

Structure Determination from Multiple-Scattering Electron Crystallography

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Structure Determination from Multiple-Scattering Electron Crystallography

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Transmission electron diffraction (TED) from large crystals involves high degrees of dynamical multiple scattering, which greatly complicates the analysis of such data. As such, TED structure solution is often limited to crystals of at most a few tens of nanometers thick, where kinematic diffraction dominates. We describe a new iterative projection algorithm (N-phaser) capable of reconstructing a scattering potential from its N-beam multiple Bragg scattered intensities [1,2]. The algorithm can recover accurate complex structure factors from a wide range of crystal thicknesses, orientations, relativistic beam energies, and does not require known thickness or atomicity constraints if sufficient multiple scattering is present.

Our approach is based on Sturkey's scattering matrix formulation of multiple scattering [3]. Here, the scattering matrix, describing high-energy transmission electron diffraction through a crystal slab of thickness t , is defined via the matrix exponential $S = \exp(2\pi i A t)$, where A is an $N \times N$ matrix indexed by the lattice vectors \mathbf{g} , contains the desired structure factors $V_{\mathbf{g}}$ of the potential V in the off-diagonal positions, and has known excitation errors $s_{\mathbf{g}} = \lambda \mathbf{g}^2/2$ along the diagonal.

The intensities of the transmitted Bragg beams are proportional to the matrix M of moduli of the elements of S . In particular, each column of M contains magnitudes from one two-dimensional Bragg diffraction pattern, where adjacent columns correspond to patterns obtained by tilting the beam by a succession of different Bragg conditions around the zone axis orientation, allowing the entire matrix M to be sampled [4]. Determining the structure of the crystal sample requires recovering the structure factors $V_{\mathbf{g}}$ from M . In this case, we seek a solution that satisfies three constraints: 1) the known inherent symmetries of A , 2) the known diagonal elements of A , and 3) consistency with the experimentally available scattering magnitude matrix M .

To solve this structure determination problem, the N-phaser algorithm starts with a random potential V and iterates between enforcing the known magnitude constraints on S and the symmetry and diagonal constraints on A via projection operations (See Fig. 1). In particular, this approach is capable of solving the structure without knowledge of the thickness t . At small thicknesses, kinematic diffraction dominates, and additional atomicity constraints are needed, achieved via charge flipping. However, we show that at larger thicknesses, the high degree of multiple scattering provides sufficient additional information for inversion without additional real-space constraints. Results are presented in reconstructing the 2D projected potentials of both GaAs (Gallium Arsenide) and spinel crystals from their corresponding scattering matrix magnitudes at multiple thicknesses and energies (See Fig. 2).

We also demonstrate the existence of a fundamental instability in the relationship between A and S that severely complicates inversion for very thick crystals, and how the N-phaser algorithm can be modified to handle it. Furthermore, we show how this instability affects the ability to retrieve chirality from

multiple-scattering data as a function of thickness. [5]

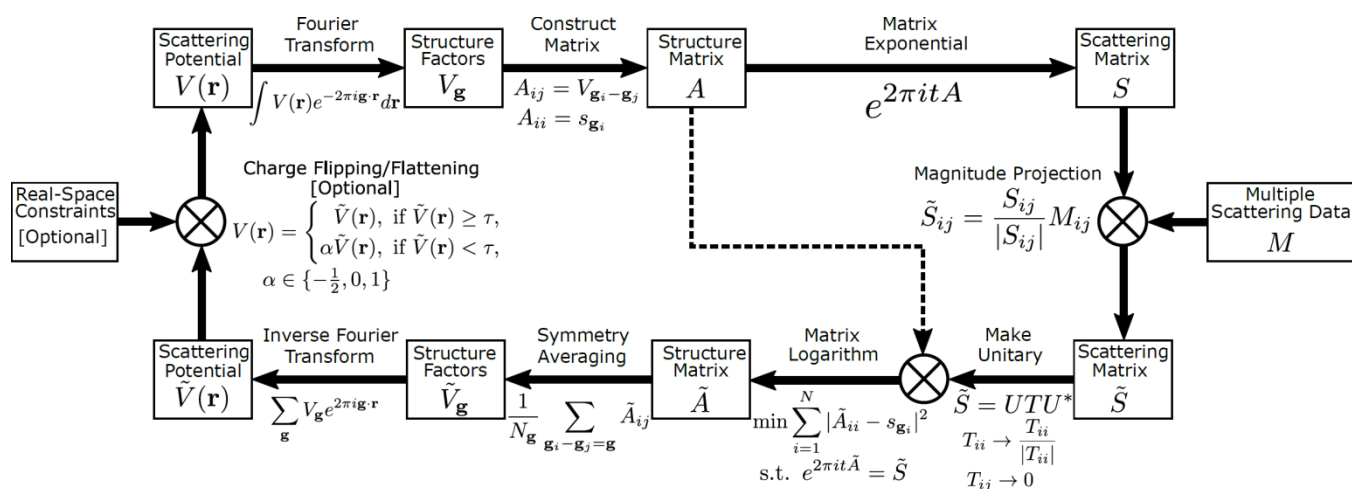


Figure 1. Flowchart description of the iterative N-phaser algorithm for reconstructing projected potential maps from N-beam multiple Bragg scattered intensities [1].

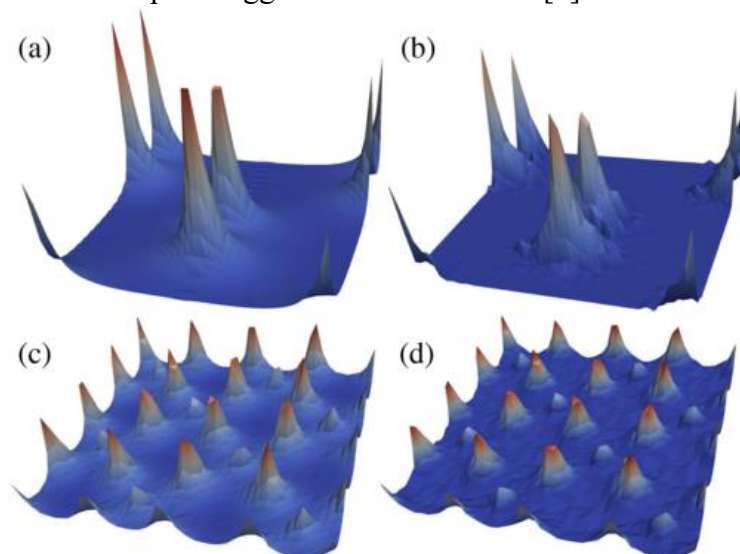


Figure 2. GaAs (a),(b) and spinel (c),(d) projected potential maps. (a),(c) are the ground truth. (b),(d) are recovered from N-beam multiple Bragg scattered intensities via N-phaser [1].

References:

- [1] J Donatelli and J Spence, PRL, **125** (2020), 065502. <https://doi.org/10.1103/PhysRevLett.125.065502>
- [2] J Spence and J Donatelli, Ultramicroscopy, **222** (2021), 113214. <https://doi.org/10.1016/j.ultramic.2021.113214>
- [3] L Sturkey, Proc Phys Soc, **80** (1962), p. 321. <https://doi.org/10.1088/0370-1328/80/2/301>
- [4] J Spence, Acta Crystallogr Sect A, **54** (1998), p. 7-18. <https://doi.org/10.1107/S010876739700874X>
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