Ultramicroscopy 134 (2013) 102–112

Contents lists available at ScienceDirect

Ultramicroscopy

journal homepage: www.elsevier.com/locate/ultramic

The influence of inelastic scattering on EFTEM images—exemplified at 20 kV for graphene and silicon

Z. Lee *, H. Rose, R. Hambach, P. Wachsmuth, U. Kaiser

Universität Ulm, Materialwissenschaftliche Elektronenmikroskopie, Albert-Einstein-Allee 11, 89081 Ulm, Germany

ARTICLE INFO

Available online 7 June 2013

Keywords:
EFTEM
Inelastic scattering
Zero-loss
Low-loss
Mutual coherence
Low voltage

ABSTRACT

We present model-based image simulations for zero-loss and plasmon-loss filtered images at 20 kV for graphene and silicon based on the mutual coherence approach. In addition, a new approximation for the mixed dynamic form factor is introduced. In our calculation multiple elastic scattering and one inelastic scattering are taken into account. The simulation shows that even the intensity of zero-loss filtered image is attenuated by the interference between inelastically scattered waves. Moreover, the intensity of plasmon-loss filtered images cannot be neglected, either.

1. Introduction

The standard simulation and interpretation of TEM images are based on the elastic scattering theory. This approximation is sufficient for most thin crystalline objects imaged at voltages larger than about 100 kV. In these cases, phase contrast dominates and the contribution of inelastic scattering to the total contrast is negligibly small. However, due to radiation damage caused by atom displacement at higher accelerating voltages >100 kV, the use of lower voltages (20–80 kV) becomes necessary as realized within the frame of the SALVE (Sub-Angstrom Low-Voltage Electron microscopy) project [1]. As the voltage drops down, the wavelength increases resulting in a decrease of resolution for a fixed usable aperture angle determined by the aberrations of the imaging system. Thanks to the new generation of C_s/C_c- corrector [2], transmission electron microscopes can now reach sub-Angstrom resolution down to about 60 kV. The novel SALVE corrector provides a usable aperture angle of about 50 mrad, which is supposed to lower the resolution limit to 1.7 Å at 20 kV and to 1.2 Å at 40 kV [3].

When the accelerating voltage is decreased to as low as 20 kV, all objects are strong scatterers [4]. In addition, the elastic model is not sufficient anymore and inelastic scattering must be taken into account especially for low-Z objects. As a result, the interpretation of the images becomes rather involved. In the case of inelastic scattering, the incident electron changes the initial state of the object to any allowed excited state. The initial wave function $\psi_t=\psi_0|0\rangle$ of the total system is the product of the wave function ψ_0 of the incident electron and the wave function $|0\rangle$ of the ground state of the object. After the scattering, the total wave function of the system does not factorize anymore and adopts the entangled form

$$\psi_t=\sum_{j=0}^{\infty}\Psi_j|j\rangle. \tag{1}$$

The interaction of the incident electron with the particles of the object results in a transition of the object state from the ground state $|0\rangle$ to an excited state $|j\rangle$ and a change of the wave function of the incident electron from ψ_0 to Ψ_j. For $j=0$, the scattering process is elastic and

$$\Psi_0=\psi_0+\psi_e \tag{2}$$

is the sum of the incident wave ψ_0 and the elastically scattered wave ψ_e. Inelastic scattering is incorporated into the image simulations by means of the Mixed Dynamic Form Factor (MDFF), introduced by Rose [5,6]. The MDFF accounts for the interference of different scattered partial electron waves. The elastically scattered partial waves can interfere with each other, whereas the partial waves of the inelastically scattered electron can only interfere with each other if they are associated with the same excited object state. A detailed discussion is presented in Section 2.1. Schattschneider [7] introduced the density matrix approach as an alternative method. The difference between the two approaches is that the MDFF describes the interference of the scattered waves in reciprocal space, whereas the density matrix method handles it in real space. The MDFF involves the coupling of two waves. Therefore the image calculation involves 4D Fourier transforms. The numerical calculation of 2D Fourier transform is usually performed very rapidly by applying the fast Fourier transform (FFT). The number of arithmetic operations required for the FFT on a $N\times N$ matrix is $2N^2\log_2 N$; however, for a $N\times N\times N\times N$ array, the number of operations increases to $4N^4\log_2 N$, which demands

* Corresponding author. Fax: +49 731 5022958.
E-mail address: zhongbo.lee@uni-ulm.de (Z. Lee).

0304-3991/$ - see front matter © 2013 Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.ultramic.2013.05.020

$2N^2$ times of computational expenditure compared with the 2D case. The image calculation for thick objects usually involves the multislice algorithm, and the computational task required for the propagation of the 4D array through all the slices will be too time-consuming.

In order to tackle the 4D problem in a computation-efficient way, different methods for the factorization of MDFF or density matrix have been proposed with different applications [7–17, 19–22]. Within the core-loss range where the scattering is highly localized, Stalknecht and Kohl [8] as well as Navidi-Kasmai and Kohl [9] proposed the calculation of the density matrix elements based on the first-order perturbation theory combined with Bloch-wave function. Schattschneider employed the dipole approximation [7] and this approximation was applied for the calculation of EFTEM images by Verbeeck et al. [10]. Dwyer et al. [11,12] calculated the density matrix elements for atomic ionization based on the work of Saldin [13]. Lugg et al. [14] computed the bound-state wave based on a relativistic Hartree–Fock model. Löffler et al. [15] introduced the method of matrix diagonalization. For the low-loss range where the inner shell structure is neglected, Müller et al. [16,17] utilized Bessel functions for the factorization of the MDFF obtained by employing the Raman–Compton approximation [18]. In [19–22] the MDFF was calculated by using precise wave functions. The result applies for all energy-losses, but at the sacrifice of efficiency.

This paper concentrates on the image simulation for the low-loss range and our simulation is based on the multislice mutual coherence method outlined in [16,17]. In addition, we introduce a new approximation for the MDFF. This approximation keeps the maximum similarity with the MDFF function, obtained by utilizing the Raman–Compton model [18] and the Wentzel model [23] for the atom potential. Our approximation can be applied to different imaging conditions, without loss of computational efficiency.

2. Theory

2.1. Coherence and incoherence

Coherence indicates that there is a fixed phase relation between the waves (wavepackets), so that the waves(wavepackets) can interfere with each other. Incoherence indicates that there is no fixed phase relation between the waves (wavepackets), and the observable intensity is a summation of the intensities of the waves (wavepackets).

The eigenstates of the object are mutually orthogonal, which leads to

$$\langle m|j\rangle = \delta_{mj}. \tag{3}$$

Therefore the observable intensity of the system in Eq. (1) can be written as

$$\begin{aligned}\psi_t^*\psi_t &= \left(\sum_{m=0}^{\infty}\langle m|\Psi_m^*\right)\left(\sum_{j=0}^{\infty}\Psi_j|j\rangle\right) = \sum_{m,j}^{\infty}\Psi_m^*\Psi_j\delta_{mj}\\ &= \sum_{j=0}^{\infty}|\Psi_j|^2 = \sum_{j=1}^{\infty}|\Psi_j|^2 + |\psi_0+\psi_e|^2.\end{aligned} \tag{4}$$

Since the intensity $\psi_t^*\psi_t$ is the superposed intensity contributed by each scattered partial wave corresponding to different object eigenstates, the conclusion is that the scattered waves are incoherent with each other as long as the coupled object states are different. The second term implies that the elastically scattered wave ψ_e can interfere with the non-scattered wave ψ_0.

For elastic scattering, the incident wave function ψ_0 propagates through the object. However, a pure wave function cannot be applied for the description of imaging process involving inelastic waves because inelastic waves are always coupled with the corresponding object states. Rose was inspired by the concept of mutual coherence function applied in optics, and extended its usage to the handling of the wave propagation in the electron microscope [5]. Mutual coherence function accounts for the spatial and temporal interference between the waves which correspond to the same object state. Unlike the pure wave function, the propagation of the mutual coherence function Γ_0 preserves the object information as well as the amplitude and phase resulting from wave interference.

$$\Gamma_0 = \langle \Psi^*(\vec{r}\,', t)\Psi(\vec{r}, t-\tau)\rangle_T. \tag{5}$$

Here τ is the temporal difference between the two waves, and $\vec{r}\,'$ and $\vec{r}$ indicate that the two waves originate from different sources. T represents the time average.

2.2. The concepts applied for the image calculation involving both elastic and inelastic scattering

In order to clarify the concepts utilized for the image calculation involving both elastic and inelastic scattering, the counterparts used for pure elastic scattering are listed in Table 1, exemplified by a thin sample.

- MCF – Mutual Coherence Function.
- MOT – Mutual Object Transparency.
- POA – Phase Object Approximation.

The time-averaged phase factor $\langle\chi\rangle$ is proportional to the static projected atomic potential defined as

$$\chi(\vec{\rho}) = -\frac{1}{\hbar v}\int_{-\infty}^{\infty}\sum_{j=1}^{n} V_j(\rho, z')\,dz', \tag{6}$$

where $\hbar$ is the Planck constant, v is the velocity of the electron and $V_j(\rho, z')$ is the position-dependent atomic potential of the jth atom.

The Taylor expansion of the MOT to the second order of χ results in Eq. (7), [6]

$$\gamma(\vec{\rho}, \vec{\rho}\,', \tau) \approx \exp[i\mu_1(\vec{\rho}) - i\mu_1(\vec{\rho}\,') - \tfrac{1}{2}\mu_2(\vec{\rho}) - \tfrac{1}{2}\mu_2(\vec{\rho}\,') + \mu_{11}(\vec{\rho}, \vec{\rho}\,', \tau)] \tag{7}$$

with the definitions

$$\mu_1(\vec{\rho}) = \langle\chi(\vec{\rho}, t)\rangle, \tag{8}$$

$$\begin{aligned}\mu_{11}(\vec{\rho}, \vec{\rho}\,', \tau) &= \langle\chi(\vec{\rho}, t)\chi(\vec{\rho}\,', t-\tau)\rangle - \langle\chi(\vec{\rho}, t)\rangle\langle\chi(\vec{\rho}\,', t-\tau)\rangle\\ &= \sum_{n\neq m} P_m e^{i\omega_{nm}\tau}\langle m|\chi(\vec{\rho}\,')|n\rangle\langle n|\chi(\vec{\rho})|m\rangle\\ &= \left(\frac{\alpha_s}{\pi\beta}\right)^2 \iiint e^{-i\omega\tau}\frac{S(\vec{K}, \vec{K}', \omega)}{K^2\,K'^2} e^{i\vec{K}\vec{\rho}} e^{-i\vec{K}'\vec{\rho}\,'}\, d\omega\, d^2\vec{K}_\rho\, d^2\vec{K}'_{\rho'},\end{aligned} \tag{9}$$

$$\mu_2(\rho) \approx \langle\chi^2(\rho, t)\rangle - \langle\chi(\rho, t)\rangle^2 = \mu_{11}(\rho = \rho', \tau = 0). \tag{10}$$

Here $\alpha_s = 1/137$ is the Sommerfeld constant, $\beta = v/c$ and c is the velocity of the light. The function μ_1 represents the static projected potential. The variable μ_2 accounts for the decrease of the intensity of the incident MCF caused by the reduction of the purely elastically scattered electrons; therefore μ_2 can be interpreted as the absorption potential. The factor μ_{11} accounts for the increase of

Table 1
The concepts utilized for the description of elastic/inelastic scattering and pure elastic scattering.

Status	Elastic/Inelastic	Pure elastic
Incoming	$\Gamma_0 = \langle\Psi_0^*(\vec{r}\,', t)\Psi_0(\vec{r}, t-\tau)\rangle_T$ (MCF)	$\Psi_0(\vec{r})$ (pure)
Target	$\gamma = \langle e^{i\chi(\vec{\rho}, t)}e^{-i\chi(\vec{\rho}\,', t-\tau)}\rangle$ (MOT)	$e^{i\chi(\vec{\rho})}$ (POA)
Outgoing	$\Gamma_f = \Gamma_0\cdot\gamma$ (MCF)	$\Psi_0 e^{i\chi}$ (pure)