Retrieval of the projected potential by inversion from the scattering matrix in electron–crystal scattering

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

Abstract

The retrieval of a unique crystal potential from the scattering matrix $\mathcal{S}$ in high-energy transmission electron diffraction is discussed. It is shown that, in general, data taken at a single orientation are not sufficient to determine all the elements of $\mathcal{S}$. Additional measurements with tilted incident beam are required for the determination of the whole $\mathcal{S}$ matrix. An algorithm for the extraction of the crystal potential from the $\mathcal{S}$ matrix measured at a single energy and thickness is presented. The limiting case of thin crystals is discussed. Several examples with simulated data are considered.

1. Introduction

The general solution of the phase and inversion problems in constructing the projected crystal potential from dynamical electron diffraction patterns has recently been discussed by Allen *et al.* (1998) and Spence (1998). Allen *et al.* considered the case where only the intensities of diffracted beams are measured. The method is based on a through-tilt series of measurements of intensities for specific well determined orientations of the incident beam which determine the moduli of all elements of the scattering matrix $\mathcal{S}$ for a chosen principal orientation of the incident beam relative to a crystalline slab of given thickness t. Using unitarity and the specific form of the scattering matrix (including symmetries), an overdetermined set of nonlinear equations is obtained from this intensity data. Solution of these equations yields the required phases of the elements of $\mathcal{S}$ and at the same time allows the determination of the Bethe matrix $\mathcal{A}$ which contains the Fourier coefficients of the (projected) crystal potential. The potential obtained is unique up to an arbitrary shift of the origin (and this is what we will mean when we say that the potential is unique in what follows). Intensity data for only a single energy and a single thickness are required. The approach of Spence proposes the use of phase information in coherent convergent-beam elec-

tron diffraction (CBED) patterns, which are such that adjacent beams overlap, to obtain not only the moduli but also the phases of the elements of the $\mathcal{S}$ matrix (once again using a through-tilt series of measurements). In Spence's approach, the extra experimental information (the phases of the elements of $\mathcal{S}$) allows the phase and inversion problems to be decoupled into two separate steps whereas in the approach of Allen *et al.* the retrieval of the phases of $\mathcal{S}$ and the inversion step $\mathcal{S} \longrightarrow \mathcal{A}$ are intertwined. Spence's method of inversion to retrieve $\mathcal{A}$ from $\mathcal{S}$ requires data at several thicknesses (or energies) to obtain a unique potential. In this paper, we will show that, by choosing the principal incident orientation appropriately, the projected potential can be obtained uniquely from a knowledge of $\mathcal{S}$ for a single energy and a single thickness *via* the solution of a set of linear equations. This is considerably simpler than the inversion method of Spence and the uniqueness of the potential obtained is easily addressed.

Measurement of all the complex elements of $\mathcal{S}$ as suggested by Spence (1998) is equivalent to determining the exit-surface wave function for the principal orientation (which defines the structure of $\mathcal{S}$) as well as for well defined secondary orientations of the incident beam. Several methods have been proposed for retrieval of the complex wave function at the exit surface of a crystal. These methods include holography (Lichte, 1991) and the combination of images recorded in a through-focus series (Kirkland, 1984; Tonomura, 1987; Lichte, 1991; Coene *et al.*, 1992; Gribelyuk & Hutchison, 1992; Van Dyck *et al.*, 1996). Several attempts have been made to exploit this information to retrieve the projected crystal potential by inversion, for example the proposals of Gribelyuk (1991), Beeching & Spargo (1993) and Lentzen & Urban (1996). In contrast to the method proposed by Spence (1998), all of these approaches only work satisfactorily for thin crystals. A common feature of these approaches is that they all start from the exit-surface wave function for a single orientation, which corresponds to knowing only a single column of $\mathcal{S}$. In this paper, we will show that when the crystal is thick enough (at a given energy of the beam)

there is, in general, insufficient information in this exit-surface wave function to determine the projected potential uniquely when dynamical effects are important. This lack of information must be overcome by measuring the exit wave function at other secondary well defined orientations of the incident beam.

So using either the method of Spence (1998) based on overlapping CBED discs or the methods of exit-plane-wave retrieval discussed in the previous paragraph, we can, in principle, determine the amplitude as well as the phase of all the complex elements of the scattering matrix $\mathcal{S}$ for a given principal orientation. The holographic and through-focus imaging methods are, however, constrained by the information resolution limit of the image-forming lens, whereas, since only adjacent orders need interfere, the CBED method is not. The CBED method provides information beyond the information resolution limit, limited ultimately only by Debye–Waller factors (Nellist *et al.*, 1995). The intensity at the midpoint of the overlap is unaffected by spherical aberration. Unlike the method of Allen *et al.* (1998), which proceeds from intensities (amplitudes) in a point diffraction pattern only, and in which the solution of the phase and inversion problems is intertwined, we are then left with the independent step $\mathcal{S} \longrightarrow \mathcal{A}$. Exploiting the fact that the diagonals of the Bethe matrix $\mathcal{A}$ are known for a given principal orientation and that $\mathcal{A}$ has certain general symmetries, we will show that for most choices of the principal orientation $\mathcal{A}$, and hence the Fourier coefficients of the projected crystal potential, can be uniquely retrieved from $\mathcal{S}$.

We do not include absorption in the considerations of the present paper because it would be an unnecessary complication for the questions of principle discussed here. Apart from this, the neglect of absorption is a justified assumption for thin crystals and has also been made in other very recent related work (Cheng *et al.*, 1996; Lentzen & Urban, 1996).

2. The direct scattering problem

The Schrödinger equation for the scattering of high-energy electrons from the periodic crystal potential can be cast in the form of an eigenvalue problem as follows (Humphreys, 1979; Allen & Rossouw, 1989; Allen *et al.*, 1998; Spence, 1998):

$$\mathcal{A}\mathcal{C} = 2K\mathcal{C}[\gamma^i]_D, \qquad (1)$$

where $[\ ]_D$ denotes a diagonal matrix. For discussion of the assumptions implicit in this equation and the constraints on its validity, see Allen & Rossouw (1989) and references therein. We will refer to the solution of the Schrödinger equation, following standard usage, as the direct scattering problem (Chadan & Sabatier, 1989; Zakhariev & Suzko, 1990).

The matrix $\mathcal{A}$ on the left-hand side of (1) is of the form

$$\mathcal{A} = \begin{pmatrix} & \vdots & \vdots & \vdots & \vdots & \vdots & \\ \cdots & -(\mathbf{k}_t + \mathbf{h})^2 & U_{\mathbf{h}-\mathbf{g}} & U_{\mathbf{h}} & U_{\mathbf{h}+\mathbf{g}} & U_{2\mathbf{h}} & \cdots \\ \cdots & U_{\mathbf{g}-\mathbf{h}} & -(\mathbf{k}_t + \mathbf{g})^2 & U_{\mathbf{g}} & U_{2\mathbf{g}} & U_{\mathbf{g}+\mathbf{h}} & \cdots \\ \cdots & U_{-\mathbf{h}} & U_{-\mathbf{g}} & -\mathbf{k}_t^2 & U_{\mathbf{g}} & U_{\mathbf{h}} & \cdots \\ \cdots & U_{-\mathbf{g}-\mathbf{h}} & U_{-2\mathbf{g}} & U_{-\mathbf{g}} & -(\mathbf{k}_t - \mathbf{g})^2 & U_{-\mathbf{g}+\mathbf{h}} & \cdots \\ \cdots & U_{-2\mathbf{h}} & U_{-\mathbf{h}-\mathbf{g}} & U_{-\mathbf{h}} & U_{-\mathbf{h}+\mathbf{g}} & -(\mathbf{k}_t - \mathbf{h})^2 & \cdots \\ & \vdots & \vdots & \vdots & \vdots & \vdots & \end{pmatrix}. \qquad (2)$$

Here the off-diagonal elements $U_{\mathbf{g}}$ are Fourier coefficients of the crystal potential resulting in elastic scattering and $\mathbf{g}$ and $\mathbf{h}$ are reciprocal-lattice vectors. The wavevector $\mathbf{k}_t$ is the tangential component of the incident electron wavevector $\mathbf{k}$ in vacuum along the plane defined by the reciprocal-lattice vectors. K is the magnitude of the incoming wavevector corrected for refraction, *i.e.* $K^2 = \mathbf{k}^2 + U_0$, where U_0 is the mean crystal potential.

Since the potential is real, $U_{\mathbf{g}} = U^*_{-\mathbf{g}}$ and consequently $\mathcal{A}$ is hermitian. Furthermore, we have made the tacit assumption that, for every reciprocal-lattice vector $\mathbf{g}$, the vector $-\mathbf{g}$ is included in the representation of $\mathcal{A}$ and this leads to symmetry across the 'anti-diagonal' of $\mathcal{A}$. In an N-beam approximation, $\mathcal{A}$ is an $N \times N$ matrix. Representing $\mathcal{A}$ by $\{\mathcal{A}_{m,n}\}$, where m and n label rows and columns, respectively, we express this symmetry as follows:

$$\mathcal{A}_{m,n} = \mathcal{A}_{N+1-n,N+1-m}, \qquad \text{with} \quad m \neq n \quad \text{if} \quad \mathbf{k}_t \neq 0. \qquad (3)$$

The matrix $\mathcal{C}$ has as columns the eigenvectors of $\mathcal{A}$ and takes the form

$$\mathcal{C} = \begin{pmatrix} \vdots & \vdots & \vdots & \vdots & \\ C^1_{\mathbf{h}} & C^2_{\mathbf{h}} & \cdots & C^i_{\mathbf{h}} & \cdots \\ C^1_{\mathbf{g}} & C^2_{\mathbf{g}} & \cdots & C^i_{\mathbf{g}} & \cdots \\ C^1_0 & C^2_0 & \cdots & C^i_0 & \cdots \\ C^1_{-\mathbf{g}} & C^2_{-\mathbf{g}} & \cdots & C^i_{-\mathbf{g}} & \cdots \\ C^1_{-\mathbf{h}} & C^2_{-\mathbf{h}} & \cdots & C^i_{-\mathbf{h}} & \cdots \\ \vdots & \vdots & \vdots & \vdots & \end{pmatrix}. \qquad (4)$$

Since $\mathcal{A}$ is hermitian, the eigenvectors of $\mathcal{A}$ are orthogonal to each other and $\mathcal{C}$ is a unitary matrix. From (1), we obtain a spectral representation of $\mathcal{A}$,

$$\mathcal{A}_{\mathbf{g},\mathbf{h}} = 2K \sum_i C^i_{\mathbf{g}} \gamma^i C^{i*}_{\mathbf{h}}, \qquad (5)$$

where the sum extends over N terms within the framework of an N-beam approximation.

The elements of the matrix $\mathcal{C}$ and the eigenvalues $2K\gamma^i$ allow us to construct the wave function of the fast electron in the crystal as a sum of Bloch states,

$$\psi(\mathbf{r}) = \sum_i \alpha^i \varphi^i(\mathbf{r}) = \sum_i \alpha^i \sum_{\mathbf{g}} C_{\mathbf{g}}^i \exp[i(\mathbf{k}^i + \mathbf{g}) \cdot \mathbf{r}]. \quad (6)$$

Each Bloch state $\varphi^i(\mathbf{r})$ is characterized by an intrinsic wavevector $\mathbf{k}^i$, which depends on the energy of the incident beam as well as on the crystal structure and can be obtained from the solution of the Schrödinger equation. The wavevectors $\mathbf{k}^i$ can be expressed in the form $\mathbf{k}^i = \mathbf{K} + \gamma^i \hat{\mathbf{n}}$ (Humphreys, 1979; Allen & Rossouw, 1989), where $\mathbf{K}$ is the wavevector of the incoming plane wave in the crystal. The unit vector $\hat{\mathbf{n}}$ is a surface normal directed into the top crystal surface and the γ^i are the *Anpassung*. The excitation amplitudes α^i are obtained from the boundary conditions at the top surface of the crystal. These require that the amplitude of the directly transmitted beam is 1 and the amplitudes of the diffracted beams are 0. From (6), it can be seen that, since $\mathcal{C}$ is unitary, $\alpha^i = C_0^{i*}$. At the exit surface of the crystal, the Bloch waves decouple into plane waves again. At this transition, the tangential components remain unchanged and therefore the amplitude of the beam $\mathbf{g}$ for a crystal of thickness t is obtained from (6) as

$$v_{\mathbf{g}}(t) = \sum_i \alpha^i C_{\mathbf{g}}^i \exp(i\gamma^i t). \quad (7)$$

Introducing the vector $\mathbf{v} = (v_{\mathbf{g}})$, we can write (7) in the compact form

$$\mathbf{v} = \mathcal{S}\mathbf{u}. \quad (8)$$

The vector $\mathbf{u} = (\delta_{\mathbf{g}0})$ characterizes the incident beam and

$$\mathcal{S} = \exp[i(1/2K)\mathcal{A}t] = \mathcal{C}[\exp(i\gamma^i t)]_D \mathcal{C}^\dagger \equiv \mathcal{C}[\Lambda^i]_D \mathcal{C}^\dagger \quad (9)$$

is the scattering matrix, where once again $[\ ]_D$ denotes a diagonal matrix. The scattering matrix relates the incident electron wave at the entrance surface of the crystal to the elastically scattered or diffracted wave at the exit surface of the crystal of thickness t (Humphreys, 1979). Schematically, we can represent $\mathcal{S}$ as

$$\mathcal{S} = \begin{pmatrix} & \vdots & \vdots & \vdots & \vdots & \vdots & \\ \cdots & S_{\mathbf{h},\mathbf{h}} & S_{\mathbf{h},\mathbf{g}} & S_{\mathbf{h},0} & S_{\mathbf{h},-\mathbf{g}} & S_{\mathbf{h},-\mathbf{h}} & \cdots \\ \cdots & S_{\mathbf{g},\mathbf{h}} & S_{\mathbf{g},\mathbf{g}} & S_{\mathbf{g},0} & S_{\mathbf{g},-\mathbf{g}} & S_{\mathbf{g},-\mathbf{h}} & \cdots \\ \cdots & S_{0,\mathbf{h}} & S_{0,\mathbf{g}} & S_{0,0} & S_{0,-\mathbf{g}} & S_{0,-\mathbf{h}} & \cdots \\ \cdots & S_{-\mathbf{g},\mathbf{h}} & S_{-\mathbf{g},\mathbf{g}} & S_{-\mathbf{g},0} & S_{-\mathbf{g},-\mathbf{g}} & S_{-\mathbf{g},-\mathbf{h}} & \cdots \\ \cdots & S_{-\mathbf{h},\mathbf{h}} & S_{-\mathbf{h},\mathbf{g}} & S_{-\mathbf{h},0} & S_{-\mathbf{h},-\mathbf{g}} & S_{-\mathbf{h},-\mathbf{h}} & \cdots \\ & \vdots & \vdots & \vdots & \vdots & \vdots & \end{pmatrix}$$

$$(10)$$

with the matrix elements

$$\mathcal{S}_{\mathbf{g},\mathbf{h}} = \sum_i C_{\mathbf{g}}^i \exp(i\gamma^i t) C_{\mathbf{h}}^{i*}. \quad (11)$$

Because of the hermiticity of $\mathcal{A}$, the scattering matrix $\mathcal{S}$ is unitary. It is also obvious from (9) that the eigenvectors of $\mathcal{S}$ are just those of $\mathcal{A}$.

3. Obtaining the matrix $\mathcal{A}$ from a knowledge of $\mathcal{S}$

From (9), we can write

$$\mathcal{A} = \frac{2K}{it} \ln(\mathcal{S}) = \frac{2K}{it} \mathcal{C} \ln[\Lambda^i]_D \mathcal{C}^\dagger = \frac{2K}{it} \mathcal{C}[i\gamma^i t]_D \mathcal{C}^\dagger. \quad (12)$$

Therefore, knowing $\mathcal{S}$, we can obtain $\mathcal{A}$ and after further evaluation the crystal potential $V(\mathbf{r})$ *via* the Fourier expansion

$$V(\mathbf{r}) = (\hbar^2/2m) \sum_{\mathbf{g}} U_{\mathbf{g}} \exp(i\mathbf{g} \cdot \mathbf{r}) = \sum_{\mathbf{g}} V_{\mathbf{g}} \exp(i\mathbf{g} \cdot \mathbf{r}).$$

$$(13)$$

However, without restrictions on the domain, the logarithm is not a unique function and yields a manifold of solutions,

$$i\gamma^i t = i(\theta^i + 2n^i\pi), \qquad n^i = 0, \pm1, \pm2, \ldots. \quad (14)$$

It is obvious that the determination of $\mathcal{A}$ *via* (12) requires knowledge of *all* the complex elements of $\mathcal{S}$. We assume that they have all been measured (both amplitude and phase). In other words, the phase problem has been solved and what remains is the inversion problem of obtaining $\mathcal{A}$ from $\mathcal{S}$. Diagonalization of $\mathcal{S}$ yields not only the eigenvectors of $\mathcal{S}$ but also those of $\mathcal{A}$. Hence, if we can find the set of parameters $\{\gamma^i\}$ in (12) unambiguously [*i.e.* resolve the ambiguities expressed in (14)], then all of $\mathcal{A}$ can be reconstructed *via* the spectral representation given by (5).

In a general case, the knowledge of all of the elements of $\mathcal{S}$ is essential for an unambiguous determination of $\mathcal{A}$ *via* inversion. This is the fundamental reason why methods of inversion based on limited information about $\mathcal{S}$, *e.g.* only the central column of $\mathcal{S}$ is known, fail in general when the crystal is not thin (Gribelyuk, 1991; Beeching & Spargo, 1993; Dorset, 1995; Peng & Wang, 1994; Peng & Zuo, 1995; Gilmore, 1996; Lentzen & Urban, 1996; Van Dyck & Op de Beeck, 1996; Zou *et al.*, 1996; Zhu & Tafto, 1997). For the incident energies (values of K) considered by these authors, if t is sufficiently small we can restrict (9) to the lowest-order term

$$\mathcal{S} \approx \mathcal{I} + (it/2K)\mathcal{A}, \quad (15)$$

where $\mathcal{I}$ is the unit matrix. This establishes a one-to-one relationship between elements of the central column of $\mathcal{S}$ and the Fourier coefficients in the central column of $\mathcal{A}$, namely $\mathcal{S}_{\mathbf{g},0} \approx (it/2K)U_{\mathbf{g}}$, $\mathbf{g} \neq 0$. This subset of the Fourier coefficients in $\mathcal{A}$ can give a limited resolution but physical representation of the projected potential. However, as soon as higher-order terms are required in (9) (*i.e.* multiple scattering is included) then the Fourier coefficients obtained *via* the approximation (15) will no longer necessarily yield a physical potential.

Consider the $\mathcal{A}$ matrix given by (2). Taking into account the fact that we know the diagonal elements of $\mathcal{A}$ (orientation information) and that $\mathcal{A}$ is hermitian and has symmetries across the 'anti-diagonal', we can count

Table 1. *The $\mathcal{S}$ matrix has been calculated for each of the principal orientations shown; the values of the integers n^i in equation (14) are shown for each orientation, then the outcome of retrieving the set γ^i uniquely using equations (17) and then both equations (17) and (18) is noted*

Case No.	$\mathbf{k}_t$	$n^i, i = 1, 2, \ldots, 7$							Equation (17)	Equations (17) and (18)
1	$(0, 0, 0)$	1	1	-1	-2	-2	-1	-1	No	No
2	$(0, 0, \bar{1})$	1	1	-3	-1	-2	-1	-2	No	Yes
3	$(\bar{1}, 1, 0)$	1	1	-3	-3	-1	-2	-2	No	Yes
4	$(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$	1	1	-3	-2	-2	-1	-1	Yes	Yes
5	$(1, \bar{1}, \frac{3}{4})$	1	1	-3	-3	-2	-1	-1	Yes	Yes

the number of open parameters in $\mathcal{A}$. Hence, *in general*, we can infer the minimum number of parameters in $\mathcal{S}$ that need to be determined to construct all of $\mathcal{S}$ so that we may then construct $\mathcal{A}$ from $\mathcal{S}$. In an N-beam approximation (N odd because of the assumption that, for every reciprocal-lattice vector $\mathbf{g}$, the vector $-\mathbf{g}$ is also included in the representation) there are $(N^2 - 1)/2$ open real parameters in $\mathcal{A}$. The number of open real parameters in a column of $\mathcal{S}$ is $2N$. For $N > 3$, measurement of a single column of $\mathcal{S}$ therefore yields less parameters than the number of open parameters in $\mathcal{A}$ and the inversion from $\mathcal{S}$ to $\mathcal{A}$ becomes underdetermined.

Assume that all the elements of $\mathcal{S}$ have been determined experimentally. This can be performed uniquely (up to an arbitrary overall phase) using either the method of Spence (1998) based on coherent overlapping CBED discs or from exit-surface wave functions. The relationship between the exit-surface wave functions and the elements of $\mathcal{S}$ is discussed in Appendix A, where we also very briefly recapitulate, in our notation, the key relation used by Spence to determine the phases of the elements of $\mathcal{S}$. Using either of these approaches, it is not necessary to know the thickness of the crystal t explicitly to determine $\mathcal{S}$ up to an arbitrary overall phase. The issue that remains to be resolved is that of the ambiguities implicit in the inversion given in (12) and which are explicitly expressed in (14). How should these ambiguities be resolved so that a unique potential can be retrieved from the scattering matrix? Firstly, we can impose the known diagonals of the $\mathcal{A}$ matrix as constraints:

$$(2K/it)(\ln \mathcal{S})_{\mathbf{g},\mathbf{g}} = \mathcal{A}_{\mathbf{g},\mathbf{g}} = -(\mathbf{k}_t + \mathbf{g})^2. \qquad (16)$$

This leads to a set of N linear equations in the N unknown γ^i's given by

$$\sum_i |C_\mathbf{g}^i|^2 \gamma^i = -(\mathbf{k}_t + \mathbf{g})^2/2K, \qquad (17)$$

where the $C_\mathbf{g}^i$ are obtained by diagonalizing $\mathcal{S}$. Furthermore, we can exploit the fact that $\mathcal{A}$ has the symmetries across its 'anti-diagonal' given by (3) to obtain a further set of homogeneous linear equations in the N unknowns γ^i,

$$\sum_i (C_{\mathbf{g}_m}^i C_{\mathbf{g}_n}^{i*} - C_{\mathbf{g}_{N+1-n}}^i C_{\mathbf{g}_{N+1-m}}^{i*}) \gamma^i = 0$$

$$\text{with} \quad m \neq n \quad \text{if} \quad \mathbf{k}_t \neq 0, \qquad (18)$$

where $\mathbf{g}_l$ refers to the reciprocal-lattice vector in the lth row of $\mathcal{C}$ given by (4). Note that the eigenvectors of $\mathcal{S}$ (and therefore also of $\mathcal{A}$) are independent of the crystal thickness t and hence (17) and (18) do not depend on t.

If $\mathbf{k}_t \neq 0$ then there are $(N^2 - 2N + 1)/4$ such complex equations. In most cases, the systems of linear equations (17) and (18) provide a linearly independent set that will allow a unique solution for the eigenvalues γ^i. This is not true for the symmetric orientation of the incident beam ($\mathbf{k}_t = 0$), where the eigenvectors of $\mathcal{S}$ satisfy (Kästner, 1993)

$$C_{-\mathbf{g}}^i = C_\mathbf{g}^{i*}. \qquad (19)$$

It is straightforward to show that (17) are then no longer linearly independent and (18) become trivial. However, by choosing the principal orientation to be other than the symmetric one, a unique solution to the inversion problem is possible using either the orientation information alone in some instances or in addition the symmetries in others. This will be made clearer by means of numerical examples in the next section.

4. Model solutions of the inversion problem

We consider the [110] zone axis in GaAs as an example. An energy of 400 keV is assumed for the incident electrons. We will work in a seven-beam approximation for simplicity of illustration but we emphasize that the method works just as well for larger values of N. We will give results for a crystalline slab of thickness 1000 Å but the approach works for arbitrary thicknesses.

The hermitian nature of $\mathcal{A}$ suggests 21 independent Fourier coefficients in the $\mathcal{A}$ matrix given by (1). These Fourier coefficients, which have been used as input to the direct problem, are shown together with the diagonal elements of $\mathcal{A}$ in Table 1 of Allen *et al.* (1998). The Fourier coefficients incorporate a Debye–Waller factor. A temperature factor $B = 0.6$ Å^2 was used for both Ga and As, as was performed by Lentzen & Urban (1996) and subsequently by Allen *et al.* (1998). If in addition we

Table 2. *The phase of the eigenvalues of the $\mathcal{S}$ matrix for case 4 in Table 1 (the eigenvalues are unimodular), the value of the corresponding n^i in equation (14) and the corresponding γ^i obtained from equations (17)*

i	θ^i	n^i	γ^i (Å^{-2})
1	2.06683	1	0.00835
2	−0.77755	1	0.00551
3	2.87168	−3	−0.01598
4	0.36297	−2	−0.01220
5	1.13330	−2	−0.01143
6	−1.51177	−1	−0.00779
7	−0.95405	−1	−0.00724

assume that for each element $U_{\mathbf{g}}$ in $\mathcal{A}$ the element $U_{-\mathbf{g}}$ is also included, then we have the symmetries across the 'anti-diagonal' of $\mathcal{A}$ indicated in (3) and therefore only 12 independent Fourier coefficients to be determined (Allen *et al.*, 1998).

The $\mathcal{S}$ matrix has been calculated for several principal orientations of the incident beam. $\mathcal{S}$ is then diagonalized to find its eigenvectors (and eigenvalues). This also yields, in each case, the eigenvectors of $\mathcal{A}$. We then use either (17) alone or both (17) and (18) to determine the parameters $\{\gamma^i\}$. If they are found unambiguously then $\mathcal{A}$ can immediately be correctly reconstructed *via* (6).

We solve the set of linear equations to obtain the $\{\gamma^i\}$ using the method of singular-value decomposition (SVD) as discussed by Press *et al.* (1992). This allows us to check whether the set of equations (17) has a coefficient matrix with determinant unequal to zero. If the determinant is zero, we can diagnose precisely what the problem is and easily examine whether using (17) *and* (18) *together* yields a unique set $\{\gamma^i\}$, since the SVD method easily accommodates the solution of more linear equations than unknowns.

The $\mathcal{S}$ matrix has been calculated for each of the principal orientations shown in Table 1. The values of the integers n^i in (14) are shown for each orientation. Then the outcome of retrieving the set $\{\gamma^i\}$ correctly (uniquely) using (17) and then both (17) and (18) is noted. As expected, there is insufficient information to determine the γ^i in the symmetric orientation. When $\mathbf{k}_t = (00\bar{1})$, *i.e.* the reflection (002) is in the exact Bragg orientation (case 2), then there is still sufficient symmetry in the system to render equations (17) linearly dependent. However, addition of the symmetry constraints, equations (18), determines the $\{\gamma^i\}$ uniquely. Tilting along the $(1\bar{1}0)$ direction gives similar results. Tilting so that the reflection $1\bar{1}1$ is in the exact Bragg orientation removes symmetries in the system so that now equations (17) are already sufficient to determine the $\{\gamma^i\}$ uniquely. The arbitrary orientation given in case 5 also breaks symmetries in such a way that the orientation constraints are sufficient to solve for the $\{\gamma^i\}$. Only with the symmetric orientation as the principal one is it not possible to uniquely determine the $\{\gamma^i\}$ and

hence to retrieve the model input potential correctly. This potential is shown in Fig. 4(*a*) of Allen *et al.* (1998).

In Table 2, we give more details for case 4 in Table 1. Shown are the phases of the eigenvalues of $\mathcal{S}$ (the eigenvalues are unimodular), the integers n^i in (14) and the corresponding γ^i which are retrieved uniquely in the inversion process *via* (17).

5. Summary and conclusions

We have shown that for a choice of principal incident orientation other than $\mathbf{k}_t = 0$, *i.e.* other than an exact zone-axis orientation (or the symmetric orientation for a systematic row), the projected potential can be recovered uniquely from the scattering matrix $\mathcal{S}$ corresponding to that principal orientation for a single thickness and energy. The solution of the phase and inversion problems can be summarized as follows.

(i) For a principal orientation other than $\mathbf{k}_t = 0$, all the complex elements of the scattering matrix $\mathcal{S}$ are measured using a through-tilt series of measurements at the principal and well defined secondary orientations of the incident beam. Either data from coherent overlapping CBED discs or image intensities measured in a through-focus series to determine exit plane-wave functions may be used to determine $\mathcal{S}$ up to an arbitrary overall phase.

(ii) $\mathcal{S}$ is then diagonalized to obtain its eigenvectors and eigenvalues $\{\Lambda^i\}$. The eigenvectors of $\mathcal{S}$ are just those of $\mathcal{A}$.

(iii) The eigenvalues $\{\gamma^i\}$ of $\mathcal{A}$ are obtained uniquely using (17) (containing known information about the principle orientation in the diagonal of $\mathcal{A}$) and (18) (derived from general symmetries of $\mathcal{A}$ across its 'anti-diagonal').

(iv) The matrix $\mathcal{A}$ is then constructed (uniquely) from its eigenvectors and eigenvalues using the spectral representation of $\mathcal{A}$ given by (5).

(v) The off-diagonal elements of $\mathcal{A}$ are the Fourier coefficients in the expansion for the projected potential given by (13).

In conclusion, two approaches to the phase and inversion problems in dynamical electron diffraction are available. Either one proceeds from the moduli of the elements of $\mathcal{S}$ only or one also performs measurements to determine the phases (*e.g. via* the CBED technique or *via* methods to determine the exit-surface wave functions). The former approach is experimentally more straightforward because it requires the measurement of intensities in simple point diffraction patterns only. However, in that case one must solve sets of nonlinear equations and the phase and inversion problems are intertwined. If the phase problem is solved using experimental data sensitive to the phases, then the inversion step can be accomplished by solving a set of linear equations and the uniqueness of the projected potential is established in a straightforward manner.

APPENDIX *A*

Firstly we show how the elements of the $\mathcal{S}$ matrix in (10) are obtained from measurements of wave functions at the exit surface. Let us assume that the principal incident direction of the incident beam is defined by the wavevector $\mathbf{K}$. The inwardly directed entrance surface unit normal $\hat{\mathbf{n}}$ is along the z direction. Assume that the origin is on the entrance surface. Any position vector $\mathbf{r}$ can be written as $\mathbf{r} = (\mathbf{r}_{xy}, z)$, where $\mathbf{r}_{xy}$ is a vector in the xy plane (parallel to the entrance and exit surfaces of the crystalline slab). For incident orientations related to the principal one by tilts of a reciprocal-lattice vector $\mathbf{h}$ (including $\mathbf{0}$) we may rewrite (6) in the form

$$
\psi(\mathbf{K} + \mathbf{h}, \mathbf{r}_{xy}, z)
$$
$$
= \exp[i(\mathbf{K} + \mathbf{h}) \cdot \mathbf{r}] \sum_{\mathbf{g}} \exp(i\mathbf{g} \cdot \mathbf{r})
$$
$$
\times \sum_{i} C_0^{i*}(\mathbf{K} + \mathbf{h}) C_{\mathbf{g}}^{i}(\mathbf{K} + \mathbf{h}) \exp[i\gamma^i(\mathbf{K} + \mathbf{h})z], \quad (20)
$$

where the dependence of C_0^{i*}, $C_{\mathbf{g}}^{i}$ and γ^i (and hence ψ) on $\mathbf{K} + \mathbf{h}$ is explicitly shown. Since $\mathbf{h}$ is a reciprocal-lattice vector, we have the following relations (Kästner, 1993):

$$
C_0^{i*}(\mathbf{K} + \mathbf{h}) = C_{\mathbf{h}}^{i*}(\mathbf{K}),
$$
$$
C_{\mathbf{g}}^{i}(\mathbf{K} + \mathbf{h}) = C_{\mathbf{g}+\mathbf{h}}^{i}(\mathbf{K}), \quad (21)
$$
$$
\gamma^i(\mathbf{K} + \mathbf{h}) = \gamma^i(\mathbf{K}).
$$

Substituting these relations into (20) and using (11), we obtain the wave function at the exit surface for a crystalline slab of thickness t:

$$
\psi(\mathbf{K} + \mathbf{h}, \mathbf{r}_{xy}, t) = \exp(i\mathbf{K} \cdot \mathbf{r}_{xy}) \exp(i\mathbf{K} \cdot \hat{\mathbf{n}} t)
$$
$$
\times \sum_{\mathbf{g}+\mathbf{h}} \exp[i(\mathbf{g} + \mathbf{h}) \cdot \mathbf{r}_{xy}] \mathcal{S}_{\mathbf{g}+\mathbf{h},\mathbf{h}}(\mathbf{K}, t),
$$
$$
(22)
$$

using the assumption that $\mathbf{g}$ and $\mathbf{h}$ are in the xy plane. By Fourier transformation, it follows that

$$
\mathcal{S}_{\mathbf{g}+\mathbf{h},\mathbf{h}}(\mathbf{K}, t) = [\exp(-i\mathbf{K} \cdot \hat{\mathbf{n}} t)/A_c] \int_{A_c} \psi(\mathbf{K} + \mathbf{h}, \mathbf{r}_{xy}, t)
$$
$$
\times \exp[-i(\mathbf{K} + \mathbf{g} + \mathbf{h}) \cdot \mathbf{r}_{xy}] \, \mathrm{d}^2 r_{xy}, \quad (23)
$$

where A_c is the area of the unit cell projected onto the xy plane. By appropriate choices of $\mathbf{g}$, we can therefore obtain, from the exit-surface wave function for the orientation defined by $\mathbf{K} + \mathbf{h}$, all complex elements of the column of $\mathcal{S}$ labelled by $\mathbf{h}$ – see (10). If t is not known then $\mathcal{S}$ is still determined up to an arbitrary overall phase by (23).

Lastly, we briefly give the key relation that is used in the method of coherent overlapping CBED discs (Spence, 1998) using the notation of this paper. Consider two adjacent columns of the $\mathcal{S}$ matrix in (10), say those labelled by $\mathbf{g}$ and $\mathbf{h}$. We can determine the relative phase of the $\mathcal{S}$-matrix elements in the columns labelled $\mathbf{g}$ and $\mathbf{h}$

and in the same row $\mathbf{f}$ by considering the overlapping CBED discs $\mathbf{f}'$ and $\mathbf{f}''$ such that $\mathbf{f} = \mathbf{f}' + \mathbf{g} = \mathbf{f}'' + \mathbf{h}$. The (measured) intensity at a point in the region of overlap of the CBED discs is given by

$$
I(\mathbf{r}_{xy}, t) = |\mathcal{S}_{\mathbf{f}'+\mathbf{g},\mathbf{g}}|^2 + |\mathcal{S}_{\mathbf{f}''+\mathbf{h},\mathbf{h}}|^2 + 2|\mathcal{S}_{\mathbf{f}'+\mathbf{g},\mathbf{g}}||\mathcal{S}_{\mathbf{f}''+\mathbf{h},\mathbf{h}}|
$$
$$
\times \cos[(\mathbf{f}' - \mathbf{f}'' + \mathbf{g} - \mathbf{h}) \cdot \mathbf{r}_{xy} + \theta_{\mathbf{f}'+\mathbf{g},\mathbf{g}} - \theta_{\mathbf{f}''+\mathbf{h},\mathbf{h}}].
$$
$$
(24)
$$

Equations (21) have been used to refer all $\mathcal{S}$-matrix quantities back to that at the principal orientation. The quantities $\theta_{\mathbf{f}'+\mathbf{g},\mathbf{g}}$ and $\theta_{\mathbf{f}''+\mathbf{h},\mathbf{h}}$ are the required phases of the $\mathcal{S}$-matrix elements. The practical implementation of such relations to uniquely determine the phases of all the elements of $\mathcal{S}$ *via* the solution of a set of linear equations has been discussed in detail and by example in Spence's paper. The moduli of the $\mathcal{S}$-matrix elements can be obtained from a point diffraction pattern without overlap of adjacent orders. Note that, while the moduli and phases of $\mathcal{S}$-matrix elements in (24) depend on the thickness of the crystal t, it is not necessary to know t explicitly in determining $\mathcal{S}$.

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