The TOPAZ experience in current TOF-Laue experiments

To validate our simulations, we performed an experiment on TOPAZ at SNS. TOPAZ is a Laue-TOF single crystal diffractometer dedicated to crystal structure studies. It has large detection coverage on one of the most advanced spallation source in operation. In this respect, it is of interest to show the possibilities of this instrument type and to compare the performance of TOPAZ with MAGiC. One should note however, that TOPAZ is not particularly dedicated for science cases related to magnetism. It cannot cover the needs for low temperatures or high magnetic fields (Anger camera detectors) and does not offer polarized neutrons.

We performed a diffraction experiment on a $(La_xSr_{14-x})Cu_{24}O_{41}$ compound. In these composite compounds, two sub-lattices are coexisting, ladders and chains, with different *c* parameters. The orthorhombic unit cell is defined by *a*=13.2 Å, *b*=11.41 Å and *c*=3.9 Å for the ladders and *c*=2.75 Å for the chains. An effective large unit cell can be defined containing both the ladders and chains with *c*=27.5 Å. The large effective lattice parameters yield a huge number of attainable reflections with a wavelength range on TOPAZ from 0.4 Å to 3.6 Å. The sample volume used was of 1.5x2.5x2 mm^3 with a close to perfect rectangular shape. (Due to relatively small beam size (3x3 mm), samples bigger than 2 mm are not used on TOPAZ.)

Unfortunately, no magnetism study is currently possible on TOPAZ as it is not equipped with proper cryogenics yet. The minimum temperature achievable on sample using nitrogen flux is 90K. The advantage tough is that in this case there is no material in the beam neither in the incident nor in the scattered beam, which decreases enormously the background. TOPAZ is equipped with a kappa goniometer (with rotation angles ω, φ and χ=135°). Samples are glued on magnetic pins and centering of samples is made by in-situ video camera using piezzo-electric drives.

Data acquisition strategy is defined by the CrystalPlan software which maximize Q-space coverage and minimize the total number of angular positions for a desirable redundancy factor. 12 positions were required in our case with an hour counting time for each, with redundancy factor of about 3. Data set obtained at each position contained a list of about 3×10^{7} events and occupied ~300 Mb. A total of 600 peaks (>3σ) were typically found and indexed for each sample orientation. Once data acquisition is launched, a live access to data is available. One can visualize slices in Q-space, find Bragg spots, find the lattice parameters by FFT and refine the UB matrix. The full data reduction suite is based on the MANTID platform. Structure factors extraction from a data set is done during experiment with a typical delay of an hour from the time of the set acquisition, essentially due to rather complex binning procedure of raw data.

After the experiment SNS is giving a free remote access to its high performance computing possibility to users, but we have transferred raw data to Saclay in order to re-treat them. The treatment using MANTID was possible only on our dedicated high performance computer (>128GB of RAM and 48 logical cores), as the memory requirements for single crystal data reduction are quite high. The full data reduction process is now operational at the LLB on this computer, which allows to:

- Load one or all datasets (20s / dataset)
- Convert each dataset to instrument Q-space (40s / dataset)
- Apply absorption correction based on wavelength, sample shape, and composition. Detector efficiency is also corrected automatically (2 mn / dataset).
- Find all peaks in a given dataset using user-defined threshold (5s / 2000 peaks)
- Refine UB matrix based on the obtained peak list (2s). A few matching UB matrix are proposed to the user.
- Index peaks using the refined UB matrix (5s).
- Integrate peaks using one of the proposed algorithms (spherical, ellipsoidal, cylindrical). This part of the process is not yet optimized and takes ~40mn for 2000 peaks (per dataset in our case) on our dedicated computer.
- Export a list of hkl and associated intensities for data treatment.

The total processing time is currently close to an hour per dataset being comparable with the acquisition time. There is a lot of room for improvement on the integration algorithm as it is not parallelized and operates only on one core. Optimization of the software should reduce the total data treatment time down to a few minutes.

Preliminary crystal structure refinement using Amma(00γ) Super Space group has been made using JANA2006 which has been recently adapted for the output format of TOPAZ data. This was necessary to account for the wavelength dependence of extinction corrections occurring in Laue-TOF method. Final R-factors of 6.9% was achieved on main sub-lattices reflections after merging and 7.2% on first order satellites. Higher satellite orders (up to fourth) were extracted and refinement is still in progress.

This shows that TOPAZ, and other Laue-TOF instruments in general, are now extremely efficient for high-resolution crystal structure determination, and in some cases competitive with X-ray scattering instruments.

Remarks:

- The whole data reduction process is operational but not yet sufficiently user friendly. Participation of local contact in data treatment is still strictly mandatory.
- Data treatment on incommensurate structures is not available yet. This problem needs to be solved on MAGiC, which will regularly deal with incommensurate magnetic structures.

MAGiC simulation: we performed a simulation of the same experiment on MAGiC to confirm the validity of our estimated counting time. Based on SNS flux and repetition rate, we estimate a 60 gain factor for MAGiC in count rates. However, this huge gain factor has to be taken with caution due to other relevant parameters:

- TOPAZ wavelength range extends down to 0.4Å. The effective 3D Q-space covered by TOPAZ is more than 3 times higher than the one of MAGiC.
- Longitudinal resolution on TOPAZ is much better at short wavelength, allowing for studying compounds with larger unit cell without pulse shaping, while MAGIC in this case will have to be used in pulse shaping mode decreasing the usable flux.

Using structure factors of reflections observed on TOPAZ we have simulated a scattering pattern on MAGIC obtained by full sample rotation. We obtained similar statistics as on TOPAZ in 11 minutes within the Q-space accessible on MAGIC ($Q_{max}<20$ Å $^{-1}$) and poorer longitudinal resolution. This resolution was, however sufficient for the proper intensity integration with the lattice parameters of our sample. It corresponds to a 66 gain factor close to the expectation. This result validates the time estimates made in the proposal for the various examples.

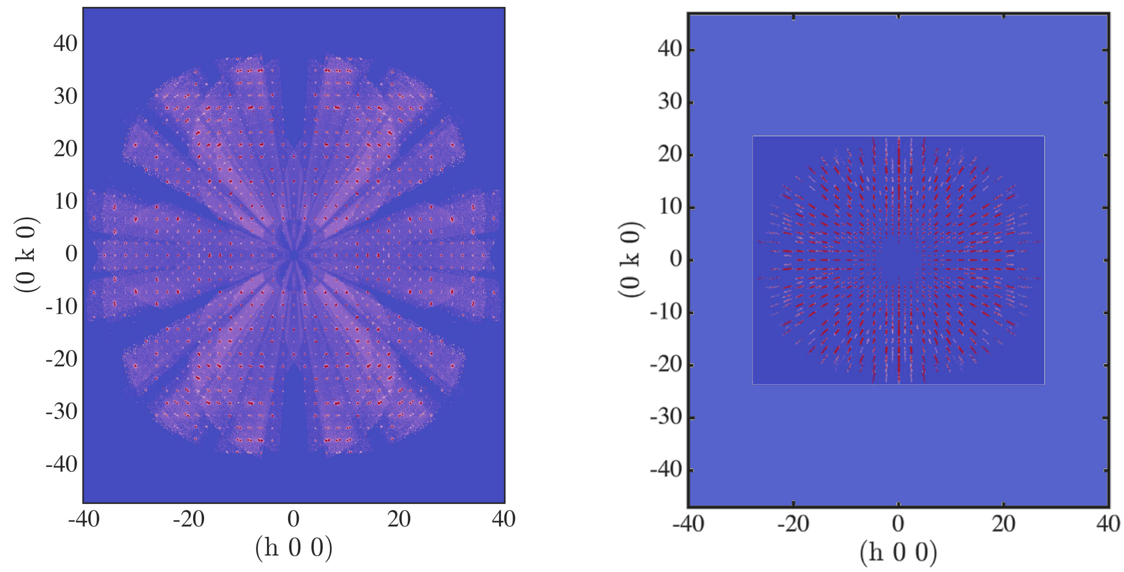

Fig. : *Left* : (hk0) diffraction pattern obtained on TOPAZ. *Right* : (hk0) diffraction pattern obtained in MAGiC simulation. TOPAZ pattern corresponds to a 12 hours acquisition time while MAGiC one corresponds to an 11 minutes one. Both patterns make use of the orthorhombic symmetries.