Machine Learning Calculation of Large Angle Convergent Beam Electron Diffraction Patterns

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The theory of electron scattering by crystalline materials is well-established, with two main approaches to the calculation of diffracted intensities: the Bloch-wave (or scattering matrix) method, and the Multislice method [1, 2]. Both have been implemented in numerous simulation programs and compute the required large-angle convergent beam electron diffraction (LACBED)patterns. While the accuracy of these simulations can be very high [3] they are also computationally intensive and relatively slow even with modern high-performance computing facilities and graphical processing units (GPUs). In Bloch-wave simulations the limiting step is the inversion of acomplex, non-Hermitian matrix, while for multislice the use of many configurations in the frozen-phonon approximation can increase simulation timesby several orders of magnitude [1, 2]. As a result, simulation times aregenerally several minutes at best. Machine learning (ML) as a computational approach has been gaining prominence in recent years and the implementation of neural networksas universal approximators holds great promise for the solution of inverse and/or computationally difficult problems. Importantly, once trained, a ML calculation is fast – typically a few milliseconds on a GPU. In the caseof modelling electron scattering, this may allow an increase in speed of 5–6 orders of magnitude. Here, we explore the simulation of electron scattering using a variational autoencoder (VAE) [4]. The VAE takes as an input a 128×128 pixel image of the projected potential of a unit cell of cubic material in the [001] orientationand gives an output of the 000 LACBED pattern of the same size. The VAE is trained end-to-end using 5527 Felix [5] Bloch-wave simulations of cubic materials taken from the inorganic crystal structure database (ICSD).[6] The simulations were split 85:10:5 into training, validation, and test sets. Similarity was quantified using a zero-mean normalised cross correlationloss function Z. [7] The position of features in reciprocal space was fixedby choosing an angular range that varied in inverse proportion with lattice parameter. Using 500 Bloch waves, each simulation typically required~400 seconds to complete running on acluster of 160 cores.The VAE uses convolutional encoder and decoder sub-models, both 2 layers deep, to access a 12-dimensional latent space of encoded LACBED patterns. Calculations on an Nvidia GTX 1080Ti GPU are~3.6×10^5 times faster than a 160-core felix simulation. In this exploratory trial, only 5527 out of the 42879 cubic materials available on the ICSD were used for model training. Even with this very limited training data set, some VAE simulations approach the accuracy of felix, while others only produce a poor approximation. We estimate that training on> 12000 simulations would produce losses Z <5%, giving a similarity equivalent to that between felix and experiment.[8]

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