Quantitative High-Resolution TEM/STEM and Diffraction

IM.1.P037
Atomic structure from low-dose exposures via maximum likelihood reconstructions - A new route to circumvent radiation damage

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Keywords: radiation damage, maximum-likelihood reconstruction, transmission electron microscopy

The recent years have seen remarkable developments in electron microscopy, such that essentially all atomic distances can be resolved by the latest generation of instruments. In particular, the correction of lens aberrations [1,2] has not only pushed spatial resolution well below atomic distances, but also reduced delocalization effects which simplifies the analysis on the level of single atoms. However, increased resolution inevitably requires higher doses on the sample. Radiation damage is a key limitation to the applicability of TEM-based methods, and becomes increasingly relevant also in the study of materials. Atomically resolved images with a reasonable signal-to-noise ratio in a TEM or STEM require on the order of 10^5 high-energy electrons to be passed through every square angstrom of the sample, and only a tiny fraction of known substances survive these extreme conditions undisturbed [3]. When knock-on damage dominates the radiation damage, low-voltage imaging is a viable solution [4,5]. However, lower voltages also increase the electron-electron scattering cross section, which leads to increased ionization damage. While reduced electron energies have enabled the atomically resolved imaging of low-dimensional materials made of light elements [6-8], significant beam-driven dynamics are clearly still present in these studies. An alternative route to circumvent radiation damage is to distribute the dose over many identical copies of the same object. This approach has been under active development for decades for the study of biological molecules [9-11] but it has never before been explored in the context of materials science. Here, we show that it is possible to exploit the multiplicity of identical configurations also in the study of non-periodic atomic configurations (e.g., defects) in materials by statistical means. The big advance of this algorithm is that it does not require the location, classification or alignment of the individual entities in the noisy data set. This new approach has the potential to overcome the limitations set by radiation damage, by reducing the dose on the sample by many orders of magnitude. It can be applied to any case where a discrete set of deviations from a regular lattice can be expected to occur repeatedly across a larger sample area. Examples of such situations encompass point defects in a material or 2D material (shown here), as well as functional groups and small molecules on the surface of a thin membrane. The presented method can be considered as a generalization of the approach in Ref. [10], with the addition of unknown object positions. The data is collected by imaging a larger area of the sample with a very low dose, where the signal-to-noise ratio in the images may be well below what is needed to recognize individual objects (defects). Then, by automatically constructing candidate structures, which evolve to maximize the probability of obtaining the observed data via low-dose exposures, the underlying configurations are revealed without a need for image alignment. An example is shown in Fig. 1. Panel a shows a noise-free case (infinite dose) of a simulated STEM image of graphene with a mixture of three different di-vacancy defects randomly distributed across the sample. Fig. 1b shows a similar simulation, but with Poisson noise corresponding to a dose of only 500 $e^-/Å^2$ on the sample. Simulated noisy data corresponding to an area of 3.2 μm^2 was generated, and split into frames of ca. 1.2nm × 1.2nm. We started with model exposures of a defect free lattice with some added noise (to ensure that different initial structures were not numerically identical). Next, the likelihood value of the model set given the low-dose data was calculated as given by Eq. 1, where the product is over all frames (index i), and the summation runs over all models m and spatial offsets xo, yo. $p(I|M)$ is the probability of recording the image data (I) frame i, given model structure M_m with a position offset xo,yo. w_m is a weight associated with each model structure. The model images and their weights are next modified step by step on a single-pixel level, and every change that increases the likelihood value is kept. Remarkably, this procedure leads to exactly the same configurations that were incorporated in the model structure. No *a priori* knowledge about the defect structures is required. We have successfully tested this method with simulated TEM and STEM data both for defects in graphene and small molecules adsorbed on a graphene sheet. For the next step, i.e., experimental realization, a low dose acquisition approach is needed, with a precision sufficient for atomic resolution.

We point out that the limiting dose and defect density, for which this approach still works, depend on the computing power available for the reconstruction, rather than the to-be expected experimental limitations. Hence, further improvements (dose reduction below values demonstrated here) appear feasible with improved algorithms or the use of larger computing resources.

Equation 1:
$$L=\prod_i \sum_m \sum_{xo,yo} p(I_i|M_m) w_m \quad (1)$$

1. M. Haider et al., *Nature* 392 (1998), p. 768.
2. O. Krivanek, *Ultramicroscopy* 78 (1999), p. 1.
3. R. F. Egerton, *Microscopy research and technique* 75 (2012), p. 1550.
4. R. F. Egerton et al., *Ultramicroscopy* 110 (2010), p. 991.
5. J. Meyer et al., *Physical Review Letters* 108 (2012), 196102.
6. K. Suenaga, *Nature nanotechnology* 6 (2007), p. 358.
7. J. C. Meyer et al., *Nano letters* 8 (2008), 3582.
8. O. Krivanek et al., *Ultramicroscopy* 110 (2010), p. 935.
9. J. Frank, "Three-dimensional electron microscopy of macromolecular assemblies: visualization of biological molecules in their native state". Oxford University Press, 2006, p. 410.
10. P. Schwander, D. Giannakis, and A. Ourmazd, *Optics Express* 20 (2012), p. 12827.
11. N.-T. D. Loh and V. Elser, *Physical Review E* 80, (2009), 026705.
12. C. T. Koch, Determination of core structure periodicity and point defect density along dislocations. Dissertation, Arizona State University 2002, p. 32.

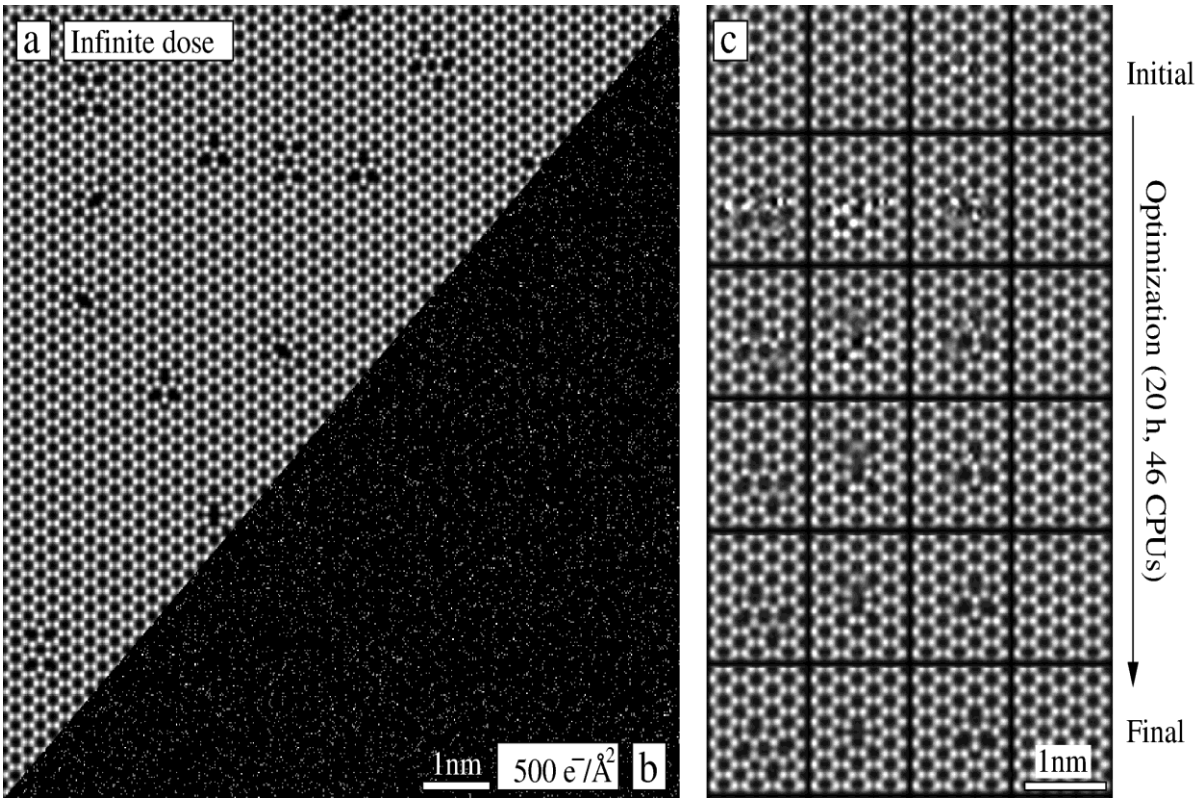

Figure 1. (a) Simulated STEM image (MAADF conditions [8], using QSTEM software [12]) of graphene with di-vacancy defecs (mixture of three different types with all possible orientations at random locations). **(b)** The same simulation for a dose of only 500 $e^-/Å^2$: Most pixels are empty (black) or show a single electron arriving at the ADF detector (gray). A total area of 3.2 μm^2 was simulated. **(c)** Maximum-likelihood reconstructions from the data set, showing 4 out of 20 model images used in the calculation (the others arrived at rotated versions of the defects or empty lattice). All defect configurations were recovered without any *a priori* knowledge, with the low-dose images being the only required input