MP_tools: from molecular dynamics simulations to diffuse scattering maps

J. Kulda
Institut Laue-Langevin, BP 156, 380042 Grenoble Cedex, France
kulda@ill.fr

To extract information on disordered structures from scattering data one often has to compare model-based intensities with the observed ones. The progress in computing techniques in last decades permits to produce realistic models of crystalline lattices by a variety of approaches ranging from *ab initio* DFT methods via molecular dynamics (MD) to phase-field models based on the Landau formalism.
With this progress in place the bottleneck has shifted from producing supercell models to generating the corresponding diffuse scattering distributions in reciprocal space. The principal issue was due to the fact that scattering amplitudes from a distorted lattice could not be summed up using fast Fourier transform algorithms (FFT) because of the displacement phase factor exp(-i$\boldsymbol{QR}$) being $\boldsymbol{Q}$-dependent.
As a consequence, many efforts in recent years have been restricted to simple models on small supercells [1,2] or to more involved pair distribution function (PDF) analysis [3-5], where the summation problem is reduced to a single dimension. Nevertheless, the efficiency of PDF model generation in direct space for large supercells and large correlation distances needed for good quality Fourier transforms was limited by the radial distribution function spherical shells volume diverging with R^2.
The *MP_tools* program suite [6] addresses these issues employing innovative algorithms. In the first case, recent developments of the non-uniform fast Fourier transform [7] permit to accelerate the summation of scattering amplitudes from large supercells by orders of magnitude, bringing in the usual FFT speed and allowing for interactive work even in case of dynamic scattering functions S($\boldsymbol{Q}$,ω) based on time sequences of thousands of frames. A similar effect in the PDF accumulation brings a Monte-Carlo algorithm with projective sampling [8], permitting to accumulate the *g(r)* pair-distribution function with uniform (*r*-independent) accuracy without passing *via* the radial distribution function.

Many motivating discussions with Marek Pasciak, Petr Ondrejkovic and Jirka Hlinka, colleagues from the Institute of Physics (AS CR) are kindly acknowledged.

[1] Welberry T.R., Butler B.: J. Appl. Cryst. 27 (1994) 205-231
[2] Neder R.B., Proffen Th.: Diffuse Scattering and Defect Structure Simulations: A cook book using the program DISCUS, Oxford, 2008; DOI:10.1093/acprof:oso/9780199233694.001.0001
[3] McGreevy R.L., J. Phys.: Condens. Matter 13 (2001) R877–R913
[4] Proffen Th. et al., Z. Kristallogr. 218 (2003) 132–143
[5] Eremenko M. et al., Nature Comm. 10 (2019) 2728
[6] Kulda J., https://github.com/jkulda/MP_tools, https://mptools.fr
[7] Barnett A.H. et al., J. Sci. Comput. 41 (2019) C479-C504, https://github.com/flatironinstitute/finufft
[8] Kulda J., Acta Cryst. A (2023) in preparation