Oral presentation

Single-pulse, single-crystal diffraction at XFELs

G. Bortel[1], M. Tegze[1], G. Faigel[1]

[1]*HUN-REN Wigner Research Centre for Physics, Budapest, Hungary*

bortel.gabor@wigner.hun-ren.hu

Conventional X-ray single-crystal diffraction measures the reflections, while the crystal is rotated. Similarly, serial crystallography at synchrotrons and X-ray free electron lasers (XFELs) takes multiple diffraction patterns of crystal replicas in random orientations. The common requirement of changing / multiple orientations is imposed by the need to swipe through the 3D reciprocal space with the 2D sampling surface of the Ewald sphere.

Although XFEL sources provide enough photons for a complete diffraction experiment in a single, few fs long X-ray pulse, the conventional way of data collection cannot be realized due to the inability to sample multiple orientations in such a short time. Aiming at a single-pulse, ultrafast diffraction experiment, one needs to rely on a parallel technique capable of simultaneously measuring all reflections. This could be achieved by either polycrystalline diffraction or a Laue single-crystal diffraction experiment, but these either provide limited 3D diffraction information (due to the orientational averaging) or are simply unfeasible for XFELs (due to the inherent monochromatic nature of the source). Therefore, a new measurement principle is required.

In our recent publication [1] we presented and demonstrated an experimental technique that measures all Bragg-reflections of a single-crystal sample in parallel, without the need for sample rotation, hence it can give the complete 3D structure of the crystal using a single XFEL pulse. This method relies on the Kossel-lines first observed and explained at the dawn of X-ray research. In the process, a secondary spherical radiation field is created that reaches the crystal from all directions, and only the components satisfying the Bragg condition are reflected and form a conical radiation field leaving the sample. The secondary sources are some atoms of the sample itself, that are excited by the XFEL pulse and emit fluorescent radiation in the X-ray energy range. Since the source atoms are part of the crystal structure, their location is in registry with the lattice, and this results in an interference signal across the Bragg-reflection, encoding the phase of the given reflection's structure factor.

In our demonstration experiment at the European XFEL facility, we used an XFEL pulse to excite Ga atoms of stationary GaAs and GaP single-crystal samples and recorded all the Kossel-lines appearing as conic sections on a flat detector surface. A Kossel- line pattern first of all contains geometrical information. The axis and opening angle of the cone for each line give a reciprocal lattice vector and ultimately define the crystal lattice and crystal orientation. The high enough angular resolution of the recorded pattern also reveals the fine interference signal across each Kossel-line enabling derivation of both the amplitude and the phase of the corresponding structure factor. This information allows direct derivation of the electron density within the unit cell using Fourier synthesis. Detailed steps to obtain the structure from a single Kossel-line pattern created by a single XFEL pulse are explained.

The presented inside source diffraction technique simultaneously enables ultrafast, parallel collection of complete diffraction data at XFELs and provides an elegant way of avoiding the phase problem when reconstructing the structure.

[1] Gábor Bortel, Miklós Tegze, Marcin Sikorski, Richard Bean, Johan Bielecki, Chan Kim, Jayanath C. P. Koliyadu, Faisal H. M. Koua, Marco Ramilli, Adam Round, Tokushi Sato, Dmitrii Zabelskii and Gyula Faigel: 3D atomic structure from a single X-ray free electron laser pulse *Nature Communications* **15**, 970 (2024). https://doi.org/10.1038/s41467-024-45229-8