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Image simulation for electron energy loss spectroscopy

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Abstract

Aberration correction of the probe forming optics of the scanning transmission electron microscope has allowed the probe-forming aperture to be increased in size, resulting in probes of the order of 1 Å in diameter. The next generation of correctors promise even smaller probes. Improved spectrometer optics also offers the possibility of larger electron energy loss spectrometry detectors. The localization of images based on core-loss electron energy loss spectroscopy is examined as function of both probe-forming aperture and detector size. The effective ionization is nonlocal in nature, and two common local approximations are compared to full nonlocal calculations. The affect of the channelling of the electron probe within the sample is also discussed.
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1. Introduction

Electron energy loss spectroscopy (EELS) is a valuable tool in materials science, providing both elemental mapping and local bonding information via near edge structure. The delocalization of the EELS interaction potential has been broadly discussed, see for example Egerton's book and references therein (Egerton, 1996). Many descriptions of the delocalization of the EELS interaction use a single interaction width, such as a classical impact parameter. The effective core-loss ionization interaction is however nonlocal in nature and not easily described in terms of a single variable (Allen and Josefsson, 1995). This is particularly the case for diffracting samples or scanning transmission electron microscopy (STEM), where EELS image formation depends on the interference of different Fourier components of the incident electron wave function.

In this paper EELS image localization and formation is examined, with particular reference to STEM imaging. Recent advances in aberration correction have allowed resolution in the STEM of less than 1 Å in the case of annular dark field (ADF) imaging (Nellist et al., 2004). The next generation of aberration corrected machines promises resolution of near 0.5 Å (TEAM Project, 2000). The localization of the EELS interaction for STEM has been examined by considering the width of single atom images (Kohl and Rose, 1984; Cosgriff et al., 2005). The core-loss EELS image width is a complicated function of binding energy, probe size and detector geometry.

These results however ignore the channelling of the incident electrons within the sample. In this paper the process of STEM image formation based on core-loss EELS is examined using an optical potential formulation, with absorption due to thermal diffuse scattering (TDS) included as an imaginary term in the potential. The presence of heavy columns with the crystalline sample leads to large angle scattering of the incident electron beyond the EELS detector, as well as an attenuation of the elastic intensity. The 'focussing' of the electron probe by the atomic columns is also considered.

2. Theory

A general expression for the inelastic cross section of fast electrons incident on a sample of thickness t and cross-sectional area A may be written as (Allen and Josefsson, 1995; Allen et al., 2006)

$$\sigma(\vec{\Gamma}) = \frac{2\pi m}{h^2 k}\int_0^t \int_A \int_A \psi_0^*(\vec{\Gamma}, \vec{r}_\perp, z) W(\vec{r}_\perp, \vec{r}'_\perp) \times \psi_0(\vec{\Gamma}, \vec{r}'_\perp, z)\, \mathrm{d}\vec{r}_\perp\, \mathrm{d}\vec{r}'_\perp\, \mathrm{d}z. \quad (1)$$

The vector $\vec{\Gamma}$ serves as a place holder for the relevant variables for a given experimental procedure. For example, for plane-wave illumination, where the incident beam is tilted with respect to the sample it describes the incident electrons wave

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vector $\vec{k}$. For the case of STEM it describes probe position $\vec{R}$. In Eq. (1), m is the relativistic electron mass, h is Planck's constant, $k = |\vec{k}|$ and $\psi_0(\vec{\Gamma}, \vec{r}_\perp, z)$ describes the elastic wave function of the incident electron within the sample.

The term $W(\vec{r}_\perp, \vec{r}'_\perp)$ is an effective nonlocal potential, in the projected potential approximation, and describes the inelastic scattering process of interest, for example EELS and energy dispersive X-ray analysis (EDX). It may be written in the form (Oxley et al., 2005),

$$W(\vec{r}_\perp, \vec{r}'_\perp) = \frac{h^2 k}{2\pi m A t} \sum_{\vec{h},\vec{g}} \mu_{\vec{h},\vec{g}} \exp(2\pi i \vec{h} \cdot \vec{r}_\perp) \times \exp(-2\pi i \vec{g} \cdot \vec{r}'_\perp), \quad (2)$$

where $\vec{h}$ and $\vec{g}$ are variables in the Fourier transform space. For inner-shell ionization from atom type β the inelastic scattering coefficients $\mu_{\vec{h},\vec{g}}$ are calculated using (Allen and Josefsson, 1995)

$$\mu_{\vec{h},\vec{g}} = \frac{1}{2\pi k V_c} \sum_j \exp[-M_{\beta_j}(\vec{g} - \vec{h})] \times \exp[2\pi i(\vec{g} - \vec{h}) \cdot \vec{\tau}_{\beta_j}] f(\vec{h}, \vec{g}). \quad (3)$$

The sum over j includes all atoms of type β within the unit cell of volume V_c. The Debye–Waller factor $M_\beta(\vec{g} - \vec{h})$ accounts for the thermal motion of the target atoms. The atomic scattering form factor $f(\vec{h}, \vec{g})$ is given by

$$f(\vec{h}, \vec{g}) = \frac{1}{2\pi^3 a_0^2} \int\int K' \frac{\sum_{i,f} F^*_{i,f}(\vec{Q}_{\vec{h}}, E_f) F_{i,f}(\vec{Q}_{\vec{g}}, E_f)}{|\vec{Q}_{\vec{h}}|^2 |\vec{Q}_{\vec{g}}|^2} \mathrm{d}E_f \, \mathrm{d}\Omega_{K'}, \quad (4)$$

where a_0 is the relativistically corrected Bohr radius. The momentum transfer to the crystal is $h\vec{Q} = h(\vec{K} - \vec{K}')$ where $\vec{K}$ and $\vec{K}'$ are the refraction corrected wave vectors of the incident and scattered electron, respectively. The vector $\vec{Q}_{\vec{h}}$ is defined as $\vec{Q}_{\vec{h}} = \vec{Q} + \vec{h}$. The indices i and f define the initial and final states of the target electron, and E_f is the energy of the ejected electron. For EELS the detector geometry is defined by the integration over $\mathrm{d}\Omega_{K'}$ and $\mathrm{d}E_f$ defines the energy window over which the EELS signal is integrated. For EDX these integrations cover all possible scattering angles and energy losses.

The transition matrix element from the initial state to final state is given by

$$F_{i,f}(\vec{Q}_{\vec{g}}, E_f) = \int u^*_f(E_f, \vec{r}) \exp[2\pi i(\vec{Q}_{\vec{g}} \cdot \vec{r})] u_i(\vec{r}) \, \mathrm{d}\vec{r} \equiv \langle f | \exp[2\pi i(\vec{Q}_{\vec{g}} \cdot \vec{r})] | i \rangle, \quad (5)$$

where $u_i(\vec{r})$ defines the initial state and $u_f(E_f, \vec{r})$ the final state of the target electron, respectively.

We describe $W(\vec{r}_\perp, \vec{r}'_\perp)$ as an effective nonlocal potential because it is a function of two independent real-space vectors and its Fourier components $\mu_{\vec{h},\vec{g}}$ are similarly a function of two independent reciprocal space vectors. Examination of Eq. (1) shows that the cross section is a function of the product of the incident wave function expressed in terms of two different real space coordinates. It is hence not a function of the intensity of the incident wave function alone. It may in this sense be considered a "coherent" cross section, dependant not only on the amplitude, but also the phase of the incident electron wave function as it propagates through the specimen. The expressions given here are a generalization of the expressions of Yoshioka, which in turn are a generalization of the Bethe scattering equations (Allen and Josefsson, 1995; Yoshioka, 1957; Bethe, 1928). Similar expressions have been derived, using different starting points, independently by other authors (Dudarev et al., 1993; Dywer, 2005). The sum over the product of transition matrix elements seen in Eq. (4) is closely related to the mixed dynamical form factor of Rose (1976). The effective nonlocality is implicit in all these formulations.

The cross-section expression in Eq. (1) can be rewritten in reciprocal space form as (Allen et al., 2003),

$$\sigma(\vec{\Gamma}) = \int_t \sum_{\vec{h},\vec{g}} \Psi^*_h(\vec{\Gamma}, z) \Psi_g(\vec{\Gamma}, z) \mu_{\vec{h},\vec{g}} \, \mathrm{d}z. \quad (6)$$

It is useful to consider some special forms of Eq. (6).

2.1. Plane-wave illumination

First let us consider the case where we have plane-wave illumination (wave vector $\vec{k}$) and channelling of the incident electron is not significant. Eq. (6) can then be reduced to the form,

$$\sigma(k) = t|\Psi_0(k)|^2 \mu_{\vec{0},\vec{0}}, \quad (7)$$

which (ignoring some normalization factors) produces results similar to Egerton's programs SIGMAK and SIGMAL (Egerton, 1979, 1981). In previous work this has also been referred to as the kinematic cross section (Allen and Josefsson, 1995). The cross section is a function of only the magnitude of the incident wave vector $k = |\vec{k}|$.

For crystalline samples, diffraction leads to significant contributions to the cross section from Fourier components of the incident electron wave function other than $\Psi_0(\vec{k})$. In this case we rewrite Eq. (6) in the form

$$\sigma(\vec{k}) = \int_t \sum_{\vec{h},\vec{g}} \Psi^*_h(\vec{K}, z) \Psi_g(\vec{K}, z) \mu_{\vec{h},\vec{g}} \, \mathrm{d}z. \quad (8)$$

The Fourier components of the incident electron wave function now become functions of the beam/crystal orientation and the refracted wave vector $\vec{K}$ as well as the depth within the crystal z. For EDX the variation with X-ray yield as a function of beam orientation has been used to develop the method of atom location by channelling enhanced microanalysis (ALCHEMI), see for example Spence and Taftø (1983); Andserson (1997); Oxley et al. (1999a) and references contained within. An example of an EDX cross section as a function of beam orientation, and simulation based on Eq. (8) is shown in Fig. 1.

Similar cross-section variation, as a function of beam orientation, is also observed for EELS, as shown in Fig. 2.

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