

Obtaining the Crystal Potential by Inversion from Electron Scattering Intensities

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Abstract

A method to obtain the crystal potential from the intensities of the diffracted beams in high-energy electron diffraction is proposed. It is based on a series of measurements for specific well determined orientations of the incident beam, which determine the moduli of all elements of the scattering matrix. Using unitarity and the specific form of the scattering matrix (including symmetries), an overdetermined set of non-linear equations is obtained from these data. Solution of these equations yields the required phase information and allows the determination of a (projected) crystal potential by inversion that is unique up to an arbitrary shift of the origin. The reconstruction of potentials from intensities is illustrated for two realistic examples, a {111} systematic row case in ZnS and a [110] zone-axis orientation in GaAs (both noncentrosymmetric crystals).

1. Introduction

Electron diffraction is increasingly being used to obtain quantitative information on the crystal potential (Spence, 1993). In particular, one is interested in the amplitude and phase of the lower-order coefficients of the Fourier expansion of the periodic potential, often referred to as the structure factors. At present, fitting procedures using intensity patterns in convergent-beam electron diffraction (CBED) are used to provide information on the amplitudes and phases of these coefficients (Spence, 1993). These fitting procedures involve the repeated solution of the forward or direct solution of the fundamental equations for electron diffraction. In this paper, we are concerned with obtaining the structure factors from measurements of dynamical beam intensities without at any stage solving the forward or direct problem. This involves the solution of both a phase and an inversion problem.

When multiple scattering can be ignored, the effective scattering amplitude is of simple form and reduces essentially to a Fourier transform of the crystal potential. This situation occurs for photon and neutron scattering on crystals where the effective interaction is

rather weak and a single-scattering (*kinematical*) approximation yields a fairly good description of these processes. In contrast to photons and neutrons, electrons experience a rather strong interaction in a crystal and in general multiple-scattering (*dynamical*) contributions are essential. However, no general relationship is known between dynamical diffracted-beam intensities and the structure factors.

For accelerating voltages around 100 keV and more, *N*-beam ($N > 2$) diffraction is dominant (Moodie & Whitfield, 1994). Certain *N*-beam cases can be reduced to two-beam form for both centrosymmetric (Fukuhara, 1966; Blume, 1966; Kogiso & Takahashi, 1977; Takeda, 1987) and noncentrosymmetric crystals (Moodie & Fehlmann, 1993; Moodie & Whitfield, 1994). In some cases, closed-form inversion from scattering intensities to structure factors is possible (Moodie, 1979; Allen & Rossouw, 1993; Moodie *et al.*, 1996).

There has been considerable work recently proposing various techniques to construct the projected crystal potential from measured data. Peng & Wang (1994) investigated the conditions under which an 'electron-density map' would provide reliable information on the potential. Peng & Zuo (1995) presented an inversion algorithm assuming small deviations from a known potential. Lentzen & Urban (1996) developed a method, based on the simulated-annealing algorithm, for reconstruction of the projected crystal potential. Zou *et al.* (1996) considered the retrieval of the projected potential from a single image of a thin sample. Huang *et al.* (1996) have also proposed an empirical approach to obtaining potential maps.

Phase information on the diffracted beams are available in electron holography or from images recorded in a defocus series (Tonomura, 1987; Lichte, 1991; Van Dyck *et al.*, 1996) since the scattered wave function at the exit surface of the crystal may be reconstructed from the data. A possible method for obtaining the crystal potential by inversion of the complex wavefield at the exit surface of the specimen has been proposed by Beeching & Spargo (1993). It is based on a reversal of the multislice algorithm and is limited to thin crystals (of the order of 100 Å or less).

The method fails when the thickness of the crystal is such that the scattering is sufficiently strong for dominant Bloch-wave structure to appear in the wave function, *i.e.* there is strong dynamical scattering (Beeching *et al.*, 1994). Other methods to obtain structural information by inversion have been discussed by Gribelyuk (1991) and Scheerschmidt & Knoll (1994). The so-called 'direct phasing' and 'Fourier' techniques to obtain structural information from nearly kinematical intensity data have been reviewed by Dorset (1995). The maximum-entropy approach to solving the phase problem has been reviewed by Gilmore (1996).

In this paper, we propose a method to obtain the crystal potential from the intensities of the diffracted beams in high-energy electron diffraction. The scattering matrix S relates the incident electron wave function to the wave function at the exit surface of the crystal. The moduli of the elements of the S matrix for a given principal orientation of the incident beam can be obtained from a series of measurements of the intensities of diffracted beams at that and other orientations of the incident beam. The unitarity of the S matrix provides constraints on the phases of the elements of S but they are insufficient to solve the phase problem. Here we demonstrate within the context of a general N -beam approximation how the unitarity of S and the specific form of the scattering matrix (including symmetries) yield constraints that allow us to solve the phase problem and to find a (projected) crystal potential by inversion. We discuss the conditions under which unique solutions to the phase and inversion problems are found. The reconstruction of potentials from beam intensities and possible ambiguities are illustrated for a {111} systematic row case in ZnS and for the case of the [110] zone-axis orientation in GaAs (both noncentrosymmetric crystals). The zone-axis results are compared with the projected crystal potential that was reconstructed recently by the simulated-annealing algorithm for the same case (Lentzen & Urban, 1996).

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

Firstly, we recapitulate some essential elements of the solution of the Schrödinger equation starting from the periodic crystalline potential pertinent to electron diffraction (Humphreys, 1979; Allen & Rossouw, 1989) using the Bloch-wave formulation. Following the standard terminology (Chadan & Sabatier, 1989; Zakhariev

& Suzko, 1990), we will refer to this as the direct problem.

The spatial distribution of the crystal potential is reconstructed from the Fourier summation,

$$V(\mathbf{r}) = (\hbar^2/2m) \sum_{\mathbf{g}} U_{\mathbf{g}} \exp(i\mathbf{g} \cdot \mathbf{r}) = \sum_{\mathbf{g}} V_{\mathbf{g}} \exp(i\mathbf{g} \cdot \mathbf{r}), \quad (1)$$

where $U_{\mathbf{g}}$ (or $V_{\mathbf{g}}$) are the Fourier coefficients of the potential due to elastic scattering and \mathbf{g} are reciprocal-lattice vectors. In practice, the sum over all reciprocal-lattice vectors is truncated to a subset of the physically important ones. Because we are not including absorption, the potential $V(\mathbf{r})$ is real and consequently the relationship $U_{\mathbf{g}} = U_{-\mathbf{g}}^*$ is valid. Furthermore, we make the assumption that if the term with coefficient $U_{\mathbf{g}}$ is included in the expansion then so is $U_{-\mathbf{g}}$. For noncentrosymmetric crystals, these coefficients are complex and for the centrosymmetric case [where there exists a center of symmetry such that $V(\mathbf{r}) = V(-\mathbf{r})$ for all \mathbf{r}], the origin can be chosen so that they are real. Consistent with the periodic nature of the potential, the total wave function for the fast electron inside the crystal is a superposition of Bloch states $\phi^i(\mathbf{r})$ with the amplitudes α^i ,

$$\psi(\mathbf{r}) = \sum_i \alpha^i \phi^i(\mathbf{r}) = \sum_i \alpha^i \sum_{\mathbf{g}} C_{\mathbf{g}}^i \exp[i(\mathbf{k}^i + \mathbf{g}) \cdot \mathbf{r}]. \quad (2)$$

Each Bloch wave is characterized by an intrinsic wavevector \mathbf{k}^i that depends on the energy of the incident beam as well as on the crystal structure and can be obtained by a solution of the Schrödinger equation. Boundary conditions allow us to express each \mathbf{k}^i in the form (Humphreys, 1979; Allen & Rossouw, 1989)

$$\mathbf{k}^i = \mathbf{K} + \gamma^i \hat{\mathbf{n}}, \quad (3)$$

where \mathbf{K} is the wavevector of the incoming plane wave in the crystal (corrected for refraction, *i.e.* $K^2 = k^2 + U_0$, where k is the magnitude of the wavevector in vacuum). The unit vector $\hat{\mathbf{n}}$ is a surface normal directed into the top crystal surface and the γ^i are the *Anpassung*.

Using the expansions for $V(\mathbf{r})$ and $\psi(\mathbf{r})$, the Schrödinger equation for high-energy electrons within the crystal can be recast in the form (Humphreys, 1979)

$$\mathcal{A}C = 2KC[\gamma^i]_D, \quad (4)$$

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.

The matrix \mathcal{A} on the left-hand side of equation (4) is of the form

$$\mathcal{A} = \begin{pmatrix} \vdots & \vdots & \vdots & \vdots & \vdots & \vdots \\ \dots & -(\mathbf{k}_i + \mathbf{h})^2 & U_{\mathbf{h}-\mathbf{g}} & U_{\mathbf{h}} & U_{\mathbf{h}+\mathbf{g}} & U_{2\mathbf{h}} & \dots \\ \dots & U_{\mathbf{g}-\mathbf{h}} & -(\mathbf{k}_i + \mathbf{g})^2 & U_{\mathbf{g}} & U_{2\mathbf{g}} & U_{\mathbf{g}+\mathbf{h}} & \dots \\ \dots & U_{-\mathbf{h}} & U_{-\mathbf{g}} & -\mathbf{k}_i^2 & U_{\mathbf{g}} & U_{\mathbf{h}} & \dots \\ \dots & U_{-\mathbf{g}-\mathbf{h}} & U_{-2\mathbf{g}} & U_{-\mathbf{g}} & -(\mathbf{k}_i - \mathbf{g})^2 & U_{-\mathbf{g}+\mathbf{h}} & \dots \\ \dots & U_{-2\mathbf{h}} & U_{-\mathbf{h}-\mathbf{g}} & U_{-\mathbf{h}} & U_{-\mathbf{h}+\mathbf{g}} & -(\mathbf{k}_i - \mathbf{h})^2 & \dots \\ \vdots & \vdots & \vdots & \vdots & \vdots & \vdots & \vdots \end{pmatrix} \quad (5)$$

The wavevector \mathbf{k}_i is the tangential component of the incident electron wavevector along the plane defined by the reciprocal-lattice vectors. The off-diagonal elements in \mathcal{A} are just the Fourier coefficients in the expansion for the potential given by equation (1). 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 a 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 $\{A_{m,n}\}$, where m and n label rows and columns, respectively, we express this symmetry as

$$A_{m,n} = A_{N+1-n, N+1-m}, \quad \text{with } m \neq n \text{ if } \mathbf{k}_i \neq 0. \quad (6)$$

Equation (4) is in principle an eigenvalue equation for \mathcal{A} with eigenvalues $2K\gamma^i$. The i th column of the matrix \mathcal{C} ,

$$\mathcal{C} = \begin{pmatrix} \vdots & \vdots & \vdots & \vdots & \vdots \\ C_{\mathbf{h}}^1 & C_{\mathbf{h}}^2 & \dots & C_{\mathbf{h}}^i & \dots \\ C_{\mathbf{g}}^1 & C_{\mathbf{g}}^2 & \dots & C_{\mathbf{g}}^i & \dots \\ C_{\mathbf{0}}^1 & C_{\mathbf{0}}^2 & \dots & C_{\mathbf{0}}^i & \dots \\ C_{-\mathbf{g}}^1 & C_{-\mathbf{g}}^2 & \dots & C_{-\mathbf{g}}^i & \dots \\ C_{-\mathbf{h}}^1 & C_{-\mathbf{h}}^2 & \dots & C_{-\mathbf{h}}^i & \dots \\ \vdots & \vdots & \vdots & \vdots & \vdots \end{pmatrix} \quad (7)$$

represents an eigenvector of \mathcal{A} associated with the eigenvalue $2K\gamma^i$ and contains the expansion coefficients $C_{\mathbf{g}}^i$ of equation (2). In an N -beam approximation, there are N such eigenvalues and eigenvectors but generally there are more than N different Fourier coefficients of the potential involved in \mathcal{A} .

Since \mathcal{A} is Hermitian, the eigenvectors of \mathcal{A} are orthogonal to each other and can be normalized to form a unitary matrix \mathcal{C} . The same eigenvectors can be used for a spectral representation of \mathcal{A}

$$A_{\mathbf{g},\mathbf{h}} = 2K \sum_i C_{\mathbf{g}}^i \gamma^i C_{\mathbf{h}}^{i*}, \quad (8)$$

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

To obtain the total electron wave function within the crystal, we need the coefficients α^i , which 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 equation (2), it can be seen that, since \mathcal{C} is unitary, $\alpha^i = C_{\mathbf{0}}^{i*}$. 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 equation (2) as

$$v_{\mathbf{g}}(t) = \sum_i \alpha^i C_{\mathbf{g}}^i \exp(i\gamma^i t). \quad (9)$$

Introducing the vector $\mathbf{v} = (v_{\mathbf{g}})$, we can write equation (9) in the compact form

$$\mathbf{v} = \mathcal{S}\mathbf{u}. \quad (10)$$

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

$$\mathcal{S} = \exp[i(1/2K)\mathcal{A}t] = \mathcal{C}[\exp(i\gamma^i t)]_D \mathcal{C}^\dagger \equiv \mathcal{C}[\Lambda^i]_D \mathcal{C}^\dagger \quad (11)$$

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 elastic scattered or diffracted wave at the exit surface of the crystal of thickness t (Humphreys, 1979). Schematically, we can represent \mathcal{S} as

$$\mathcal{S} = \begin{pmatrix} \vdots & \vdots & \vdots & \vdots & \vdots & \vdots \\ \dots & S_{\mathbf{h},\mathbf{h}} & S_{\mathbf{h},\mathbf{g}} & S_{\mathbf{h},\mathbf{0}} & S_{\mathbf{h},-\mathbf{g}} & S_{\mathbf{h},-\mathbf{h}} & \dots \\ \dots & S_{\mathbf{g},\mathbf{h}} & S_{\mathbf{g},\mathbf{g}} & S_{\mathbf{g},\mathbf{0}} & S_{\mathbf{g},-\mathbf{g}} & S_{\mathbf{g},-\mathbf{h}} & \dots \\ \dots & S_{\mathbf{0},\mathbf{h}} & S_{\mathbf{0},\mathbf{g}} & S_{\mathbf{0},\mathbf{0}} & S_{\mathbf{0},-\mathbf{g}} & S_{\mathbf{0},-\mathbf{h}} & \dots \\ \dots & S_{-\mathbf{g},\mathbf{h}} & S_{-\mathbf{g},\mathbf{g}} & S_{-\mathbf{g},\mathbf{0}} & S_{-\mathbf{g},-\mathbf{g}} & S_{-\mathbf{g},-\mathbf{h}} & \dots \\ \dots & S_{-\mathbf{h},\mathbf{h}} & S_{-\mathbf{h},\mathbf{g}} & S_{-\mathbf{h},\mathbf{0}} & S_{-\mathbf{h},-\mathbf{g}} & S_{-\mathbf{h},-\mathbf{h}} & \dots \\ \vdots & \vdots & \vdots & \vdots & \vdots & \vdots & \vdots \end{pmatrix} \quad (12)$$

with the matrix elements

$$S_{\mathbf{g},\mathbf{h}} = \sum_i C_{\mathbf{g}}^i \exp(i\gamma^i t) C_{\mathbf{h}}^{i*}. \quad (13)$$

Because of the hermiticity of \mathcal{A} , the scattering matrix \mathcal{S} is unitary. It is also obvious from equation (11) that the eigenvectors of \mathcal{S} are just those of \mathcal{A} .

For a given orientation of the incident beam, the quantities that can be measured are the intensities of the diffracted beams, *i.e.* we obtain for the beam \mathbf{g} that

$$I_{\mathbf{g}} = |v_{\mathbf{g}}(t)|^2 = \left| \sum_i C_{\mathbf{0}}^{i*} C_{\mathbf{g}}^i \exp(i\gamma^i t) \right|^2 = |S_{\mathbf{g},\mathbf{0}}|^2. \quad (14)$$

This means that, for a given principal orientation of the incident beam, only the moduli of the elements in the central column of the scattering matrix \mathcal{S} shown in equation (12) are measured. However, by tilting the incident beam to appropriate secondary orientations

related to the principal one by tilts of a reciprocal-lattice vector, we can measure the moduli of all the elements of S . This is discussed further in the next section.

3. The phase and inverse scattering problems

The experimental determination of the phase and recovery of the potential is one of the most important questions in electron-crystal diffraction. The key points that need to be addressed in the solution of the phase and inversion problems are the existence of a solution, the uniqueness of any solution and an effective algorithm to find any solution(s). Firstly, let us consider the question of obtaining the matrix \mathcal{A} from a knowledge of S . The matrix \mathcal{A} contains as its off-diagonal elements Fourier coefficients $U_{\mathbf{g}}$ of the crystal potential and this allows evaluation of $V(\mathbf{r})$. From equation (11), we can write

$$\begin{aligned} \mathcal{A} &= (2K/it) \ln(S) \\ &= (2K/it) \mathcal{C} \ln[\Lambda^i]_D \mathcal{C}^\dagger \\ &= (2K/it) \mathcal{C} [i\gamma^i t]_D \mathcal{C}^\dagger. \end{aligned} \quad (15)$$

Therefore, knowing S , we can obtain \mathcal{A} and after further evaluation the crystal potential $V(\mathbf{r})$. However, without restrictions on the domain, the logarithm is not a unique function and yields a manifold number of solutions,

$$i\gamma^i t = i(\theta^i + 2n^i\pi), \quad n^i = 0, \pm 1, \pm 2, \dots \quad (16)$$

Apart from this unpleasant ambiguity, the determination of \mathcal{A} via equation (15) requires knowledge of all the complex elements of S . Measuring the intensities of the diffracted beams at the principal orientation of the incident beam gives us the moduli of the central column of S -matrix elements in equation (12). Assuming an N -beam approximation, the moduli of the remaining $N^2 - N$ elements can be obtained by the fact that intensities of the beams at other orientations are related to the remaining S -matrix elements for the principal orientation. This follows from the fact that (Kästner, 1993)

$$\begin{aligned} C_{\mathbf{g}-\mathbf{h}}^i(\mathbf{K} + \mathbf{h}) &= C_{\mathbf{g}}^i(\mathbf{K}) \\ \gamma^i(\mathbf{K} + \mathbf{h}) &= \gamma^i(\mathbf{K}), \end{aligned} \quad (17)$$

where we now indicate the orientation dependence of the $C_{\mathbf{g}}^i$ and γ^i explicitly. Therefore, by orientating the incident beam in directions $\mathbf{K} + \mathbf{h}$, where \mathbf{h} are reciprocal-lattice vectors, the moduli of all elements of the S matrix for the principal orientation (at which we wish to solve the phase and inversion problems) can be

obtained.† Besides the moduli of the S -matrix elements, we also need their phases to determine the \mathcal{A} matrix and hence the potential (up to an arbitrary choice of origin).

An obvious constraint on the phases of the elements of the S matrix is provided by the unitarity of S and this leads to the following equations:

$$\sum_l S_{i,l} S_{j,l}^* = \delta_{ij}. \quad (18)$$

These equations can be recast into the form

$$\begin{aligned} \sum_f |S_{\mathbf{g},f}| |S_{\mathbf{h},f}| \cos(\theta_{\mathbf{g},f} - \theta_{\mathbf{h},f}) &= \delta_{\mathbf{g},\mathbf{h}} \\ \sum_f |S_{\mathbf{g},f}| |S_{\mathbf{h},f}| \sin(\theta_{\mathbf{g},f} - \theta_{\mathbf{h},f}) &= 0, \end{aligned} \quad (19)$$

where $\theta_{\mathbf{g},f}$ denotes the phase of $S_{\mathbf{g},f}$. In an N -beam approximation, unitarity of S is not exactly satisfied but it is reasonable to assume that the defect is negligible. Hence, equation (19) leads to a system of $2N^2$ equations. Actually, there are only $N^2 - N$ independent constraint equations because the interchange of \mathbf{g} and \mathbf{h} leads to the same equations and for $\mathbf{g} = \mathbf{h}$ equations (19) either contain no phase information or are trivial. Therefore, unitarity on its own is in general not sufficient to determine all of the phases.

The fact that unitarity of the S matrix is not sufficient to determine the phases of its elements is contrary to the recent conjecture by Tivol (1995). This conjecture was based on a dispersion relation similar to that for central potentials where the phase problem can be solved up to some well understood ambiguities (Newton, 1968; Lun *et al.*, 1994; Huber *et al.*, 1996). However, there is a crucial difference between the scattering from a central potential and the scattering off a crystal. In the former, the scattered waves are not spatially separated from the incident wave (Newton, 1968) since in point scattering the S matrix takes into account the scattered wave as well as contributions of the incident wave. This is completely different in electron-crystal scattering, where the crystalline slab provides a natural separation of both regimes. Hence, a measurement of electron intensities after transmission through the crystal yields $|S_{\mathbf{g},\mathbf{h}}|^2$ directly and not, as in point scattering, the absolute value of the scattering amplitude ($|S - 1|^2$). This was not taken into account by Tivol (1995).

The S matrix depends on both the crystal potential (the off-diagonal elements of \mathcal{A}) and the energy and orientation of the incident beam (information

† Since this paper was submitted, we have become aware of related work by J. C. H. Spence (1998) entitled *Direct Inversion of Dynamical Diffraction Patterns to Structure Factors*. In that paper, the periodicity relations of the Bloch waves are also exploited and there is a clear discussion in that paper that explains precisely how such a through-tilt series of measurements would be implemented.

contained in the diagonal elements of \mathcal{A}). It seems reasonable that this latter information should also be exploited in the phase and inversion problem. This can be accomplished by imposing the known diagonals of the \mathcal{A} matrix as constraints on \mathcal{S} :

$$(2K/it)(\ln \mathcal{S})_{\mathbf{g},\mathbf{g}} = -(\mathbf{k}_t + \mathbf{g})^2, \quad (20)$$

which yields $2N$ real constraint equations. However, this introduces N further unknowns into the problem *via* equation (16). Nevertheless, equations (19) and (20) together provide us with $(N^2 - N) + 2N$ equations containing $N^2 + N$ unknowns. However, they do not provide a unique solution to the phase and inversion problem, as we will show by example in the next section. They allow a discrete set of \mathcal{A} matrices that all have the same diagonal elements and are Hermitian [this is implied by equation (15) and the unitarity of \mathcal{S}] but do not all have the symmetries given in equation (6). The potentials obtained this way can be unphysical. To ensure that \mathcal{A} has the correct symmetries across its 'anti-diagonal', we require that the \mathcal{S} matrix satisfy

$$[\ln \mathcal{S}]_{m,n} = [\ln \mathcal{S}]_{N+1-n,N+1-m} \\ \text{with } m \neq n \text{ if } \mathbf{k}_t \neq 0. \quad (21)$$

An \mathcal{S} matrix that satisfies equations (18), (20) and (21) is found by exhaustive testing to yield an \mathcal{S} matrix that is related to a test input \mathcal{S} by a similarity transform, as we will see in §§4.1, 4.2. These \mathcal{S} matrices yield projected potentials that are related to each other by a shift of origin. This shift of origin is of course unimportant since the choice of origin is arbitrary and does not affect observed intensities.

If we use the constraint equations (20) or (21) in the solution of the phase and inversion problem then we need to evaluate $\ln \mathcal{S}$. This then introduces the parameters n^i in equation (16) into the problem. Therefore, in addition to the N^2 phases in \mathcal{S} we have to simultaneously determine these N parameters. Unitarity together with equation (20) provide as many constraint equations as independent parameters, *i.e.* $N^2 + N$. The symmetry constraints given by equation (21) yield $\frac{1}{2}N^2 - N + \frac{1}{2}$ constraint equations (real and imaginary parts). Therefore, we have $N^2 + N$ unknowns and, in principle, $\frac{3}{2}N^2 + \frac{1}{2}$ constraint equations. (N is implicitly assumed to be odd, which is consistent with our assumption that, for every \mathbf{g} included in our basis set, we have also included $-\mathbf{g}$.)

It should be noted that for a centrosymmetric crystal the \mathcal{S} matrix is symmetric and the number of unknown phases is reduced to $N(N+1)/2$. We also point out that, for an exact zone-axis orientation (or the symmetric orientation for a systematic row) ($\mathbf{k}_t = 0$), the eigenvectors of \mathcal{S} satisfy

$$C_{-\mathbf{g}}^i = C_{\mathbf{g}}^{i*}. \quad (22)$$

It then follows that the relation

$$S_{m,n} = S_{N+1-n,N+1-m} \quad (23)$$

holds. Consequently, \mathcal{S} has a similar symmetry across the 'anti-diagonal' as \mathcal{A} . The product of two such matrices is again a matrix of the same symmetry. It therefore follows that equation (21) is immediately satisfied if we impose equation (23) on the problem from the outset. This means that the problem can be solved with fewer unknowns.

We will illustrate the discussion in this section by means of two model solutions of the phase and inversion problems in the next section.

4. Model solutions of phase and inversion problem

4.1. Systematic row case

For tilts along a systematic row with beams $\{\dots, -2\mathbf{g}, -\mathbf{g}, \mathbf{0}, \mathbf{g}, 2\mathbf{g}, \dots\}$, the \mathcal{A} matrix becomes a band matrix of the form

$$\mathcal{A} = \begin{pmatrix} \ddots & \vdots & \vdots & \vdots & \vdots & \vdots & \vdots \\ \dots & -(k_t + 2\mathbf{g})^2 & U_{\mathbf{g}} & U_{2\mathbf{g}} & U_{3\mathbf{g}} & U_{4\mathbf{g}} & \dots \\ \dots & U_{-\mathbf{g}} & -(k_t + \mathbf{g})^2 & U_{\mathbf{g}} & U_{2\mathbf{g}} & U_{3\mathbf{g}} & \dots \\ \dots & U_{-2\mathbf{g}} & U_{-\mathbf{g}} & -k_t^2 & U_{\mathbf{g}} & U_{2\mathbf{g}} & \dots \\ \dots & U_{-3\mathbf{g}} & U_{-2\mathbf{g}} & U_{-\mathbf{g}} & -(k_t - \mathbf{g})^2 & U_{\mathbf{g}} & \dots \\ \dots & U_{-4\mathbf{g}} & U_{-3\mathbf{g}} & U_{-2\mathbf{g}} & U_{-\mathbf{g}} & -(k_t - 2\mathbf{g})^2 & \dots \\ \vdots & \vdots & \vdots & \vdots & \vdots & \vdots & \ddots \end{pmatrix}. \quad (24)$$

As an example, we will consider a five-beam case $\{-2\mathbf{g}, -\mathbf{g}, \mathbf{0}, \mathbf{g}, 2\mathbf{g}\}$ with the incident beam in the symmetric orientation ($k_t = 0$). There are four independent Fourier coefficients in \mathcal{A} . The corresponding \mathcal{S} matrix, given by

$$\mathcal{S} = \begin{pmatrix} S_{2\mathbf{g},2\mathbf{g}} & S_{2\mathbf{g},\mathbf{g}} & S_{2\mathbf{g},\mathbf{0}} & S_{2\mathbf{g},-\mathbf{g}} & S_{2\mathbf{g},-2\mathbf{g}} \\ S_{\mathbf{g},2\mathbf{g}} & S_{\mathbf{g},\mathbf{g}} & S_{\mathbf{g},\mathbf{0}} & S_{\mathbf{g},-\mathbf{g}} & S_{\mathbf{g},-2\mathbf{g}} \\ S_{\mathbf{0},2\mathbf{g}} & S_{\mathbf{0},\mathbf{g}} & S_{\mathbf{0},\mathbf{0}} & S_{\mathbf{0},-\mathbf{g}} & S_{\mathbf{0},-2\mathbf{g}} \\ S_{-\mathbf{g},2\mathbf{g}} & S_{-\mathbf{g},\mathbf{g}} & S_{-\mathbf{g},\mathbf{0}} & S_{-\mathbf{g},-\mathbf{g}} & S_{-\mathbf{g},-2\mathbf{g}} \\ S_{-2\mathbf{g},2\mathbf{g}} & S_{-2\mathbf{g},\mathbf{g}} & S_{-2\mathbf{g},\mathbf{0}} & S_{-2\mathbf{g},-\mathbf{g}} & S_{-2\mathbf{g},-2\mathbf{g}} \end{pmatrix}, \quad (25)$$

has the symmetries given by equation (23).

For the $\{111\}$ systematic row case in the noncentrosymmetric crystal ZnS, we have calculated a model \mathcal{A} matrix for a sample at 300 K. The potential coefficients incorporate a Debye-Waller factor to account for the thermal motion of the atoms. A temperature factor $B = 0.8676 \text{ \AA}^2$ was used for Zn and $B = 0.7477 \text{ \AA}^2$ for S (Reid, 1983). From the \mathcal{A} matrix, we have evaluated the corresponding \mathcal{S} matrix for 100 keV electrons incident in the symmetric orientation on a crystalline slab 100 \AA thick. The moduli of these model \mathcal{S} -matrix elements have been used as input to test the feasibility of the proposed algorithm for the solution of the phase and inversion problem. Firstly, we have used the unitarity of

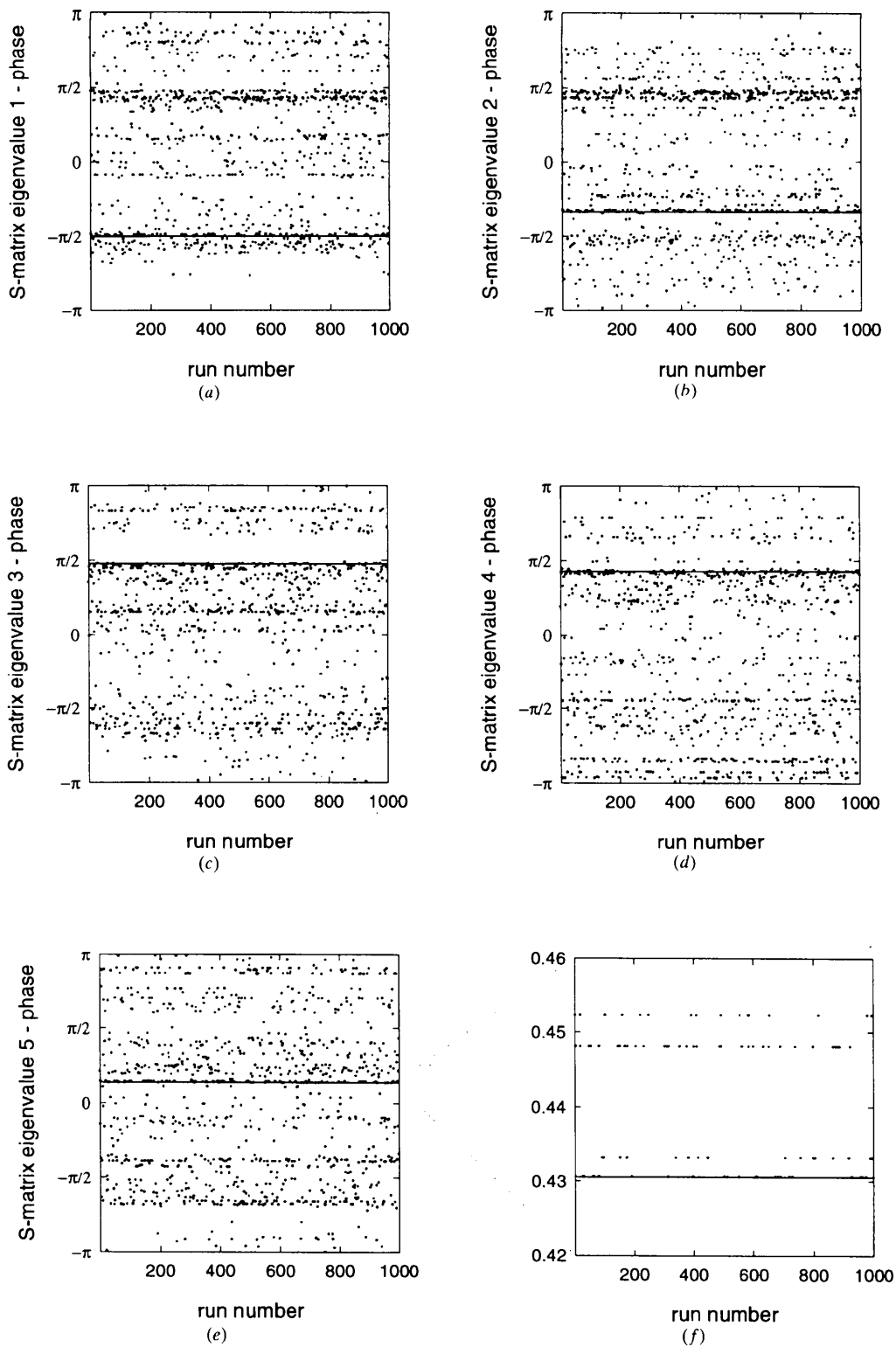


Fig. 1. The phases of the eigenvalues of the S matrix for 1000 solutions of the phase problem satisfying the unitarity constraint given by equation (19) and the orientation constraint on S given by equation (20) but not requiring the symmetries on S given by equation (21). The intensities (moduli of S) were calculated for a five-beam $\{111\}$ systematic row in ZnS. The energy of the incident beam is 100 keV and the crystal is in the symmetric orientation. The crystal is assumed to be 100 Å thick. (f) is a magnification of (e) around the model value of the phase.

S given by equations (18) or (19) and the orientation constraints on S given by equation (20), but not the symmetry constraints given by equation (21), to find solutions of the phase problem. In total, we have $N^2 + N$ nonlinear constraint equations, $N^2 - N$ from unitarity and $2N$ equations from equation (20), containing $N^2 + N$ unknowns.

We use a *global* method to solve the system of nonlinear equations. The algorithm [implemented in

subroutine *newt*, described by Press *et al.* (1992)] combines the rapid local convergence of Newton's method for finding the roots of a system of nonlinear equations with a globally convergent strategy that will guarantee progress towards a solution at each iteration. To further ensure that the whole phase space is thoroughly explored in finding possible solutions of these nonlinear equations, we have used a random-number generator to generate a series of sets of randomly chosen initial guesses for the solution of the phase problem.

We expect the phase problem to only be solved up to a similarity transformation since an arbitrary translation of the origin gives \mathcal{A} matrices and hence, *via* equation (11), S matrices that are related by a similarity transformation. Matrices that are similar have the same eigenvalues and therefore a comparison of the eigenvalues of S with those of the input S is a test of the reasonableness of the solution. However, using the method outlined above [*i.e.* equations (19) and (20) only], unitary S matrices are obtained that are not related by a similarity transformation. Starting from a thousand sets of arbitrarily chosen guesses for the phases of S , we have found solutions to the phase problem that are consistent with S being unitary and which on inversion yield the correct diagonal elements of \mathcal{A} . The phases of the eigenvalues of S for these solutions are shown in Fig. 1 (all eigenvalues have modulus unity). Banded solutions, which on closer examination show further discrete substructure are

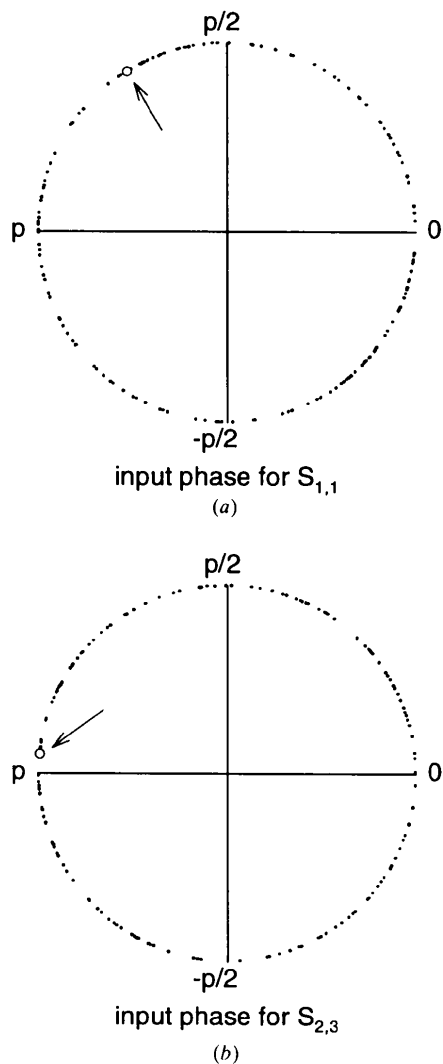


Fig. 2. Starting guesses for the phases of the S matrix for 1000 solutions of the phase problem satisfying the constraint of unitarity of S given by equations (19), the orientation constraint on S given by equation (20) and also the symmetry constraint on S given by equation (21). The intensities (moduli of S) were calculated for a five-beam $\{111\}$ systematic row in ZnS. The energy of the incident beam is 100 keV and the crystal is in the symmetric orientation. The crystal is assumed to be 100 Å thick. The arrows point to the starting guesses for the phases of two of the S -matrix elements for one of the arbitrarily chosen sets of initial phases. The potential obtained starting from this particular set of initial guesses is shown in Fig. 3.

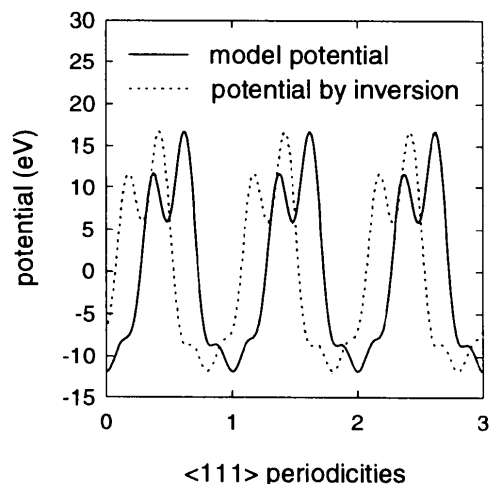


Fig. 3. The input model potential and a potential obtained from intensities calculated from the input potential by solving the phase and inversion problem for a five-beam $\{111\}$ systematic row in ZnS. The constraint of unitarity of S given by equations (19) as well as the constraints on S given by equations (20) and (21) were used. The potential found by inversion is the same as the input potential up to shift of origin. The guessed starting phases for two of the S -matrix elements are indicated in Fig. 2. The potential has negative values since the mean inner potential V_0 is not included.

Table 1. *A* matrix for the $[110]$ zone axis in GaAs in a seven-beam approximation

The modulus (units \AA^{-2}) and phase of each element of the model \mathcal{A} matrix are compared with those of an \mathcal{A} matrix found by solving the phase and inversion problems.

i	j	Model		Found	
		Modulus	Phase	Modulus	Phase
1	1	0.1251262	3.141593	0.1251262	3.141593
1	2	8.2210027E-02	0.7325953	8.2210034E-02	1.171833
1	3	4.5383908E-02	-0.7456781	4.5383919E-02	-1.231796
1	4	5.7383953E-03	0.0000000	5.7384139E-03	-0.9253497
1	5	8.2210027E-02	0.7325953	8.2210042E-02	-0.6319979
1	6	4.5383908E-02	-0.7456781	4.5383941E-02	-3.035627
1	7	5.1696863E-02	0.0000000	5.1696822E-02	-1.850710
2	2	9.3844675E-02	3.141593	9.3844689E-02	3.141593
2	3	5.7383953E-03	0.0000000	5.7383878E-03	-0.9253545
2	4	8.2210027E-02	0.7325953	8.2210034E-02	-0.6319982
2	5	7.6010779E-02	0.0000000	7.6010756E-02	-1.803830
2	6	2.2602889E-03	0.0000000	2.2602931E-03	-2.729195
2	7	4.5383908E-02	-0.7456781	4.5383923E-02	-3.035626
3	3	9.3844675E-02	3.141593	9.3844675E-02	-3.141593
3	4	8.2210027E-02	-0.7325953	8.2210034E-02	-1.171833
3	5	2.2602889E-03	0.0000000	2.2602708E-03	-0.8784839
3	6	7.6010779E-02	0.0000000	7.6010779E-02	-1.803831
3	7	8.2210027E-02	0.7325953	8.2210034E-02	-0.6319979
4	4	0.0000000E+00	0.0000000	1.7557491E-08	-0.4374708
4	5	8.2210027E-02	-0.7325953	8.2210056E-02	-1.171833
4	6	8.2210027E-02	0.7325953	8.2210027E-02	-0.6319980
4	7	5.7383953E-03	0.0000000	5.7384237E-03	-0.9253498
5	5	9.3844675E-02	3.141593	9.3844704E-02	-3.141593
5	6	5.7383953E-03	0.0000000	5.7383808E-03	-0.9253526
5	7	4.5383908E-02	-0.7456781	4.5383930E-02	-1.231795
6	6	9.3844675E-02	3.141593	9.3844660E-02	-3.141593
6	7	8.2210027E-02	0.7325953	8.2210034E-02	1.171832
7	7	0.1251262	3.141593	0.1251262	3.141593

obtained. Fig. 1(*f*), a magnification of Fig. 1(*e*) around the model value of the phase, clearly shows the discrete nature of the broader bands evident in Figs. 1(*a*)–(*e*). The phases of the eigenvalues of the model \mathcal{S} matrix are shown by the solid lines. Solutions of the phase problem can be found that give \mathcal{S} matrices not related by a similarity transformation and hence do not represent the same physical potential. The n^i values for all the solutions found were the set $\{0, 0, 0\}$.

We repeated the calculations just discussed for a thickness of 400\AA and banded solutions were once again obtained. However, the only solutions for \mathcal{A} that occur at both thicknesses are correct ones. Therefore, data at different thicknesses provide us with a unique solution to the inversion problem (up to an arbitrary choice of origin). Furthermore, this solution is the only one that yields intensities correctly at orientations other than the symmetric orientation.

Consider again now the case where we only have intensity measurements at one thickness. If we now impose the further constraint that the \mathcal{S} matrix has the symmetries evident in equation (21), then we once again find different solutions to the phase problem but now all the \mathcal{S} matrices have the same eigenvalues irrespective of the starting guesses for the phases in our numerical solution of the phase problem. In practice, we have implemented equations (20) and (21) by

requiring the modulus of the difference between the left- and right-hand sides of each equation be zero. The real and imaginary parts of the unitarity equations were each included separately.

In Fig. 2, we show the arbitrary guessed phases of two of the elements of \mathcal{S} which were input into the numerical procedure to solve the nonlinear constraint equations which yield \mathcal{S} matrices and, by inversion, \mathcal{A} matrices with all the correct symmetries. Clearly, the phase space is well covered by our starting guesses for these two phases. Such is also the case for all the other elements of the \mathcal{S} matrix. The \mathcal{S} matrices finally found from all these sets of starting guesses are related to the model by similarity transformations and the corresponding \mathcal{A} matrices all give potentials corresponding to the original potential by a shift of origin. This indicates that, irrespective of our starting point, if we find a solution to the nonlinear constraint equations, then it always leads to a potential that is unique up to a choice of origin.

The arrows in Fig. 2 point to the starting guesses for the phases of two of the \mathcal{S} -matrix elements for one particular set of arbitrarily chosen initial phases. The input model potential and the potential corresponding to the solution of the phase and inversion problem obtained starting from this set of initial guesses for the phases of the \mathcal{S} matrix is shown in Fig. 3. The potential

Table 2. S matrix for the $[110]$ zone axis in GaAs for 400 keV electrons incident in the exact zone-axis orientation on a crystal 112 Å thick

The modulus and phase of elements of the model S matrix are compared with those of an S matrix found by solving the phase problem. These S matrices correspond to the A matrices in Table 1.

i	j	Model		Found	
		Modulus	Phase	Modulus	Phase
1	1	0.7144645	-0.8613200	0.7144645	-0.8613202
1	2	0.3965959	1.980233	0.3965959	2.419471
1	3	0.2070741	0.6754036	0.2070741	0.1892851
1	4	8.4514052E-02	-2.339094	8.4514052E-02	3.018733
1	5	0.3965959	1.980233	0.3965959	0.6156400
1	6	0.2070741	0.6754036	0.2070741	-1.614546
1	7	0.2864644	1.639925	0.2864644	-0.2107853
2	1	0.3758914	0.4858674	0.3758914	4.6629861E-02
2	2	0.6914203	-0.6738561	0.6914203	-0.6738564
2	3	2.8726360E-02	1.427802	2.8726360E-02	0.5024490
2	4	0.4196112	2.273396	0.4196112	0.9088027
2	5	0.4005086	1.607214	0.4005086	-0.1966167
2	6	2.1400725E-02	2.202648	2.1400725E-02	-2.5265385
3	1	0.2399583	2.207322	0.2399583	2.693440
3	2	2.7373556E-02	2.689571	2.7373556E-02	-2.668251
3	3	0.6914203	-0.6738561	0.6914203	-0.6738561
3	4	0.3797395	0.7808390	0.3797395	0.3416016
3	5	4.1365739E-02	3.122441	4.1365739E-02	2.243968
4	1	0.1064531	1.487735	0.1064531	2.413092
4	2	0.3797395	0.7808390	0.3797395	2.145433
4	3	0.4196112	2.273396	0.4196112	2.712634
4	4	0.5839296	1.9742457E-02	0.5839296	1.9742429E-02
5	1	0.3758914	0.4858674	0.3758914	1.850461
5	2	0.4005086	1.607214	0.4005086	-2.872141
5	3	2.1400725E-02	2.202648	2.1400725E-02	3.081137
6	1	0.2399583	2.207322	0.2399583	-1.785915
6	2	4.1365739E-02	3.122441	4.1365739E-02	-0.4315567
7	1	0.2836539	1.680123	0.2836539	-2.752351

obtained is identical to the model potential up to a shift of the origin.

The calculations described above have all proceeded from intensities calculated within a five-beam approximation. Intensities have also been calculated using 51 beams in the direct problem. Using the appropriate subset of these intensities as input to a solution of the phase and inversion problem in a five-beam approximation, we were able, to some extent, to assess the effects of truncation of the phase and inversion problems to a smaller (finite) number of beams when applied to more realistically calculated data. The sum of the truncated set of intensities was, of course, such that the sum of the intensities for a given orientation was less than one. Nevertheless, inversion from these intensities gave stable and physical solutions to the phase problem similar to those obtained above, albeit at a lower level of accuracy. Renormalization of the intensities to sum to one before solving the phase and inversion problem once again led to physical solutions and with improved accuracy.

4.2. Zone-axis case

Lentzen & Urban (1996) used a method based on the simulated-annealing algorithm to reconstruct the

projected crystal potential from the exit-plane-wave function for the $[110]$ zone axis in GaAs (incident electron energy of 400 keV). They tested their algorithm for thicknesses of 8, 56 and 112 Å. For the 112 Å case, they found an incorrect solution. They suggested that their reconstruction of the crystal potential belongs to a class of inverse scattering problems that have more than one solution.

We consider the 112 Å case discussed by Lentzen & Urban (1996) in a seven-beam approximation, assuming an exact zone-axis orientation. Taking into account only the Hermitian nature of A suggests 21 independent Fourier coefficients in the A matrix given by equation (5). These Fourier coefficients, which have been used as input to the direct problem, are shown together with the diagonal elements of A in Table 1. The Fourier coefficients incorporate a Debye-Waller factor. A temperature factor $B = 0.6 \text{ Å}^2$ was used for both Ga and As, as was performed by Lentzen & Urban (1996). If in addition we assume that for each element $U_{\mathbf{g}}$ in A the element $U_{-\mathbf{g}}$ is also included, then we have the symmetries across the 'anti-diagonal' of A indicated in equation (6). Then, as can be seen by inspection of Table 1, there are 12 independent Fourier coefficients to be determined.

Exploring the solutions to the nonlinear constraint equations for the phases of the S matrix from a global point of view was time consuming. Starting with arbitrary guesses for the phases of the S -matrix elements, of the order of 100 solutions were found to the problem, so that the whole of the solution space was reasonably explored. All these S matrices are related to the model input S matrix, which is listed in Table 2 [equation (23) is satisfied], by a similarity transformation. An example of one of the solutions to the phase problem is shown in Table 2 and the corresponding A matrix obtained by inversion is given in Table 1. The potentials corresponding to solutions of the phase problem only differ

by a translation in real space. The projected potential corresponding to the A matrix found by solution of the phase and inversion problems and given in Table 1 is compared to the model potential in Fig. 4.

5. Conclusions

We have proposed a method to obtain the projected crystal potential from intensities of the diffracted beams in high-energy electron diffraction. At no stage during this procedure is it necessary to solve the direct or forward problem of electron diffraction. The unitarity of the S matrix together with constraints on S that make it consistent with information about the orientation of the incident beam and on further general structure of the matrix A are sufficient to determine the projected potential up to an arbitrary shift of origin. Since we have not solved the problem algebraically, a general mathematical statement on the existence and uniqueness of the solution for the phase and inversion problem (for data from a single thickness) is not straightforward. The question of uniqueness has been addressed by finding solutions starting from many different sets of arbitrarily chosen phases of the S -matrix elements. No spurious solutions, which also fulfilled all our requirements for a physical solution, were found.

We have demonstrated the method for a systematic row case in a five-beam approximation and a zone-axis case in a seven-beam approximation (in both cases for noncentrosymmetric crystals). Although we chose as principal orientations the symmetric and exact zone-axis orientations, successful tests of the method have also been performed for arbitrary incident-beam directions.

In principle, the method can be applied for larger numbers of beams. However, the algorithm to solve the phase and inversion problems involves the solution of a set of nonlinear equations. In general, increasing the number of open parameters means that such a method tends to ill posedness. As the number of beams becomes larger, greater accuracy will be required to achieve the excellent reconstruction of the Fourier coefficients in Table 1 for a seven-beam case. The computation time needed to find solutions of the nonlinear equations was typically of the order of tens of seconds for the five-beam case and for the seven-beam case it was a few minutes. Solutions in a 13-beam approximation were tried and solution times were several hours. Calculations were performed on a Pentium-based personal computer. However, no attempt was made to optimize the solution method with respect to speed, with accuracy of the solutions found being the main consideration. Further work will explore more efficient methods of solution and also study the conditions under which analytic solutions may be possible.

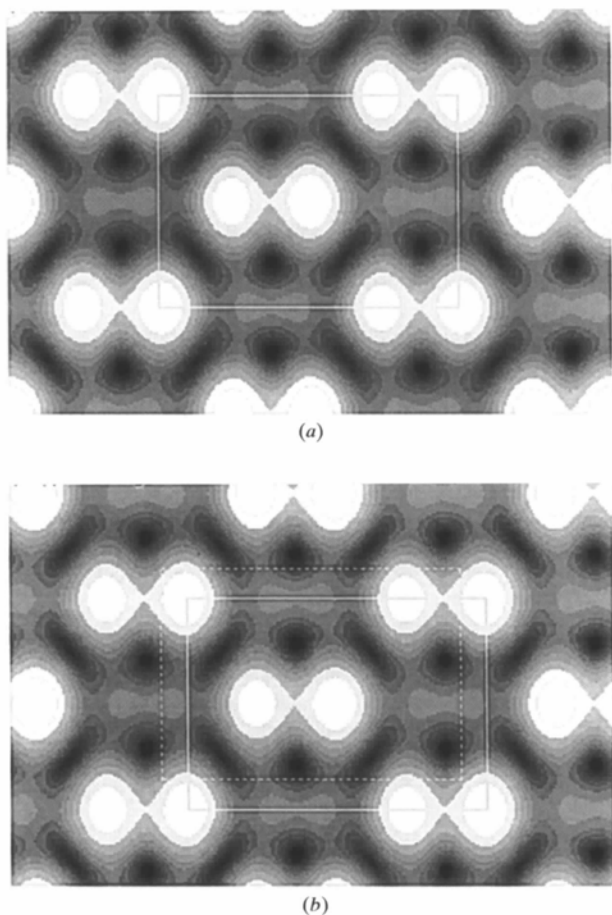


Fig. 4. (a) The model projected potential for the [110] zone axis in GaAs at 300 K. The rectangle joins four equivalent potential points corresponding to the position of As atoms. (b) A potential obtained from intensities calculated from the input potential, assuming 400 keV incident electrons in the exact zone-axis orientation and a crystal 112 Å thick, by solving the phase and inversion problems. The constraint of unitarity of S given by equations (19) as well as the constraints on S given by equations (20) and (21) were used. The potential found by inversion is the same as the input potential up to shift of origin, as indicated by the shift of the rectangle in (a) from its former position, indicated by the dashed rectangle. The calculations were performed in a seven-beam approximation.

The stability of this approach in the presence of errors in measured intensities, the precision needed in the energy and orientation of the incident beam and inelastic scattering effects are important considerations for future work.

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References

- Allen, L. J. & Rossouw, C. J. (1989). *Phys. Rev. B*, **39**, 8313–8321.
- Allen, L. J. & Rossouw, C. J. (1993). *Ultramicroscopy*, **48**, 341–346.
- Beeching, M. J. & Spargo, A. E. C. (1993). *Ultramicroscopy*, **52**, 243–247.
- Beeching, M. J., Spargo, A. E. C. & Allen, L. J. (1994). *Ultramicroscopy*, **55**, 329–333.
- Blume, J. (1966). *Z. Phys.* **191**, 248–272.
- Chadan, K. & Sabatier, P. C. (1989). *Inverse Problems in Quantum Scattering Theory*, 2nd ed. New York: Springer-Verlag.
- Cheng, Y. F., Nüchter, W., Mayer, J., Weickenmeier, A. & Gjønnes, J. (1996). *Acta Cryst. A* **52**, 923–936.
- Dorset, D. L. (1995). *Acta Cryst. A* **51**, 869–879.
- Fukuhara, A. (1966). *J. Phys. Soc. Jpn.* **21**, 2645–2662.
- Gilmore, C. J. (1996). *Acta Cryst. A* **52**, 561–589.
- Gribelyuk, M. A. (1991). *Acta Cryst. A* **47**, 715–723.
- Huang, D. X., Liu, W., Gu, Y. X., Xiong, J. W., Fan, H. F. & Li, F. H. (1996). *Acta Cryst. A* **52**, 152–157.
- Huber, H., Lun, D. R., Allen, L. J. & Amos, K. (1996). *Phys. Rev. A*, **54**, 1363–1371.
- Humphreys, C. J. (1979). *Rep. Prog. Phys.* **42**, 1825–1887.
- Kästner, G. (1993). *Many Beam Electron Diffraction Related to Electron Microscope Diffraction Contrast*. Berlin: Akademie Verlag.
- Kogiso, M. & Takahashi, H. (1977). *J. Phys. Soc. Jpn.* **42**, 223–229.
- Lentzen, M. & Urban, K. (1996). *Ultramicroscopy*, **62**, 89–102.
- Lichte, H. (1991). *Advances in Optical and Electron Microscopy*, edited by T. Mulvey & C. J. R. Sheppard, Vol. 12, pp. 25–91. London: Academic Press.
- Lun, D. R., Allen, L. J. & Amos, K. (1994). *Phys. Rev. A*, **50**, 4000–4006.
- Moodie, A. F. (1979). *Chem. Scr.* **14**, 21–22.
- Moodie, A. F., Etheridge, J. & Humphreys, C. J. (1996). *Acta Cryst. A* **52**, 596–605.
- Moodie, A. F. & Fehlmann, M. (1993). *Acta Cryst. A* **49**, 376–378.
- Moodie, A. F. & Whitfield, A. J. (1994). *Acta Cryst. A* **50**, 730–736.
- Newton, R. G. (1968). *J. Math. Phys.* **9**, 2050–2055.
- Peng, L.-M. & Wang, S. Q. (1994). *Acta Cryst. A* **50**, 759–771.
- Peng, L.-M. & Zuo, J. M. (1995). *Ultramicroscopy*, **57**, 1–9.
- Press, W. H., Teukolsky, S. A., Vetterling, W. T. & Flannery, B. P. (1992). *Numerical Recipes in Fortran*, 2nd ed., pp. 375–381. Cambridge University Press.
- Reid, J. S. (1983). *Acta Cryst. A* **39**, 1–13.
- Scheerschmidt, K. & Knoll, F. (1994). *Phys. Status Solidi A*, **146**, 491–502.
- Spence, J. C. H. (1993). *Acta Cryst. A* **49**, 231–260.
- Spence, J. C. H. (1998). *Acta Cryst. A* **54**, 7–18.
- Takeda, M. (1987). *Phys. Status Solidi A*, **101**, 25–36.
- Tivol, W. F. (1995). *Acta Cryst. A* **51**, 708–716.
- Tonomura, A. (1987). *Rev. Mod. Phys.* **59**, 639–669.
- Van Dyck, D., Lichte, H. & van der Mast, K. D. (1996). *Ultramicroscopy*, **64**, 1–15.
- Zakhariev, B. N. & Suzko, A. A. (1990). *Direct and Inverse Problems*. Berlin: Springer-Verlag.
- Zou, X., Sunberg, M., Larine, M. & Hovmöller, S. (1996). *Ultramicroscopy*, **62**, 103–121.