Experimentally refined density functional theory on strongly correlated materials

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Quantitative convergent-beam electron diffraction (QCBED) is regarded as the most accurate and precise experimental technique for measuring low-order bonding charge density-sensitive structure factors [1,2], i.e., the Fourier coefficients of the periodic charge distribution in crystalline materials. Density functional theory (DFT) is ubiquitous in materials science as a theoretical means of calculating electron distribution and thus making predictions about the properties of materials. Here, we combine both techniques to produce a form of DFT where the approximations it uses are directly refinable by QCBED.

The intersection of QCBED and DFT is the set of structure factors that are required by QCBED to calculate a theoretical diffraction pattern to be matched with an experimental one. DFT-generated structure factors can be used to fulfill this requirement. Different approximations used in DFT calculations will result in different theoretical charge distributions and therefore, different structure factors which are then used as input for QCBED.

In this work, we examine the strongly correlated electron material of nickel oxide (NiO). Strongly correlated electron materials have proved challenging for DFT to model accurately and in order to do so, the local density approximation is coupled with a Hubbard term (LDA+U [3]). In demonstrating our combination of QCBED+DFT, we refine the Hubbard energy term, U, which describes the Coulomb interaction of Ni-localized 3d electrons, and we reveal the sensitivity of the low-order structure factors to this U parameter, and therefore how precisely QCBED can be used to refine it.

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