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Persistent misconceptions about incoherence in electron microscopy

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ABSTRACT

Incoherence in electron microscopic imaging occurs when during the observation the microscope and the object are subject to fluctuations. In order to speed up the computer simulation of the images, approximations are used that are considered as valid. In this paper we will question the validity of these approximations and show that in specific cases they can lead to erroneous results.

It is shown in particular in the case of one single vibrating atom that the thermal diffuse scattering that causes the signal in HAADF STEM is not only dependent on Z but also on the mean square displacement of the atom so that it can even be large for light atoms in soft matter, provided the right HAADF aperture is used.

In HREM imaging the diffuse scattering leaks out of the coherent (elastic) wave and is redistributed in the background. This might explain the mismatch in elastic contrast (Stobbs factor) especially for crystals with a thickness beyond the extinction distance, where also the HAADF signal saturates and the elastic (coherent) component vanishes.

1. Introduction

An electron microscope is in principle a coherent imaging device in which the image is formed by a coherent electron wave that propagates from the source, through the object and the electron lenses to the detector. But during the recording of the image (or the diffraction pattern) both the instrument and the object can be subject to fluctuations which have an effect on the imaging process. This is called incoherence.

The only correct way to describe and simulate these effects is by repetitive calculations of the coherently formed images for each of the different states of the microscope and of the object and subsequently add all the calculated intensities at the level of the detector. Usually this has a deteriorating effect on the image quality. The term "partial coherence" is somewhat misleading since it gives the impression that it describes a situation somewhere between coherence and incoherence, but in reality it remains a purely incoherent superposition of imaging conditions in which the coherent details in the image are not completely destroyed.

Repetitive calculations over all possible states of microscope and object can be prohibitively time consuming. For this reason one often tries to handle the problem by performing the averaging over the fluctuations in the object and the microscope independently so as to yield an "effective" averaged object coherently imaged by an "effective" averaged microscope.

Usually the intuitive feeling is that such approximation is valid for most experimental conditions.

However, it is the purpose of this paper to show that in many cases it can lead to erroneous results that can have a consequence in the interpretation of the experiments.

2. Incoherence in HREM and STEM

We will now consider two different imaging modes in the electron microscope, HREM and STEM.

In HREM, the intensity in the image plane is given by [2]

$$I_{\mathrm{HREM}}(\mathbf{r}) = |\psi(\mathbf{r}) * P(\mathbf{r})|^2 \tag{1}$$

where $*$ represents convolution, $\psi(\mathbf{r})$ is the exit wave of the object and $P(\mathbf{r})$ is the complex point spread function of the electron microscope. If the electron microscope is subject to fluctuations during the recording of the image, we have to average the image intensity over the various states of the microscope, we can write this average as $\langle\ \rangle_M$. But also the object can be subject to variations during the recording of the image so that the intensity has to be averaged over the different states of the object. We will write this average as $\langle\ \rangle_O$. We can also assume that the states of the microscope and of the object are uncorrelated. Hence the total averaged intensity is given by

$$I(\mathbf{r}) = \langle\langle |\psi(\mathbf{r}) * P(\mathbf{r})|^2 \rangle_O \rangle_M \tag{2}$$

Since the fluctuations of microscope and object are independent, the averages over M and O are commutative.

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As mentioned above, the numerical calculation of these averages requires a repetitive calculation for the coherent image intensity over all the states of microscope and object which is very time consuming. Therefore, the usual way to speed up this process is by performing the averages of the microscope and the object separately

$$\tilde{I}(\mathbf{r}) = |\langle\psi(\mathbf{r})\rangle_O * \langle P(\mathbf{r})\rangle_M|^2 \tag{3}$$

In this way the point spread function of the microscope is replaced by an effective point spread function $\langle P(\mathbf{r})\rangle$ and the object wave is replaced by an effective object wave $\langle\psi(\mathbf{r})\rangle$. Then (3) can be considered again as a coherent imaging process where an "averaged" microscope images an "averaged" object. Since both microscope and object become much "smoother" the Nyquist criterium enables us to perform the image simulations with much less sampling points and larger slice thickness, to approximate the atom scattering factors by Gaussian functions, to limit the number of images in focal series exit wave simulations and so on. However, it is clear that

$$\langle\langle|\psi(\mathbf{r}) * P(\mathbf{r})|^2\rangle_O\rangle_M \neq |\langle\psi(\mathbf{r})\rangle_O * \langle P(\mathbf{r})\rangle_M|^2 \tag{4}$$

so that the approximation (4) may be inaccurate which can have an important consequence. In more sophisticated simulation programs the average over the microscope fluctuations is carried out correctly but the averaging over the object fluctuations is still approximate

$$I_{\mathrm{HREM}}(\mathbf{r}) = \langle|\psi(\mathbf{r}) * \langle P(\mathbf{r})\rangle_O|^2\rangle_M \tag{5}$$

We will discuss this further in Section 4.

In incoherent HAADF STEM imaging [2] the image intensity is given by

$$I_{\mathrm{STEM}}(\mathbf{r}) = |\psi(\mathbf{r})|^2 * |P(\mathbf{r})|^2 \tag{6}$$

One now similarly has for the averaging over microscope and object fluctuations

$$I_{\mathrm{STEM}}(\mathbf{r}) = \langle\langle|\psi(\mathbf{r})|^2 * |P(\mathbf{r})|^2\rangle_O\rangle_M \tag{7}$$

$$= \langle|\psi(\mathbf{r})|^2\rangle_O * \langle|P(\mathbf{r})|^2\rangle_M \tag{8}$$

and one can similarly speed up the calculation by approximation

$$\tilde{I}_{\mathrm{STEM}}(\mathbf{r}) = |\langle\psi(\mathbf{r})\rangle_O|^2 * |\langle P(\mathbf{r})\rangle_M|^2 \tag{9}$$

and also in this case

$$\langle\langle|\langle\psi(\mathbf{r})\rangle|^2 * |\langle P(\mathbf{r})\rangle|^2\rangle_O\rangle_M \neq |\langle\psi(\mathbf{r})\rangle_O|^2 * |\langle P(\mathbf{r})\rangle_M|^2 \tag{10}$$

so this approximation may be inaccurate and lead to incorrect interpretations.

3. Fluctuations of the object

In order to make our point very clear, we will describe it by way of the simplest EM experiment possible: the imaging of one single atom.

The incident electron interacts with the electrostatic potential of the atom $V(\mathbf{r})$. For fast electrons it is a valid assumption to state that the electron sees only the projected electrostatic potential of the atom. Hence $V(\mathbf{r})$ represents the projected electrostatic potential and $\mathbf{r}$ is the position of vector of the atom in the plane perpendicular to the electron beam. It is the purpose of any imaging mode (TEM, STEM) of the electron microscope to visualise this atom potential as faithfully as possible in order to determine both the position of the atom and the interaction potential (atom type).

As model for the changes in the structure of the object we will assume that the atom is moving as function of time around its equilibrium position.

In principle we can thus simulate the effect of the atom motion by repetitive coherent simulations for all atom positions independently and add the images afterwards. In crystals, the atoms move collectively in so called phonons. Electron diffraction at moving atoms is therefore called phonon scattering. In HAADF STEM phonon scattering constitutes the dominant signal so that quantitative agreement with simulations can only be obtained by integrating simulated intensities using tedious repetitive multislice calculations for different frozen phonon configurations [3–6].

However, this leads to an apparent conflict between two different views. The multislice programs treat the interaction between the electron and the rigorously displaced atoms as an elastic (coherent) scattering process in which the scattered wave still interferes with the unscattered wave. The other view is based on the "which way" reasoning of Feynman [1] in which inelastic phonon scattering exchanges energy with the crystal and by altering the crystal state, the interference with the non-interacting wave is destroyed. This then leads to the paradox why inelastic phonon scattering, which dominates the HAADF-STEM signal, can be calculated accurately by means of an "elastic" multislice calculation. This paradox is solved in several papers [7–9] in which it is shown that the frozen phonon approach is fully equivalent to a rigorous inelastic scattering treatment based on the Yoshida theorem. In the former, the interference is destroyed by thermal averaging and in the latter it is destroyed by the orthogonality of the crystal state but the consequences are formally and mathematically the same. Hence we can further use the frozen atom model since it is the simplest model for inelastic scattering and is intuitively much more transparent for our purpose.

In order to make our point even more clear without losing the essence, we will assume as in [10] that the object is a weak phase object, so that the electron wave can be approximated as

$$\psi(\mathbf{r}(t)) = 1 + iV(\mathbf{r}(t)) \tag{11}$$

where $V(\mathbf{r}(t))$ is proportional to the projected potential of the object, t is the time dependence. For simplicity we will omit constant factors. We will now separate the potential in a time averaged and a time dependent part as suggested in [11,12]

$$V(\mathbf{r}(t)) = \langle V(\mathbf{r}(t))\rangle_t + W(\mathbf{r}(t)) \tag{12}$$

so that

$$\langle W(\mathbf{r}(t))\rangle_t = 0 \tag{13}$$

where $\langle\ \rangle_t$ denotes averaging over time. It is a special but representative case of averaging over the possible object fluctuations $\langle\ \rangle_O$ as described in Eq. (2).

Fourier transformation to diffraction space now yields for (11) and (12)

$$\psi(\mathbf{g},t) = \delta(\mathbf{g}) + iV(\mathbf{g},t) \tag{14}$$

$$\psi(\mathbf{g},t) = \delta(\mathbf{g}) + i\langle V(\mathbf{g},t)\rangle_t + iW(\mathbf{g},t) \tag{15}$$

with

$$\langle W(\mathbf{g},t)\rangle_t = 0 \tag{16}$$

For the intensity at $\mathbf{g} \neq 0$ we then have

$$I(\mathbf{g}) = |V(\mathbf{g},t)|^2 = |\langle V(\mathbf{g},t)\rangle_t + W(\mathbf{g},t)|^2 \tag{17}$$

Since $\langle|V(\mathbf{g},t)|\rangle$ is independent of t, integration over time then yields, using (16)

$$\langle I(\mathbf{g})\rangle_t = \langle I(\mathbf{g},t)\rangle_t = |\langle V(\mathbf{g})\rangle_t|^2 + \langle|W(\mathbf{g},t)|^2\rangle_t \tag{18}$$

In the case of a perfect crystal $|\langle V(\mathbf{g},t)\rangle_t|^2$ represents the Bragg scattering and $\langle|W(\mathbf{g},t)|^2\rangle_t$ the thermal diffuse scattering background [3].