

Inversion of dynamical electron scattering to obtain the crystal potential using data from two thicknesses

L. J. Allen,^{a*} C. Koch,^b M. P. Oxley^a and J. C. H. Spence^b

^aSchool of Physics, University of Melbourne, Victoria 3010, Australia, and ^bDepartment of Physics and Astronomy, Arizona State University, Tempe, Arizona 85287, USA. Correspondence e-mail: lja@physics.unimelb.edu.au

A method is given to invert the multiple scattering of electrons in a crystalline slab to obtain the projected potential. For a fixed orientation of the incident beam of electrons, scattering data are required for two thicknesses of the crystal.

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1. Introduction

The inversion of dynamical (multiple) scattering in high-resolution transmission electron microscopy was identified as recently as 1997, in a review article by Smith (1997), as ‘unquestionably the major unresolved problem confronting the field of atomic resolution microscopy’. Solutions to the inversion problem have subsequently been given by Spence (1998) and independently by Allen and co-workers (Allen *et al.*, 1998, 1999, 2000). For a crystalline slab of fixed thickness and working at a fixed incident energy within an N -beam approximation, the exit-surface wavefunction must be determined for a well defined set of N incident-beam orientations. For zone-axis conditions, N is typically of the order of 100. Retrieval of exit-surface wavefunctions using images implies a loss of resolution. Within a linear imaging approximation, it has recently been shown how to extend the resolution by using, in addition, a high-resolution diffraction pattern (Chen *et al.*, 1999). To be useful under multiple scattering conditions, the linear imaging constraint needs to be removed, *e.g.* using a combination of the through-focal series and Gerchberg–Saxton algorithms (Allen *et al.*, 2001). Here we will show that, for a fixed incident energy, the dynamical inversion problem can be solved by obtaining the exit-surface wavefunction for two different thicknesses at a *single* orientation.

2. Theory

The fundamental wave equation that describes the scattering of electrons in a crystalline slab is the Schrödinger equation:

$$[\nabla^2 + U(\mathbf{r}) + K^2]\Psi(\mathbf{r}) = 0. \quad (1)$$

Here $U(\mathbf{r})$ is the potential in the crystal and K is the wavenumber of the incident electrons corrected for refraction, *i.e.* $K^2 = k^2 + U_0$, where U_0 is the mean inner potential. The wavenumber k in vacuum is related to the wavelength of the incoming electrons by $k = 2\pi/\lambda$.

Let us assume that the xy plane coincides with the entrance surface of the crystal and that the electron beam is incident along the z axis. Considering the wavefunction to be a modification of an incident plane wave, we write

$$\Psi(\mathbf{r}) = \exp(i\mathbf{K} \cdot \mathbf{r})\Phi(\mathbf{r}), \quad (2)$$

which has the effect of factoring out the rapid variation in $\Psi(\mathbf{r})$ along the z direction. Writing $\mathbf{r} = (\mathbf{r}_\perp, z)$ and substituting (2) into (1), we obtain

$$[\nabla_\perp^2 + U(\mathbf{r}_\perp, z)]\Phi(\mathbf{r}_\perp, z)|_{(\text{cryst})} = -2iK\partial_z\Phi(\mathbf{r}_\perp, z)|_{(\text{cryst})}, \quad (3)$$

where ∇_\perp operates in the xy plane, ∂_z denotes a partial derivative with respect to z and we have assumed that $\partial_z^2\Phi \simeq 0$ (paraxial approximation). For propagation through free space, the corresponding equation is

$$\nabla_\perp^2\Phi(\mathbf{r}_\perp, z)|_{(\text{free})} = -2ik\partial_z\Phi(\mathbf{r}_\perp, z)|_{(\text{free})}. \quad (4)$$

At the exit surface of the crystalline slab of thickness t , $\Phi(\mathbf{r}_\perp, t)|_{(\text{cryst})} = \Phi(\mathbf{r}_\perp, t)|_{(\text{free})} \equiv \Phi(\mathbf{r}_\perp, t)$. Assuming incident electron energies of hundreds of keV, $K \simeq k$ to high accuracy. Then, using (3) and (4) we obtain

$$U(\mathbf{r}_\perp, t) = -2ik \left[\frac{\partial_z\Phi(\mathbf{r}_\perp, z)|_{(z=t, \text{cryst.})} - \partial_z\Phi(\mathbf{r}_\perp, z)|_{(z=t, \text{free})}}{\Phi(\mathbf{r}_\perp, t)} \right]. \quad (5)$$

We now note the fact that $\nabla_\perp^2 \equiv -\mathcal{F}^{-1}\mathbf{q}_\perp^2\mathcal{F}$, where \mathcal{F} denotes a Fourier transform and \mathbf{q}_\perp is the variable conjugate to \mathbf{r}_\perp in the Fourier space [we have used formula 33.20 in Spiegel (1968)]. Then from (4) we see that we may write

$$\partial_z\Phi(\mathbf{r}_\perp, z)|_{(z=t, \text{free})} = \frac{1}{2ik}\mathcal{F}^{-1}\mathbf{q}_\perp^2\mathcal{F}\Phi(\mathbf{r}_\perp, t). \quad (6)$$

Using (6) and approximating the derivative in the crystal in terms of the wavefunctions at two planes a distance Δt apart, we can rewrite (5) in the form

$$U(\mathbf{r}_\perp, t) = \frac{2ik}{\Delta t} \left[1 - \frac{\Phi(\mathbf{r}_\perp, t + \Delta t)}{\Phi(\mathbf{r}_\perp, t)} \right] + \frac{\mathcal{F}^{-1}\mathbf{q}_\perp^2\mathcal{F}\Phi(\mathbf{r}_\perp, t)}{\Phi(\mathbf{r}_\perp, t)}. \quad (7)$$

This gives the (projected) potential over the slice between t and $t + \Delta t$ (which for a perfect crystal is the projected potential for the crystal). A crucial issue in using (7) to construct the potential is that we must be able to correctly phase the wavefunctions retrieved for thicknesses t and $t + \Delta t$ relative to each other. In retrieving the exit-surface wavefunction at each thickness, the phase can only be determined up to an arbitrary overall constant. Assume that the wavefunction retrieved at $t + \Delta t$ (indicated by a prime) is related to that which is correctly phased relative to the wave function at t by $\Phi'(\mathbf{r}_\perp, t + \Delta t) = \Phi(\mathbf{r}_\perp, t + \Delta t)\exp(-i\alpha)$. For conditions where

absorptive effects are small, the potential $U(\mathbf{r}_\perp, t)$ is real and this implies that we must have

$$\mathcal{R}e \left[\frac{\Phi'(\mathbf{r}_\perp, t + \Delta t) \exp(i\alpha) + i\Delta t / (2k) \mathcal{F}^{-1} \mathbf{q}_\perp^2 \mathcal{F} \Phi(\mathbf{r}_\perp, t)}{\Phi(\mathbf{r}_\perp, t)} \right] = 1. \quad (8)$$

This constraint could be used in an approximate way in the presence of absorption.

Care has to be taken in evaluating the potential at points where $\Phi(\mathbf{r}_\perp, t)$ is small or has zeros. This can be achieved by avoiding thicknesses where such features are evident in the exit-surface images or by using numerical regularization. The difference between the two thicknesses Δt needs to be accurately determined, for example using convergent-beam electron diffraction (Williams & Carter, 1996). We note also that an effective change in thickness of the sample can be obtained by varying the voltage of the incident beam (Rez, 1999).

3. Model example

Let us illustrate the inversion of the multiple scattering using a model example. Consider 200 keV electrons incident along the [110] zone axis in GaAs at room temperature. Figs. 1(a) and (b) show the exit-surface image and phase calculated in an $N = 27$ beam approximation for a thickness of 200 Å. The image and phase for a thickness of 250 Å are shown in Figs. 1(c) and (d), where intensity zeros in the image with corresponding vortices in the phase are evident (this does

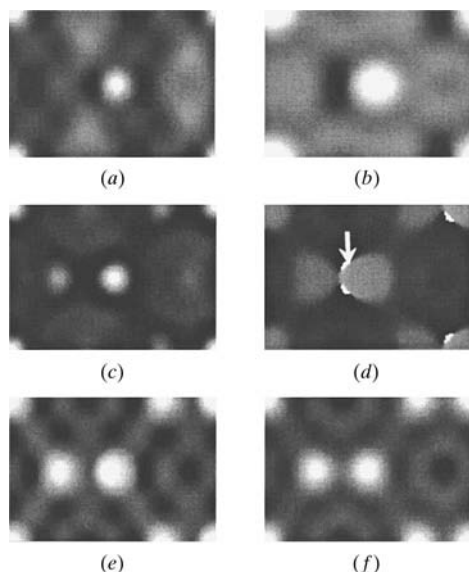


Figure 1

Shown in (a) and (b) are the exit surface image and phase along the [110] zone axis for a 200 Å thick slab of GaAs. The image and phase for a thickness of 250 Å are shown in (c) and (d). The reconstructed potential is shown in (e). The input model potential is shown in (f). The arrow in (d) indicates a vortex in the phase.

not occur at 200 Å). A vortex, joined to a counter rotating partner by a branch line, is indicated by the arrow in (d). We have checked that (8) retrieves α correctly. As Δt increases, a further component in α arises due to inaccuracies in the estimation of the derivative of the wavefunction in the crystal. For example, adding a constant phase of 1.9 rad to the phases in Fig. 1(b), we retrieve a value for α of 1.76 rad. The potential then constructed from these two exit-surface wavefunctions is shown in Fig. 1(e). The input model potential calculated using the 27 Fourier coefficients corresponding to the reciprocal-lattice vectors defining our 27-beam approximation (with maximum magnitude 5.44 \AA^{-1}) is shown in Fig. 1(f) for purposes of comparison. Information on higher-order Fourier coefficients is not retrieved, as is the case in a 27-beam approximation using inversion methods based on a through-tilt series of measurements (Spence, 1998; Allen *et al.*, 1998, 1999, 2000). For $t = 200 \text{ \AA}$, the exit-surface image and the potential do not show similar contrast, due to multiple scattering. However, the potential retrieved using (7) and (8) is in good agreement with the corresponding model potential – there is in fact a damping of the potential (maximum range reduced by 84%), which becomes smaller as Δt is reduced. A reasonable (but further damped) representation of the projected potential is still obtained for $\Delta t = 100 \text{ \AA}$.

4. Conclusions

A method has been given to invert the multiple scattering in many-beam dynamical electron diffraction to obtain the crystal potential. Scattering data are required for two thicknesses of the crystalline slab for a fixed orientation of the beam. The difference in thickness Δt needs to be accurately known. For nonperiodic objects, this approach provides the projected potential between the two thicknesses.

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