Per-pixel XFEL diffraction data processing for wavelength deconvolution

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Advances in XFEL and detector technologies are on track to be able to enable diffraction experiments in the kHz and even MHz regimes at several linear accelerators worldwide in the coming years (Zastrau et al. 2021; Zhang et al. 2020). The computational demands of such experiments are daunting from many angles, but their scientific potential is equally incredible (Blaschke et al. 2021). One type of experiment piloted at current LCLS pulse repetition rates aims to pinpoint oxidation states of individual metal ions in a crystal structure without scanning the metal edge - or, more accurately, by collecting diffraction data from all wavelengths simultaneously and deconvoluting their contributions after the fact (Sauter et al. 2020). The natural bandwidth of LCLS pulses is already optimal for spanning the metal edge, and the stochastic individual spectra can be recorded in order to track their contributions at each wavelength. The diffraction signal attributable to each wavelength accumulates at a slightly different position on the detector for each Bragg peak, since each wavelength produces slightly different diffracting conditions, producing radially streaked reflections. The distribution of signal depends on several factors: the spectrum of the X-ray pulse irradiating the crystal, the precise crystal parameters and orientation, the wavelength-dependent parallax effect and the point-spread function on the detector. Selecting the values for each of these parameters that best fit the pixel-wise image data allows us to track and individually recover the contributions of each wavelength to each Bragg peak. This process has been validated on simulated data with the program nanoBragg (Mendez et al. 2020). To extend it to experimental data, we require the ability to refine individual pixel observations against the crystal model in a computationally feasible manner. The development of diffBragg addresses this step by computing first derivatives of complete (pixel-wise) images to use in global minimization. Finally, incorporation of this approach into the existing xfel.stills_process pipeline and the codevelopment of these tools with the exascale computing framework necessary to use them at scale are expected to make this approach tractable at upcoming XFEL experiments.

Blaschke, Johannes P., Aaron S. Brewster, Daniel W. Paley, Derek Mendez, Asmit Bhowmick, Nicholas K. Sauter, Wilko Kröger, Murali Shankar, Bjoern Enders, and Deborah Bard. 2021. "Real-Time XFEL Data Analysis at SLAC and NERSC: A Trial Run of Nascent Exascale Experimental Data Analysis." ArXiv, August. https://www.ncbi.nlm.nih.gov/pubmed/34189183.

Mendez, Derek, Robert Bolotovsky, Asmit Bhowmick, Aaron S. Brewster, Jan Kern, Junko Yano, James M. Holton, and Nicholas K. Sauter. 2020. "Beyond Integration: Modeling Every Pixel to Obtain Better Structure Factors from Stills." IUCrJ 7 (Pt 6): 1151–67.

Sauter, Nicholas K., Jan Kern, Junko Yano, and James M. Holton. 2020. "Towards the Spatial Resolution of Metalloprotein Charge States by Detailed Modeling of XFEL Crystallographic Diffraction." Acta Crystallographica. Section D, Structural Biology 76 (Pt 2): 176–92.

Zastrau, Ulf, Karen Appel, Carsten Baehtz, Oliver Baehr, Lewis Batchelor, Andreas Berghäuser, Mohammadreza Banjafar, et al. 2021. "The High Energy Density Scientific Instrument at the European XFEL." Journal of Synchrotron Radiation 28 (Pt 5): 93–1416.

Zhang, Z., A. S. Fisher, M. C. Hoffmann, B. Jacobson, P. S. Kirchmann, W. S. Lee, A. Lindenberg, et al. 2020. "A High-Power, High-Repetition-Rate THz Source for Pump-Probe Experiments at Linac Coherent Light Source II." Journal of Synchrotron Radiation 27 (Pt 4): 890–901.