Direct optimisation of physical parameters in TLS models

Ying Luo, Nicholas M. Pearce

Physics, Chemistry and Biology (IFM), SciLifeLab, Linköping University.

Translation-Libration-Screw (TLS)_models produce group anisotropic atomic displacement parameters (a-ADPs) using relatively few parameters compared to individual atomic a-ADPs. However, there has been a long-standing problem with the interpretation of refined TLS matrices in terms of physical motions, with refined TLS models often physically invalid. This problem stems from the refinement approach used in modern refinement programs, where they refine elements of the intermediate TLS matrices, rather than the underlying physical parameters.

To address this challenge, we restructured the TLS matrices in terms of physical motions in a spherical coordinate system. This allows for the direct optimisation of a TLS model by gradient descent with easily validated outputs. These changes are implemented in the ECHT model, a hierarchical TLS disorder analysis model, where the enhancements in parameter reliability and the streamlining of optimisation operations reduce the need for multiple optimisation regimes and a laborious validation process, while simultaneously providing more precise and reliable results.