Towards an exit wave in closed analytical form

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Dedicated to Professor A. F. Moodie on the occasion of his 75th birthday

Abstract

A simple but sufficiently accurate expression is obtained for the exit wave of a crystal in zone-axis orientation. The exit wave at each atom column can be parametrized with only one parameter, which is a function of the projected 'weight' of the column.

1. Introduction

If an analytical expression could be found for the exit wave of a crystal in a zone-axis orientation in terms of the projected structure of the object, this could have enormous advantages for quantitative high-resolution electron microscopy. Firstly, it would allow one to 'invert' to some extent the exit wave into a projected structure model, which can be used as a seed for further quantitative refinement. Secondly, it would speed up enormously the quantitative refinement procedure. A quantitative refinement consists of searching for the best fit between simulated and experimental data sets (images and/or diffraction patterns) in which all model parameters (atom coordinates, specimen orientation and thickness, imaging parameters *etc.*) are varied. In fact, one searches for a global optimum in a high-dimensional space. This search is performed in an iterative way in which each step requires full calculation of the dynamical electron diffraction in the crystal. At present, this is performed with standard multislice programs (*e.g.* Zandbergen *et al.*, 1997), which, if repeated thousands of times, presents a real bottleneck for flexible applications.

A simpler expression for the exit wave, and hence for the image and the diffraction pattern, allows not only a drastic increase in speed of calculation but also calculation of the change (gradient) of the fitness function with respect to change in the parameters in an analytical and hence more robust way so as to improve the convergence of the refinement procedure.

Thus far, no attempts have been successful in obtaining an expression for the exit wave in closed analytical form except for very thin objects (phase-object approximation) and for crystals in two- or three-beam orientations but neither approach is of practical value for realistic high-resolution situations.

The reason for this is that most of the theories and/or simulation programs used to date are based on plane-wave expansions for the electron wavefield. This approach stems from X-ray work and is only suitable if the scattering is weak. However, in a zone-axis orientation, where the projected crystal structure is simplest, the atom cores exactly superimpose along the beam direction and hence the scattering is very dynamical. It would be much better to look for a more appropriate quantum-mechanical basis to describe the dynamical wavefield.

In this work, we try to exploit the fact that, in a zone-axis orientation, the electrons are trapped in the electrostatic potential of the atom columns parallel to the electron beam. Therefore, we will expand the wavefield in eigenstates of the Hamiltonian of the projected atom columns. As has been observed before, the dynamical motion of the electron in a column can be expressed primarily in terms of the 1*s* bound state of the two-dimensional projected potential of the columns.

Furthermore, it has already been observed that for a variety of atom column types the 1*s* bound state has a simple scaling behaviour.

In this work, we try to parametrize this 1*s* state and hence the exit wave at the corresponding column in terms of a minimal number of parameters. Ideally, the exit wave of a column should be characterized by three parameters, two coordinates of the projection of the column and one parameter related to the 'weight' of the projected column. However, it is known that high-energy electrons only see an averaged potential along their path and are not sensitive to potential variations along this path. Hence, one can expect to obtain only projected information and ambiguity about the types and distances of atoms along a column can only be removed by combining information from different zone-axis orientations.

Finally, we can tolerate a precision in an analytical exit-wave expression of the order of say 5% since many other sources of error such as noise, inaccuracy of atom potentials, inelastic scattering, thermal effects *etc.* are at least of the same order of magnitude.

Scaled 1s wavefunction (normalized)
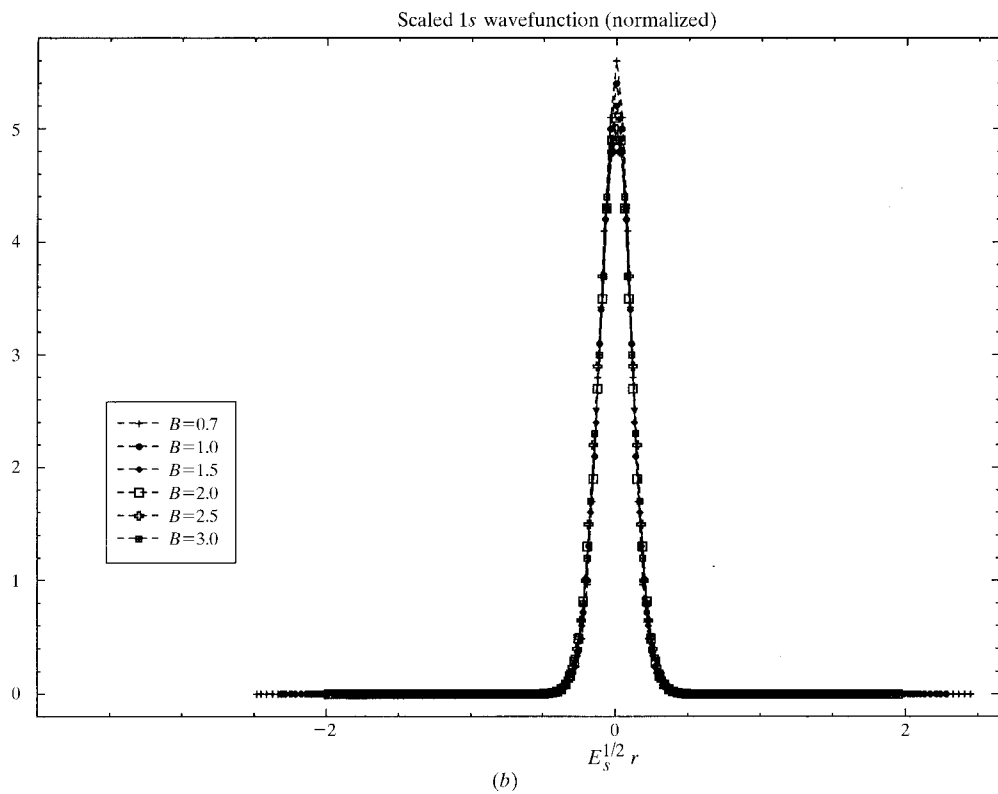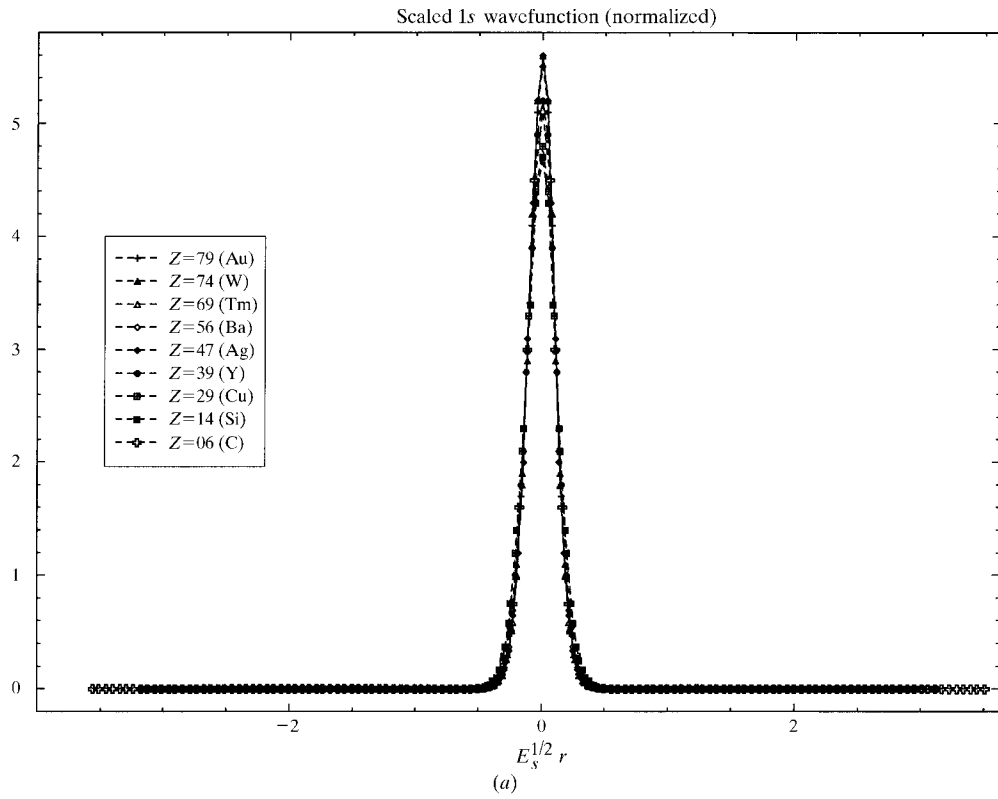

(a)

Scaled 1s wavefunction (normalized)

(b)

Fig. 1. Scaled 1s wavefunction (normalized) calculated using Doyle–Turner (1968) parameters for the potential: (a) for different types of atoms; (b) for gold but with different Debye–Waller factors.

2. Theory

It has been shown by Van Dyck & Op de Beeck (1996) that for a crystal in a zone-axis orientation the exit wave for one particular column located at the origin can be approximated as

$$\psi(\mathbf{r}, z) = 1 + \varphi_s(\mathbf{r})[\exp(-i\pi E_s k z/E_0) - 1], \qquad (1)$$

where $\varphi_s(\mathbf{r})$ is the 1s eigenstate of the Hamiltonian $H(\mathbf{r})$ of the projected column potential.

$$H(\mathbf{r})\varphi_s(\mathbf{r}) = E_s\varphi_s(\mathbf{r}). \qquad (2)$$

$\mathbf{r}$ is the vector in the two-dimensional plane perpendicular to the electron beam, k is the electron wavevector, E_0 the incident electron energy and z the crystal thickness. This approach goes back to the work of Howie (1966) and has been elaborated by Fujimoto (1978).

A two-dimensional potential of this form has one strongly bound state, which, by analogy to atoms, is called the 1s state. Note that the other states are not neglected but for thin crystals will not build up and are incorporated in the term 1 in (1). If needed, we can improve (1) so as to account for these states up to first order (Van Dyck & Op de Beeck, 1996). The 1s state is radially symmetric and can be expressed as a function of the radial coordinate $\mathbf{r}$.

By comparing the 1s states calculated for various atom column types, we have observed empirically that all the 1s states obey an empirical scaling behaviour (Van Dyck & Op de Beeck, 1996):

$$\varphi_E(\mathbf{r}) = \varphi_0(E^{1/2}\mathbf{r}) \qquad (3)$$

and we further observed that

$$E \propto Z/d^{5/4} \qquad (4)$$

or, for the extinction distance ξ (periodicity along Z),

$$\xi \propto 1/E \propto d^{5/4}/Z, \qquad (5)$$

where Z is the mass of the atoms along the column and d is the repeat distance. We can write (2) in an abbreviated notation as

$$(\nabla + V_E)\varphi_E = E\varphi_E \qquad (6)$$

with ∇ the Laplacian operator in the plane, V_E the projected potential and E the 'eigenenergy'. Assuming φ_E to be radially symmetric, we have for (6) in polar coordinates

$$\frac{1}{r}\frac{d}{dr}r\frac{d\varphi_E}{dr} + V_E\varphi_E = E\varphi_E, \qquad (7)$$

or, if we put

$$rE^{1/2} = r', \qquad (8)$$

$$\frac{1}{r}\frac{d}{dr'}r'\frac{d\varphi_E}{dr'} + \frac{1}{E}V_E\left(\frac{r'}{E^{1/2}}\right)\varphi_E = \varphi_E. \qquad (9)$$
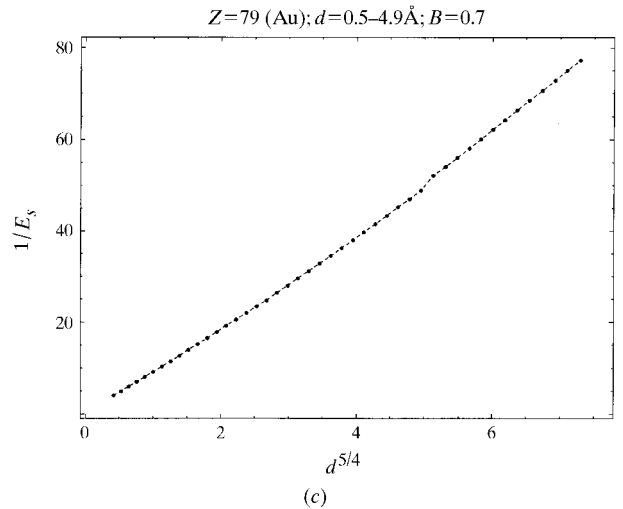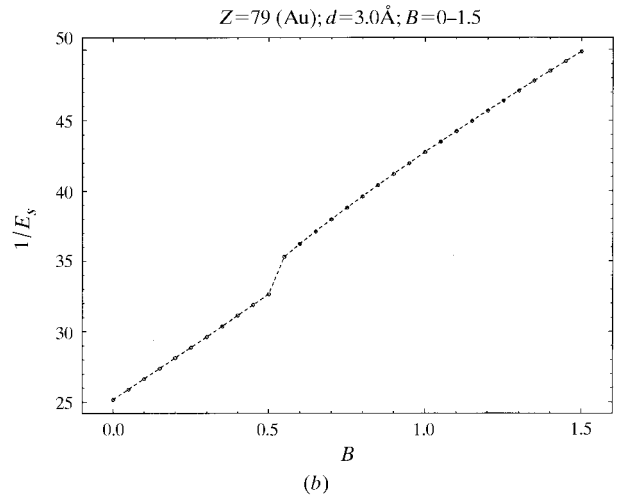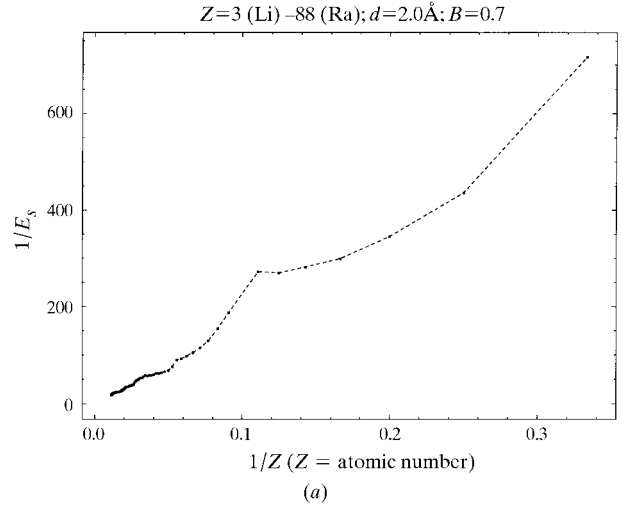
Since we have found empirically that

Fig. 2. Extinction distance along the atom column ($1/E_s$), plotted against (a) $1/Z$, (b) B, and (c) $d^{5/4}$. Slight changes in (b) and (c) are due to changes of the accuracy of the calculation method (see text).

$$\varphi_E(r') = \varphi_0(rE^{1/2}) \qquad (10)$$

is independent of E, we can conclude that

$$(1/E)V_E(r'/E^{1/2}) = V_0(r') \qquad (11)$$

or

$$V_E(r) = EV_0(rE^{1/2}). \qquad (12)$$

If we assume that $V_0(\mathbf{r}')$ is approximately a Gaussian function

$$V_0(r') = U_0 \exp(-r'^2/r_0^2), \qquad (13)$$

where U_0 and r_0 are constants, we have from (12)

$$V_E(r) = U_0 E \exp(-Er^2/r_0^2). \qquad (14)$$

If we now introduce a temperature factor B (Debye–Waller factor), $V_E(r)$ is convoluted with a Gaussian spread function proportional to $\exp(-4\pi^2 r^2/B)$. The energy is then expected to become, using (4),

$$E = \frac{Z/d^{5/4}}{\alpha + BZ/d^{5/4}} \qquad (15)$$

and the extinction distance becomes

$$\xi \propto \alpha(d^{5/4}/Z) + B, \qquad (16)$$

where α is a constant. For large factors, ξ will be determined by B; for small B factors, ξ will be determined by $d^{5/4}/Z$.

3. Results

From the foregoing, we expect that the exit wave at a particular column is radially symmetric and has a universal shape, with only one extra parameter E (or ξ) related to the projected 'weight' of the column.

From fitting with experimentally obtained exit waves, it should thus be possible to derive accurate values for the column positions and reasonable values for their weights. The extinction distance (16) is related in a simple way to Z, d and B but does not allow unambiguous determination of these values individually without prior knowledge.

Fig. 1 shows a superposition of scaled $\varphi_s(E^{1/2}r)$ functions obtained from various types of columns. It is clear that within an acceptably small error all functions are identical. Fig. 2 shows (a) ξ as a function of $1/Z$, (b) ξ as a function of B, and (c) ξ as a function of $d^{5/4}$. In all three cases, the functional relation is close to linear, which confirms the empirical relation (16).

Note that to calculate the eigenvalues and wavefunctions of $1s$ eigenstates one can use different methods, such as the standard matrix method for both the eigenvalue and wavefunction, the standard multislice method for the eigenvalue through measuring the periodicity of intensity oscillation along Z (but not for the eigenfunction) and the Bessel-function method (Op de Beeck & Van Dyck, 1995) for both the eigenvalue and wavefunction. However, these methods become very time-consuming when used to investigate the relation (16) and to obtain the accurate $1s$ eigenfunctions of heavy columns, which are sharply peaked and therefore need very fine sampling intervals (large number of beams) to describe them. We have found a fast Fourier transform method for this purpose, which works with errors less than 5% for light columns, 1% for heavy columns and 10^{-4} for very heavy columns so that the sharply peaked universal $1s$ wavefunctions can be obtained accurately. We will publish this calculation method in a separate paper.

4. Conclusions

We have established a simple analytical expression for the exit wave of a crystal in a zone-axis orientation. The exit wave at each column can be parametrized with only one parameter which is related to the 'weight' of the column and which is a simple function of the atom number, atom distance in the column and temperature factor.

These preliminary results have now to be tested in dynamical calculations for realistic crystals, and possibly extended so as to include small tilts from the zone axis.

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