Poster

Assignment of partial charges with 3D ED

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It is well know that electron interact with matter through the electrostatic potential. In principle, the map resulting from electron diffraction experiments displays the electrostatic potential of the crystalline molecule. For minerals, this fact was demonstrated in the 1990s, pioneered by Spence et al with the development of converged beam electron diffraction (CBED). The rise of 3D ED triggered a boost to the development of electron diffraction. Electron diffraction produces a crystal structure, quite similar to X-ray and neutron diffraction, providing information about the 3D atomic coordinates.

Here we present an experimental method to assign partial charges to individual atoms of the molecule from electron diffraction data. Our method is based on a linear superposition of ionic and atomic scattering factors. We demonstrate that the fraction of ionic scattering factors directly relate to the partial charge of an atom in the crystallographic model. We confirm our result with ED data from a ZSM5, a zeolite of framework MFI with 12 independent T-sites and 26 independent oxygen sites in the asymmetric unit, as well as ED data from L-histidine andD-tyrosine.

Our approach works with data at atomic resolution, i.e., 0.7A - 0.8A. This makes it applicable to a very broad range of chemical compounds, in particular to systems with a complexity that escapes DFT and similar simulations. It is based on refinement with SHELXL, using its concept of free variables, and thus can be easily adapted by any crystallographer familiar with this concept. Not only does our method provide experimental values of the chemically universal concept of partial charges, it also offers the possibility to improve and calibrate algorithms for DFT computations and to scale theoretical results.