Mail-in synchrotron powder diffraction at the APS

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As everyone in the field knows, the information obtained from a diffraction experiment is often limited by how much can be measured. This is especially true for powder diffraction experiments, where the information obtained from a powder pattern can be limited by several experimental effects. These effects may include: signal-to-noise, determined by the source intensity and ability to separate diffraction signal from air scatter and fluorescence; measurement range, limited by the source wavelength; and poor instrumental resolution, which reduces the number of observable intensity measurements due to the overlap of separate reflections.

The use of synchrotron source affords an opportunity to minimize the impact of all these limitations by offering a very intense and nearly parallel beam of high-energy X-rays. Armed with this intense beam, three-crystal diffraction optics (two-crystal monochromation combined with a perfect-crystal analyzer) can be added to enable the detection of diffraction peaks with very high resolution. The use of such a crystal analyzer design also enhances signal *vis-a-vis* background, in that fluorescence and incidental scatter is rejected, since only photons with the right energy and coming along the right path are diffracted by the analyzer. The use of high-energy X-rays allows measurements to be conducted to higher-Q values ($Q = 4\pi\sin\theta/\lambda = 2\pi/d$) and allows greater penetration into high-Z materials. The nearly parallel beam intrinsic to a synchrotron source avoids use of Bragg-Brentano focusing, with its inherent uncertainty for the location of zero for 2θ. Imprecision in $2\theta_0$ is reduced by typically two orders of magnitude with a synchrotron. Finally, synchrotron powder diffraction measurements typically employ a Debye-Sherrer transmission geometry (with a rotating capillary sample), which often minimizes problems of preferred sample orientation. However, the triple-crystal optic is very inefficient and even with the great intensity of a synchrotron source an average scan might require close to a day. For this reason, new synchrotron powder diffractometers are now constructed with anywhere from 10 to 40 sets of analyzer crystals and detectors, offering much faster data collection times of approximately 1 hour. Readers are referred to Fig. 1 for a visual A visual example of just how good a one-hour scan can be. This shows a one-hour scan of the NIST LaB_6 standard.

Thus synchrotron measurements are the preferred technique for structure determination from powder diffraction. The improved accuracy of the 2θ measurement, along with the improved ability to resolve individual peaks, greatly simplifies the task of indexing patterns for new materials. These measurements also offer an order of magnitude better sensitivity and precision for phase detection and quantification.

A problem with synchrotron experiments can be access to the instrument. Historically, this requires writing a proposal, a delay of at many months while proposals are reviewed; and then further delay for successful proposals to be scheduled. Typically, the scientist must finally travel to the synchrotron to perform the measurements. This is expensive and time consuming. Not surprisingly, relatively few research groups perform such measurements and those that do, often have significant delays between when a material is first identified for study and when the measurements are finally made.

The Advanced Photon Source (APS) synchrotron facility at the Argonne National Laboratory has made progress in alleviating some of these problems. Nearly a decade ago, several forward-thinking scientists (lead by John Mitchell, Jim Jorgensen, and Peter Lee), proposed adding a synchrotron powder diffraction beamline at the Argonne National Laboratory's Advanced Photon Source (APS) and in that proposal they envisioned a solution to this historical barrier. They called for an instrument equipped with an industrial sample-loading robot, a barcode reader, and cryo-cooling/heating device so that unattended powder diffraction measurements, even at non-ambient temperatures, could be performed. An engineering model for the instrument is shown in Fig. 2. They also proposed that access to the instrument could be provided without the need for users to come to the beamline. Their proposal was funded by the US Department of Energy and the APS beamline 11-BM was selected for its location. The prospect of offering scientific access to this instrument was sufficient to attract one of us (BHT) to oversee its construction and design the workflow and the other (MRS) to oversee its operation.

Fast-forward to today; the 11-BM powder diffraction instrument is operational and available to users from around the world. Access can be made without travel ("mail-in access") for short experiments that meet certain criteria, or with travel to the APS ("on-site access") for more complex or longer experiments. Proposals from potential users are required in either case, but mail-in users may submit via a rapid access process, which removes the long delay for the proposal review and scheduling. Extensive information about the beamline and how to obtain access to its user program can be found on website http://11bm.xor.aps.anl.gov/. Experiments suitable for mail-in are cases where:

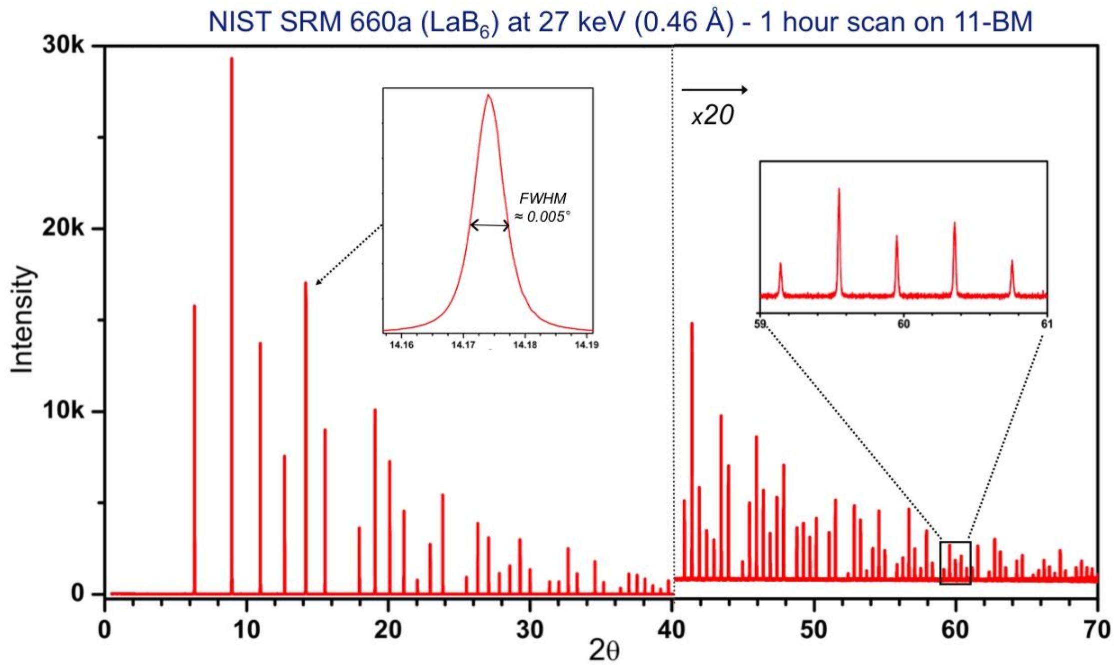
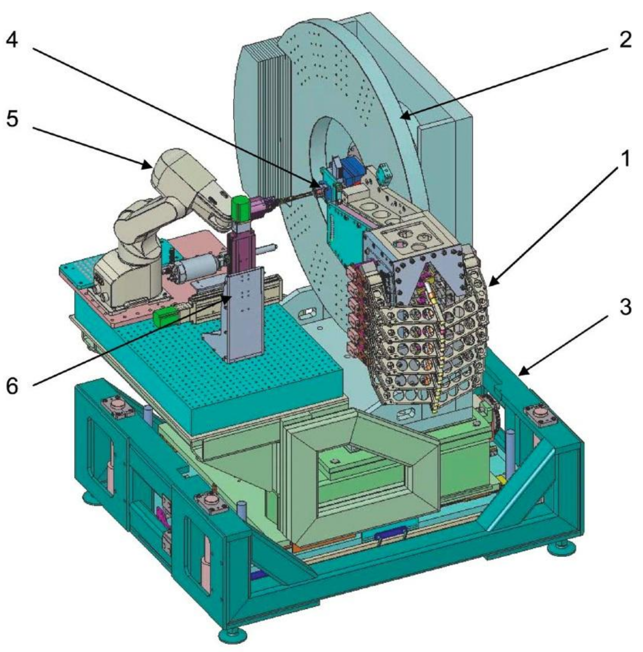


Figure 1. A scan of the NIST 660a LaB_6 Standard Reference Material, where data were collected in one hour of the APS 11-BM diffractometer. Sections of the diffraction pattern have been enlarged to show additional detail. Vertical axis shows the approximate number of counts collected at each angle.



11-BM Diffractometer Schematic: (1) 12-analyzer detector system, (2) two-circle goniometer, (3) supporting table, (4) sample stages, (5) sample mounting robot, (6) stages for cryostream etc.

Figure 2. An engineering schematic of the 11-BM diffractometer showing (1) the 12-analyzer detection system, (2) the two-circle vertical goniometer, (3) the supporting table with vertical and lateral motion adjustments, (4) the modular sample mounting stage, (5) the sample mounting robot and (6) the sample environment positioning device that allows the cryostream to move out of the way of the robot.

1) shipping the material does not present a hazard,
2) measurements can be completed in 8 samples or < 8 hours,
3) scan temperatures are between 100 K and 450 K, and
4) where data collection conditions can be planned in advance and order of measurements is not important.

In contrast, if review of one measurement is needed to plan the next, or where a sample heating or cooling protocol must be used, this defines an experiment that needs an on-site participant on-site at the beamline. Likewise, use of the more complex equipment needed to cool samples to below 100 K or to heat up to 950 C is only accessible to on-site users.

For mail-in use, the user must prepare the sample(s) using a kit and instructions provided by the beamline. This is described in more detail on the beamline website. All mail-in measurements are made at ~30 KeV (~0.4 Å); the user has some discretion on data collection parameters.

The beamline web site also provides copious information on how to submit a proposal for use of the APS and on how the mail-in submission process works. One suggestion to interested readers is that they investigate if their institution has a signed agreement in place for access to the APS (a "User Agreement") early in the process. This is a legal document and it is required that the lead institution on every proposal must have this agreement in place to access our facility. This is required for both mail-in and on-site access, and can be time consuming to process depending on your institution's requirement.

Readers of this newsletter may be pleased to note that the US does not prioritize access to APS facilities by scientists from any particular country. We wish to encourage access to users, both domestic and international, who will perform the best science and will publish their results to the greater community. However, overseas mail-in users must abide by a few additional constraints. They must handle all paperwork to have their samples shipped into the US (we do not sufficient resources to respond to U.S. Customs or 3rd party shippers; as such, all such requests will be directed to the shipper.) Likewise, return shipping of samples to samples to addresses outside the U.S. is not available. More information can be obtained on the webpage or by contacting beamline staff.

In addition to the extensive information on the beamline web site, the reader is also referred to three papers [1-3] that have been published describing the detector system, the instrumental performance and the automation design.

References

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