Inclusion of radiation damage dynamics in HRTEM image simulations

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Computer image simulations are routinely used to interpret experimental high-resolution transmission electron microscopy (HRTEM) images. The variety of well established theoretical methods describe the scattering of electrons by atomic potential, which is the main step in conventional image simulation. It is recognized as well, both in "soft matter" electron microscopy [1, 2] and in the field of nanomaterials [3], that fast electrons of imaging beam in turn cause a modification of atomic structure under observation. Implementation of aberration correction in electron microscopy opened a road for direct experimental imaging of atomic dynamics under the influence of electron beam in controllable conditions with sufficient space and time resolution [4-8]. The ability of AC-TEM to observe the dynamics of individual atoms under the controlled influence of the e-beam provide a mean for direct measurements of diffusion coefficients, cross-sections, chemical constants and o ther characteristics of the dynamic processes that take place at the atomic scale. These advances in experimental AC-TEM techniques require theoretical solutions capable of combined treatment of the dynamic evolution of structures under the e-beam and their subsequent imaging. We propose a computational framework, which combines an event driven molecular dynamics (MD) approach similar to kinetic Monte-Carlo for prediction of the path and the rate of structure evolution under e-beam, MD simulations and DFT for structure refinement, MD for generation of temperature induced conformations ("frozen phonons") and multislice for image simulations. Both simulation parts (atomic dynamics caused by fast electrons and scattering of electrons by atomic potential) are linked via electron energy and electron dose rate. This approach has a potential to incorporate a variety of processes induced in a sample by high-energy electrons such as atom removal by a direct knock-on, atom rearrangements, ionization and consequent chemical reactions. As a proof of principle, in order to demonstrate the feasibility and simplicity of the proposed approach, we select a system in which direct knock-on damage is the principal effect of electron irradiation - a recently observed process of structural transformation of a small graphene flake into a fullerene cage in HRTEM.

We show that proposed approach reproduces the path of the flake evolution into the fullerene cage, the rate of this evolution and the signal-to-noise ratio for given experimental conditions. By varying experimental conditions under simulation (accelerating voltage and dose rate) we conclude that observation of this process at higher beam energies would be hardly possible due to fast evolution rate or low signal-to noise ratio. The proposed approach adds a new dimension to image simulation concept as it includes robustness of a sample as one of the input parameters. Thus, the HRTEM imaging conditions can be evaluated and optimized not only in respect to the instrument, but also in respect to the sample or the process under study.

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