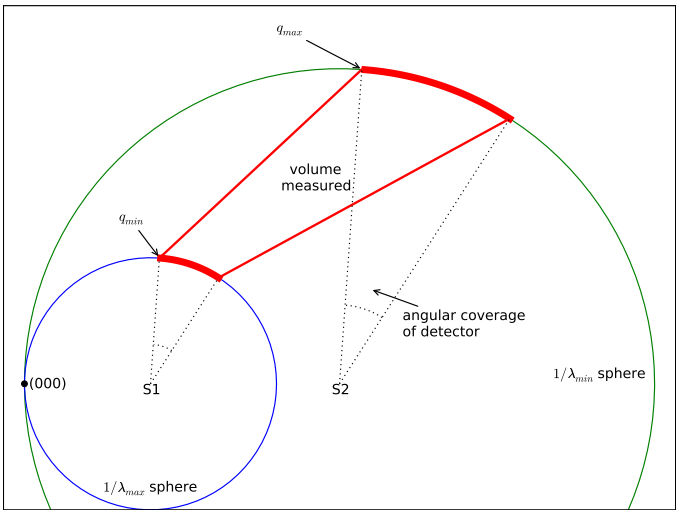


Figure 1
2D Ewald construction showing reciprocal-space detector coverage for a wide-bandwidth Laue diffractometer. The smaller blue circle represents the $1/\lambda_{max}$ limit of measurement, with the larger green circle representing $1/\lambda_{min}$. The origin of reciprocal space (000) is common to both spheres, but the "virtual" sample position in the construction (S1 or S2) depends on λ. The angular coverage of the detector is represented by the dashed wedge, and projects an arc onto each sphere of radius $1/\lambda$. The red wedge between the two λ is built by drawing straight lines between the q_{min} and q_{max} limits for each pixel on the detector face, giving the volume in reciprocal space that is measured at a single sample orientation.

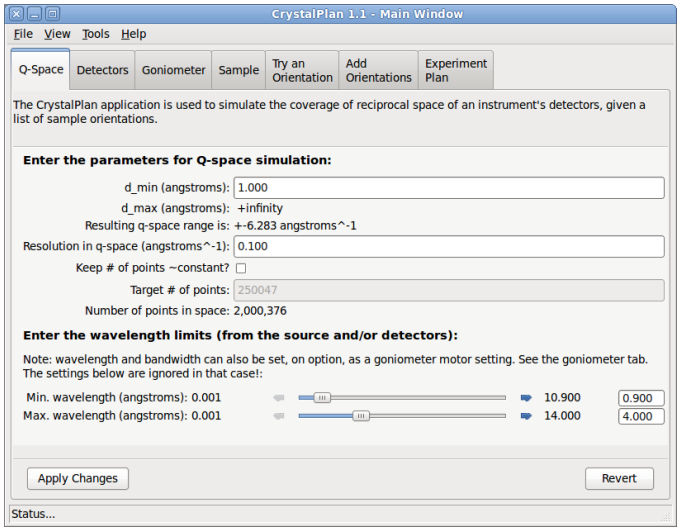
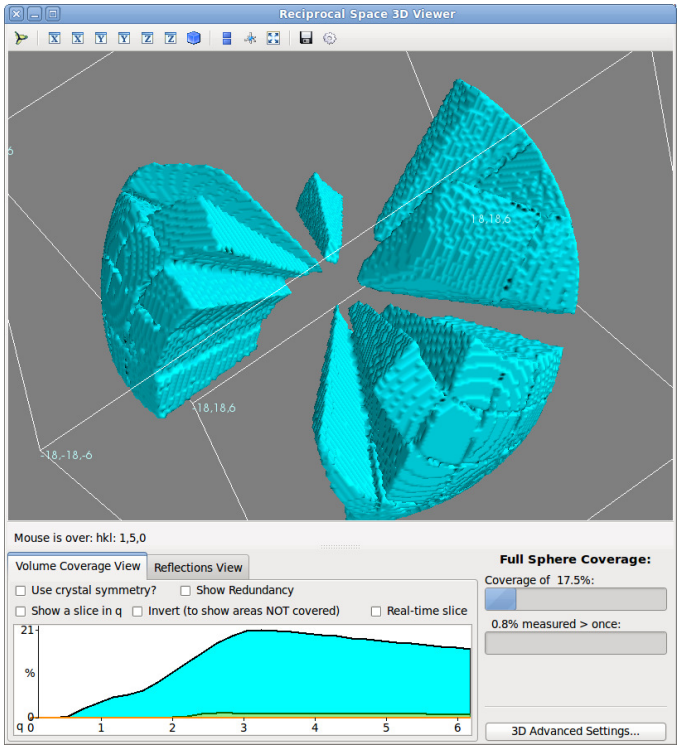
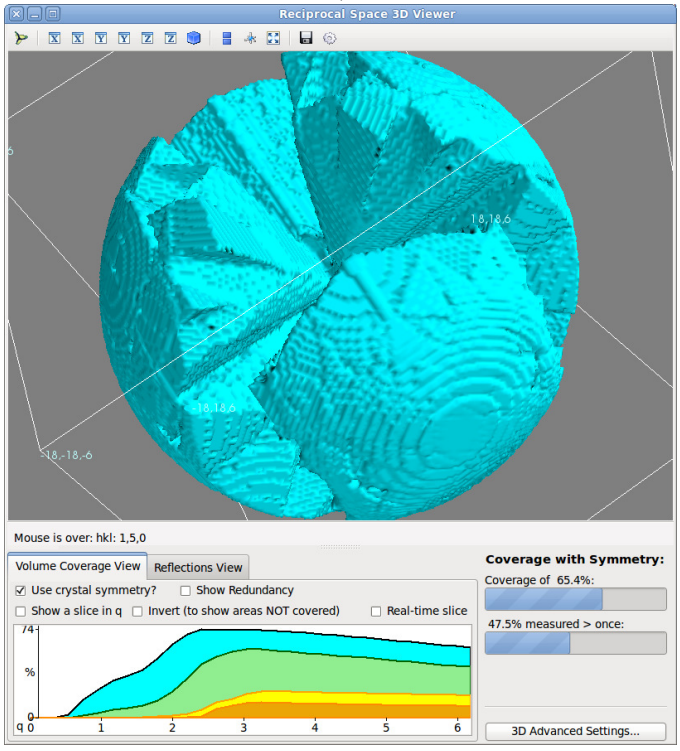


Figure 2
Screenshot of the main GUI window. Six more interface windows (not shown) are presented to the user when he or she clicks on the tabs at the top of the main window.



a)



b)

Figure 3
Sample screenshot showing the 3D view of the predicted coverage of reciprocal space volume for 3 sample orientations and 14 detectors on TOPAZ, from 0.9 Å to 4.0Å: a) without crystal symmetry, 17% coverage; b) with crystal symmetry (mmm 8-fold symmetry), coverage increases to 65%.

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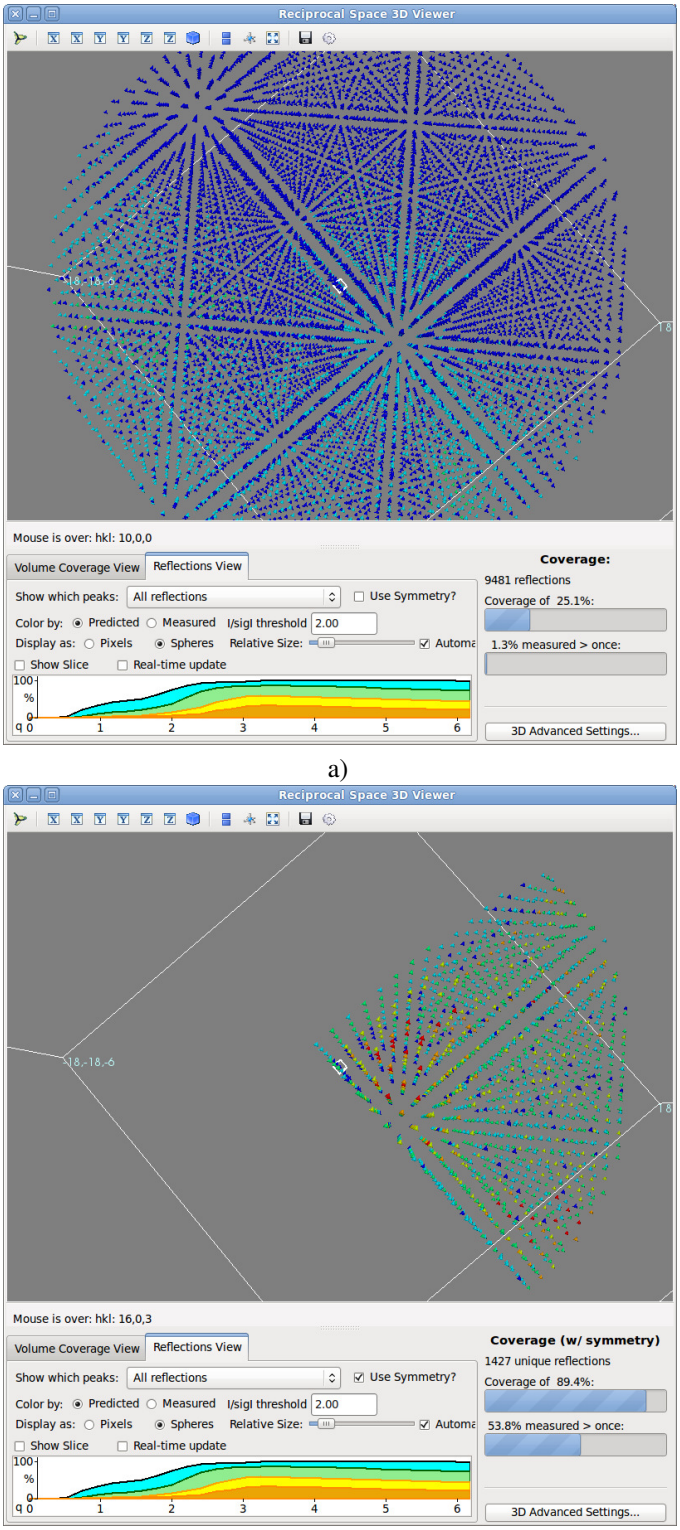

Figure 4
Sample screenshot showing the 3D view of single-crystal reflection measurements. Each sphere represents an *hkl* reflection, color-coded to indicate the number of measurements (dark blue: 0, cyan: 1, green: 2, yellow: 3, red: 4 or more). a) without crystal symmetry, 2375 of 9481 reflections are measured, or 25%. b) with crystal symmetry, 1276 of 1427 primary reflections are measured, or 89%, and redundancy is also increased.

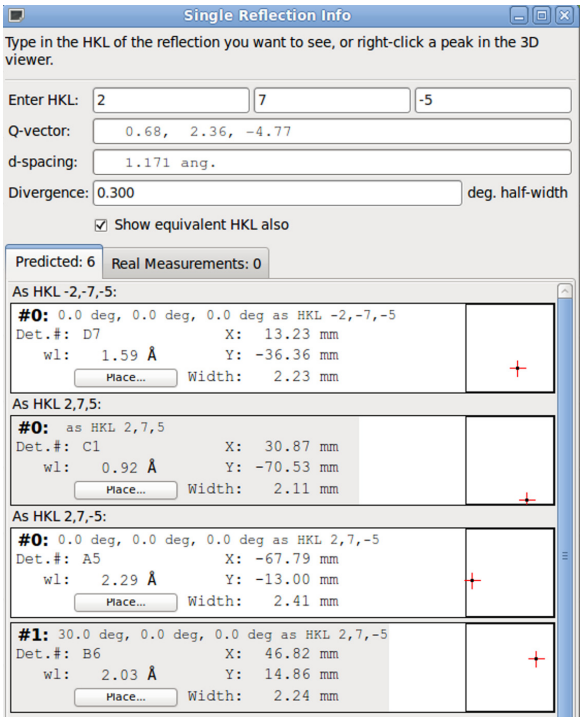

Figure 5
Part of a GUI showing details of the predicted measurement positions for a single *hkl* and some of its equivalents.

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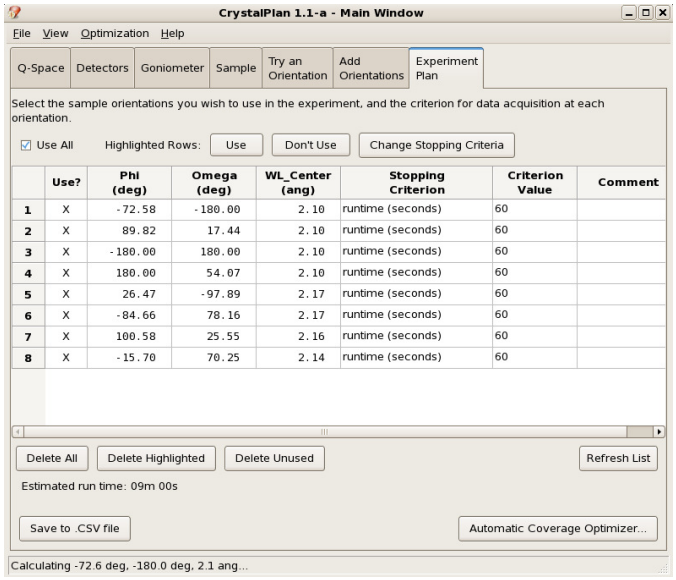
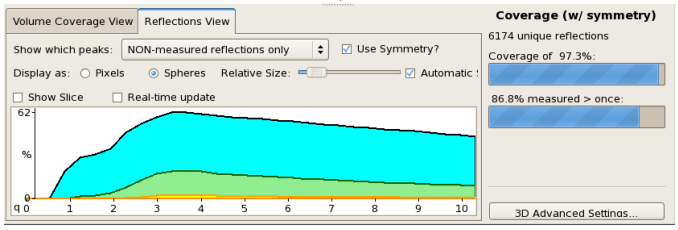


a)



b)

Figure 6
a) Screenshot of the Natrolite experiment plan generated using the Automatic coverage Optimizer. b) Screenshot of the resulting coverage statistics.

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