Contrary to the case of $n=72$, for $n=54$ the same locally extremal packing is obtained with tetrahedral $\{3,3+\}_{4,2}$ and octahedral $\{3,4+\}_{3,1}$ surface lattices. It should be noted that this packing also represents a four-branched spherical helix structure (Székely, 1974; Tarnai, 1985).

The applied method did not result in Danzerian rigid packings for $n=114$ and 282. The result for $n=114$ is not of interest since the circle diameter for $n=114$ is less than the circle diameter for $n=120$. But, the arrangement of 282 circles is quite good, so it is worth improving it, by giving up the icosahedral symmetry.

Terms of the packing sequences $\{3,q+\}_{c+1,c}$, $\{3,q+\}_{c+2,c}$ defined with removal of the vertices of the base polyhedra $\{3,q\}$ present Danzerian rigid arrangements and quite large densities in all of the investigated cases. On the basis of the results obtained it is expected that Danzerian rigid packings will also be obtained in these sequences for values $c>3$.

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Theory of Electron Diffraction from Planar Ideal Crystals

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Abstract

A theory of electron diffraction from a planar ideal crystal of arbitrary thickness is presented. It is based on Schrödinger's equation. Both the relativistic corrections in energy and wavelength and the electron 'absorption' due to the presence of inelastic scattering may be incorporated as usual. This theory is constructed in an exact differential-equation approach known as rigorous coupled-wave analysis. This is an exact method of diffraction analysis that has been extensively tested for its numerical calculation scheme. The exact solution for electron wave amplitudes of all diffraction orders is formally presented in terms of a standard eigenvalue problem and explicitly expressed in matrix form. Numerical calculation can be implemented on digital computers in a straightforward manner. An approximate conservation law is given for the transmittance and reflectance, which are then the relevant dynamical quantities to be measured in a realistic time-dependent diffraction process and to be calculated in this time-independent diffraction theory for comparison. Two derivations of the well known Bragg law are sketched.

1. Introduction

In theories of electron diffraction from a planar ideal crystal, as in all wave-motion problems, the wave field is usually expanded into certain elementary waves when a differential-equation approach is adopted. The amplitudes of the elementary-wave components are to be determined, exactly or approximately, by a wave equation and boundary conditions. A particular relativistically corrected form of Schrödinger's wave equation is used when electron polarization may be ignored (Hirsch, Howie, Nicholson, Pashley & Whelan, 1977; Cowley, 1981). As for the wave expansion, there are three main types. One type is

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the semi-reciprocal approach of Tournarie (1962), in which the wave field is expanded into Fourier series with respect only to the coordinates parallel to the crystal surfaces. The amplitudes are then depth dependent and governed by second-order differential equations. Lynch & Moodie (1972) converted these equations to first order, and obtained an exact but formal solution of the latter.

Another type is obtained by expanding the wave field in Bloch waves with constant amplitudes. This approach was initiated by Ewald (1917) for X-ray diffraction and adopted by Bethe (1928) for electrons. An exact and formal solution for electron diffraction in this approach was given by Colella (1972), in which the numerical calculation is essentially reduced to a standard eigenvalue problem. The third type expands the wave field as a single depth-dependent Bloch wave. The physical idea of this approach was originated by Darwin (1914) for X-rays, and further developed by Howie & Whelan (1961) for electrons. In an exact treatement of electron diffraction in this approach, van Dyck (1976) directly solved the resulting second-order equations of the depth-dependent amplitudes exactly and formally.

In this paper we solve the same problem, also exactly and formally, by adopting still another approach. We expand the wave field as a single depth-dependent Bloch wave, just as in the aforementioned van Dyck (1976) theory. The resulting second-order equations of the coupled amplitudes, however, are converted to first order, as in Lynch & Moodie's (1972) semi-reciprocal approach. The resulting first-order ordinary differential equations with constant coefficients are then solved formally and exactly in matrix form. Finally, the diffraction amplitudes of all diffraction orders in the vacuum regions outside the crystal are determined in terms of the amplitude of the incident wave by application of boundary conditions. Numerical calculation in this theory is essentially reduced to a standard eigenvalue problem, as in Colella's (1972) Bloch-wave expansion.

2. Coupled-amplitude equations and solutions

Schrödinger's equation may be cast in the form

$$\nabla^2\psi(\mathbf{r})+(2\pi k_0)^2\varepsilon(\mathbf{r})\psi(\mathbf{r})=0, \qquad (1)$$

with $\varepsilon(\mathbf{r})=1+eV(\mathbf{r})/E$ and $k_0=1/\lambda=(2m_0E)^{1/2}/h$. Here E, m_0 and $-e$ are respectively the energy, mass and charge of the electron. $V(\mathbf{r})$ is the potential, and h is Planck's constant. The crystal slab of arbitrary but finite thickness d is assumed to have infinite lateral extension (region II with $0\le z\le d$), and it divides the vacuum space into two regions (region I with $-\infty<z\le 0$ and region III with $d\le z<\infty$). The electron is incident from region I and assumed to be an arbitrary monoenergetic plane wave, $\psi_{\rm inc}(\mathbf{r})=\exp(i2\pi\mathbf{k}_0.\mathbf{r})b$. The coordinate frame is chosen such that $\mathbf{k}_0=k_0(\sin\theta^i, 0, \cos\theta^i)$.

In each region, the wave function $\psi(\mathbf{r})$ and the permittivity $\varepsilon(\mathbf{r})$ are expanded in the same forms,

$$\psi(\bar{\mathbf{r}})=\sum_{m,n,t}\varphi_{mnt}(\bar{z})\exp(i\mathbf{n}_{mnt}.\bar{\mathbf{r}}) \qquad (2)$$

$$\varepsilon(\bar{\mathbf{r}})=\sum_{m,n,t}\varepsilon_{mnt}\exp[i(\mathbf{n}_{mnt}-\mathbf{n}_{000}).\bar{\mathbf{r}}], \qquad (3)$$

where $\bar{\mathbf{r}}=2\pi k_0\mathbf{r}$ and $\mathbf{n}_{mnt}=(\mathbf{K}+m\mathbf{b}_1+n\mathbf{b}_2+t\mathbf{b}_3)/k_0$ with m, n, $t=0$, ±1, ±2, ... and $\mathbf{K}=k_0[\sin\theta^i, 0, +(\varepsilon_{000}-\sin^2\theta^i)^{1/2}]$. The explicit form $m\mathbf{b}_1+n\mathbf{b}_2+t\mathbf{b}_3$ of the reciprocal-lattice vectors $\mathbf{g}$ is used here for later convenience in the discussions concerning the diffraction order numbers m, n, t of the diffraction-wave components. Substituting (2) and (3) into (1), we obtain

$$\begin{aligned}&{\rm d}^2\varphi_{mnt}/{\rm d}\bar{z}^2+i2n_{mntz}\,{\rm d}\varphi_{mnt}/{\rm d}\bar{z}\\&\qquad-\mathbf{n}^2_{mnt}\varphi_{mnt}+\sum_{m,n,t}\varepsilon_{m-u,n-v,t-w}\varphi_{uvw}=0.\end{aligned} \qquad (4)$$

The form of the wave-function expansion, (2), and, consequently, the coupled-amplitude equations, (4), is almost exactly the same as that of van Dyck (1976). Equation (2) is in the form of a single depth-dependent Bloch wave. To be more precise, it is a single modified Bloch wave in which the depth-dependent amplitudes have taken the places of the constant coefficients in the space-harmonic expansion of the Bloch wave. It is known as the coupled-wave expansion in electromagnetic wave diffraction (Gaylord & Moharam, 1982).

The set of second-order differential equations in (4) can be readily converted into an equivalent set of first-order ones by using the so-called state variables (*e.g.* Liu & Liu, 1975). In fact, Lynch & Moodie (1972) used such a technique in solving the amplitude equations in semi-reciprocal space. For later convenience in expressing the boundary conditions in § 3, we define the state variable 'conjugate' to φ_{mnt} as

$$\chi_{mnt}=n_{mntz}\varphi_{mnt}-i\,{\rm d}\varphi_{mnt}/{\rm d}\bar{z}. \qquad (5)$$

Thus, from (4) and (5), a set of linear and homogeneous first-order differential equations can be obtained and then combined into a matrix equation

$${\rm df}/{\rm d}\bar{z}=i{\rm Cf}, \qquad (6)$$

with

$${\rm f}=\begin{bmatrix}{\rm f}_1\\{\rm f}_2\end{bmatrix},\qquad {\rm C}=\begin{bmatrix}-{\rm n}_z & 1\\ \varepsilon-{\rm n}_x^2-{\rm n}_y^2 & -{\rm n}_z\end{bmatrix}, \qquad (7)$$

where the elements of the column matrices f_1 and f_2 and the diagonal elements of the diagonal matrices n_x, n_y and n_z are respectively φ_{mnt}, χ_{mnt}, n_{mntx}, n_{mnty} and n_{mntz}, while the elements ε_{mntuvw} of the square matrix ε are related to the Fourier coefficients ε_{mnt} of $\varepsilon(\mathbf{r})$ in (3) as $\varepsilon_{mntuvw}=\varepsilon_{m-u,n-v,t-w}$.

Inside the crystal, $\varepsilon = \varepsilon^{\mathrm{II}}$. In accordance with matrix theory, the solution of (6) in this region can be formally expressed as

$$\mathrm{f}^{\mathrm{II}}(\bar{z}) = \mathrm{T}^{\mathrm{II}} \exp(i\kappa^{\mathrm{II}}\bar{z})\mathrm{a}^{\mathrm{II}}, \tag{8}$$

with square matrix $\mathrm{T}^{\mathrm{II}} = [\nu_1^{\mathrm{II}}\ \nu_2^{\mathrm{II}} \ldots]$ where the column matrices $\nu_1^{\mathrm{II}}, \nu_2^{\mathrm{II}}, \ldots$ are eigenvectors of the constant square matrix C^{II}, belonging respectively to eigenvalues $\kappa_1^{\mathrm{II}}, \kappa_2^{\mathrm{II}}, \ldots$, which constitute the diagonal elements of the diagonal matrix κ^{II} (arranged in that order). a^{II} is an arbitrary constant column matrix. This results show that the numerical calculation in this diffraction problem is essentially reduced to a standard eigenvalue problem, as in the exact Bloch-wave-expansion approach of Colella (1972).

In the vacuum regions, $\varepsilon = 1$. Here we may set $\mathrm{n}_z = 0$ for convenience. This amounts to absorbing the z-dependent phase factors in (2) into the z-dependent amplitudes. Then the corresponding C matrix is simplified to

$$\mathrm{C}^{v} = \begin{bmatrix} 0 & 1 \\ 1 - \mathrm{n}_x^2 - \mathrm{n}_y^2 & 0 \end{bmatrix}, \tag{9}$$

where the superscript v refers to a vacuum region. Since all the submatrices in C^{v} are diagonal, (6) can be decomposed into

$$\mathrm{d}\mathrm{f}_{mnt}^{v}/\mathrm{d}\bar{z} = i\mathrm{C}_{mnt}^{v}\mathrm{f}_{mnt}^{v} \tag{10}$$

with $\mathrm{f}_{mnt}^{v} = [f_{mnt1}^{v}\ f_{mnt2}^{v}]^{t} = [\varphi_{mnt}^{v}\ \chi_{mnt}^{v}]^{t}$, where the superscript t stands for matrix transpose, and with

$$\mathrm{C}_{mnt}^{v} = \begin{bmatrix} 0 & 1 \\ 1 - n_{mntx}^2 - n_{mnty}^2 & 0 \end{bmatrix}. \tag{11}$$

The eigenvalues of C_{mnt}^{v} and their corresponding eigenvectors are $\kappa_{mnt1}^{v} = \xi_{mnt}$, $\kappa_{mnt2}^{v} = -\xi_{mnt}$, $\nu_{mnt1}^{v} = [1\ \ \xi_{mnt}]^{t}$ and $\nu_{mnt2}^{v} = [1\ \ -\xi_{mnt}]^{t}$, where $\xi_{mnt} = +(1 - n_{mntx}^2 - n_{mnty}^2)^{1/2}$ is positive (real or imaginary). The explicit solution of (10) is then

$$\mathrm{f}_{mnt}^{v}(\bar{z}) = \mathrm{T}_{mnt}^{v} \exp[i\kappa_{mnt}^{v}(\bar{z} - \bar{z}_0^{v})]\mathrm{a}_{mnt}^{v}, \tag{12}$$

where $\kappa_{mnt}^{v} = [\kappa_{mntr}^{v}\delta_{rr'}]$ with r and $r' = 1$ or 2, $\mathrm{T}_{mnt}^{v} = [\nu_{mnt1}^{v}\ \ \nu_{mnt2}^{v}]$, and $\mathrm{a}_{mnt}^{v} = [a_{mnt1}^{v}\ \ a_{mnt2}^{v}]^{t}$. Here $\delta_{rr'}$ denotes a Kronecker delta, while the constant $\bar{z}_0^{v}$ assumes the values 0 and $\bar{d}$ $(=2\pi k_0 d)$ in regions I and III, respectively. Thus, the solution of (6) in a vacuum region is

$$\mathrm{f}^{v}(\bar{z}) = \mathrm{T}^{v} \exp\{i\kappa^{v}(\bar{z} - \bar{z}_0^{v})\}\mathrm{a}^{v} \tag{13}$$

with

$$\mathrm{T}^{v} = \begin{bmatrix} 1 & 1 \\ \xi & -\xi \end{bmatrix}, \qquad \mathrm{a}^{v} = \begin{bmatrix} \mathrm{a}^{v+} \\ \mathrm{a}^{v-} \end{bmatrix}, \tag{14}$$

and $\kappa^{v} = [\kappa_r^{v}\delta_{rr'}]$, where the elements of the column matrices a^{v+}, a^{v-} and the diagonal elements of the diagonal matrices κ_r^{v}, ξ are a_{mnt1}^{v}, a_{mnt2}^{v}, κ_{mntr}^{v} and ξ_{mnt}, respectively.

By substituting the first component of the matrix solution (12) into the wave-function expansion (2) we obtain the wave solution in a vacuum region

$$\psi^{v}(\bar{\mathbf{r}}) = \sum_{m,n,t,r} a_{mntr}^{v} \exp[i\mathbf{n}_{mntr}^{v}\cdot(\bar{\mathbf{r}} - \mathbf{i}_z\bar{z}_0^{v})], \tag{15}$$

where $\mathbf{n}_{mnt1}^{v} = (n_{mntx}, n_{mnty}, \xi_{mnt})$ and $\mathbf{n}_{mnt2}^{v} = (n_{mntx}, n_{mnty}, -\xi_{mnt})$ are the wave vectors of the plane-wave components (normalized by k_0), and $\mathbf{i}_z$ is the unit vector in the z direction. Although n_{mntx} and n_{mnty} are real, ξ_{mnt} can be either real or imaginary, corresponding respectively to propagating and evanescent waves. From these results we see that, for a real ξ_{mnt}, the coefficients a_{mnt1}^{v} and a_{mnt2}^{v} are the amplitudes of the plane waves that propagate with the sense along the z axis being positive and negative, respectively.

3. Boundary conditions and formal solution

The boundary conditions are $\psi^{\mathrm{I}} = \psi^{\mathrm{II}}$ and $\partial\psi^{\mathrm{I}}/\partial z = \partial\psi^{\mathrm{II}}/\partial z$ at $z = 0$, and $\psi^{\mathrm{II}} = \psi^{\mathrm{III}}$ and $\partial\psi^{\mathrm{II}}/\partial z = \partial\psi^{\mathrm{III}}/\partial z$ at $z = d$. These, together with the definitions of φ_{mnt} and χ_{mnt} given in (2) and (5), imply the continuity of $\mathrm{EXP}(i\mathrm{n}_z\bar{z})\mathrm{f}(\bar{z})$, where $\mathrm{EXP}(i\mathrm{n}_z\bar{z}) = [\exp(i\mathrm{n}_z\bar{z})\delta_{rr'}]$. Then we have

$$\mathrm{f}^{\mathrm{I}}(0) = \mathrm{f}^{\mathrm{II}}(0), \tag{16}$$

$$\mathrm{f}^{\mathrm{III}}(\bar{d}) = \mathrm{EXP}(i\mathrm{n}_z^{\mathrm{II}}\bar{d})\mathrm{f}^{\mathrm{II}}(\bar{d}). \tag{17}$$

The simplicity of this presentation of boundary conditions is a result of defining the state variable χ_{mnt} in the form of (5). Substituting (8) and (13) into (16) and (17), and eliminating the common factor a^{II}, we obtain a direct relation between the constant amplitudes of the plane waves in the vacuum regions outside the crystal as

$$\mathrm{a}^{\mathrm{I}} = \mathrm{M}\mathrm{a}^{\mathrm{III}} \tag{18}$$

with

$$\begin{aligned} \mathrm{M} &= \begin{bmatrix} \mathrm{M}_1 & \mathrm{M}_2 \\ \mathrm{M}_3 & \mathrm{M}_4 \end{bmatrix} \\ &= (\mathrm{T}^{v})^{-1}\mathrm{T}^{\mathrm{II}} \exp(-i\kappa^{\mathrm{II}}\bar{d})(\mathrm{T}^{\mathrm{II}})^{-1} \\ &\quad \times \mathrm{EXP}(-i\mathrm{n}_z^{\mathrm{II}}\bar{d})\mathrm{T}^{v}. \end{aligned} \tag{19}$$

The matrix M is related to a scattering matrix S which will be briefly discussed in Appendix *A*.

In this diffraction problem, the given 'initial condition' is that the incident wave is $\psi_{\mathrm{inc}}(\mathbf{r}) = \exp(i2\pi\mathbf{k}_0.\mathbf{r})b$. This implies $\mathrm{a}^{\mathrm{III}-} = 0$ and $\mathrm{a}^{\mathrm{I}+} = b[0 \ldots 0\ 1\ 0 \ldots 0]^{t}$ when the $(0, 0, 0)$th element is arranged in the middle of the column matrix. Then, finally, a formal solution of all the diffraction-wave amplitudes in the vacuum regions outside the crystal can be solved exactly from (18):

$$\mathrm{a}^{\mathrm{III}+} = \mathrm{M}_1^{-1}\mathrm{a}^{\mathrm{I}+}, \tag{20}$$

$$\mathrm{a}^{\mathrm{I}-} = \mathrm{M}_3\mathrm{M}_1^{-1}\mathrm{a}^{\mathrm{I}+}. \tag{21}$$

4. Transmittance, reflectance and Bragg condition

The probability-current conservation law for a stationary state is

$$\oint \mathbf{j}.\mathrm{d}\mathbf{A}=0, \tag{22}$$

where $\mathbf{j}=\mathrm{Re}\,(-ih\psi^*\nabla\psi)/2\pi m_0$ and the integration is over an arbitrary closed surface. The probability current density $\mathbf{j}$ of a stationary scattering state in a coupled-wave expansion involves cross-product terms between all of the plane-wave components. Now we take the integration surface in the shape of a box with its plane surfaces either parallel or perpendicular to the crystal slab, and with the two planes that are parallel to the slab being different vacuum regions. When the lateral sizes of the latter two planes in the x and y directions are much larger than the inverses of the reciprocal-lattice periods in the corresponding directions, the cross-product terms in $\mathbf{j}$ will be approximately zero after integration (see Appendix *B*). The result is a simple but approximate equality, $j_z^{\mathrm{inc}}=\sum(j_{zmnt}^T-j_{zmnt}^R)$, where $j_z^{\mathrm{inc}}=(hk_0/m_0)\xi_{000}|b|^2$, $j_{zmnt}^T=(hk_0/m_0)\,\mathrm{Re}\,(\xi_{mnt})|a_{mnt}^{\mathrm{III}+}|^2$ and $j_{zmnt}^R=-(hk_0/m_0)\,\mathrm{Re}(\xi_{mnt})|a_{mnt}^{\mathrm{I}-}|^2$ are constants. This is an approximate conservation law for the spatially averaged electron density. It becomes

$$\sum_{m,n,t}(T_{mnt}+R_{mnt})=1 \tag{23}$$

when normalized by the incident current, where

$$T_{mnt}=\mathrm{Re}\,(\xi_{mnt}/\xi_{000})|a_{mnt}^{\mathrm{III}+}/b|^2, \tag{24}$$

$$R_{mnt}=\mathrm{Re}\,(\xi_{mnt}/\xi_{000})|a_{mnt}^{\mathrm{I}-}/b|^2. \tag{25}$$

In a practical experiment, it is the electron flux impinging on one of the detecting or recording elements of a finite size which is measured. In other words, one measures only the current density that is spatially averaged over the surface of a detecting element. Thus, when the size of the detecting elements is much larger than the inverses of the reciprocal-lattice periods along the x and y directions, the approximate conservation law (23) is well satisfied. Moreover, a realistic diffraction phenomenon is a time-dependent process of scattering off an incident wave packet. Only the propagating diffraction waves, for which the T_{mnt} and R_{mnt} are non-zero, correspond to the truly transmitted/reflected electrons. Hence, it is physically meaningful to interpret T_{mnt} and R_{mnt} as the transmittance and reflectance (or the diffraction efficiencies) of the (m, n, t)th-order forwardly and backwardly diffracted waves, respectively. These are then the only relevant dynamical quantities to be measured in a realistic time-dependent diffraction process and to be calculated in this time-independent diffraction theory for comparison.

In the extreme case that one of the basic reciprocal-lattice vectors, say $\mathbf{b}_3$, is exactly perpendicular to the crystal surfaces (*i.e.* $b_{3x}=b_{3y}=0$), all the transmitted/reflected waves with the same order numbers m and n but different order number t collapse into a single plane wave. In consequence, the interferences between this subset of waves must be taken into account. That is, their amplitudes must be summed first before calculating the transmittance/reflectance of the (m, n)th-order diffraction waves.

According to the coupled-wave expansion (2), the wave vectors of the (depth-dependent) diffraction waves of all orders inside the crystal are $\mathbf{K}^{\mathrm{II}}+\mathbf{g}$. Owing to the definition of $\mathbf{K}^{\mathrm{II}}$, the zeroth-order diffraction wave may be interpreted as the 'refraction wave' of the given incident wave. It is this refraction wave that may be considered as the wave incident upon the 'unbiased' periodic structure of the permittivity to excite the other diffraction waves. The well known phase-matching requirement of the Bragg (1913) law in diffraction processes (see Kittel, 1976), which corresponds to momentum conservation, should be applied to this incident wave and a single diffraction wave. That is, $(\mathbf{K}^{\mathrm{II}})^2=(\mathbf{K}^{\mathrm{II}}+\mathbf{g})^2$ or $(\mathbf{n}_{mnt}^{\mathrm{II}})^2-\varepsilon_{000}^{\mathrm{II}}=0$. Or, more explicitly,

$$\begin{aligned}
&2\sin\theta^i[m(\lambda/\Lambda_1)\sin\theta_1\cos\varphi_1\\
&\quad+n(\lambda/\Lambda_2)\sin\theta_2\cos\varphi_2+t(\lambda/\Lambda_3)\sin\theta_3\cos\varphi_3]\\
&\quad+2(\varepsilon_{000}^{\mathrm{II}}-\sin^2\theta^i)^{1/2}\\
&\quad\times[m(\lambda/\Lambda_1)\cos\theta_1+n(\lambda/\Lambda_2)\cos\theta_2\\
&\quad+t(\lambda/\Lambda_3)\cos\theta_3]\\
&=-[m(\lambda/\Lambda_1)\sin\theta_1\cos\varphi_1+n(\lambda/\Lambda_2)\sin\theta_2\cos\varphi_2\\
&\quad+t(\lambda/\Lambda_3)\sin\theta_3\cos\varphi_3]^2\\
&\quad-[m(\lambda/\Lambda_1)\sin\theta_1\sin\varphi_1+n(\lambda/\Lambda_2)\sin\theta_2\sin\varphi_2\\
&\quad+t(\lambda/\Lambda_3)\sin\theta_3\sin\varphi_3]^2\\
&\quad-[m(\lambda/\Lambda_1)\cos\theta_1+n(\lambda/\Lambda_2)\cos\theta_2\\
&\quad+t(\lambda/\Lambda_3)\cos\theta_3]^2,
\end{aligned} \tag{26}$$

where we have used $\mathbf{b}_j=(\sin\theta_j\cos\varphi_j, \sin\theta_j\sin\varphi_j, \cos\theta_j)/\Lambda_j$ (with $j=1, 2, 3$) for the basic reciprocal-lattice vectors. This kinematical condition determines the (m, n, t)th-order Bragg angle of incidence, θ_{mnt}^i, if it exists.

When weak dynamical couplings are assumed (*i.e.* $\varepsilon_{mnt}^{\mathrm{II}}\simeq 1$ with the possible exception of $\varepsilon_{000}^{\mathrm{II}}$), the Bragg law may be derived analytically from the coupled-amplitude equation (4) for region II by noting the fact that the coefficient of the amplitude $\varphi_{mnt}^{\mathrm{II}}$ in this equation is $(\mathbf{n}_{mnt}^{\mathrm{II}})^2-\varepsilon_{000}^{\mathrm{II}}$. [For a similar derivation in a Bloch-wave-expansion approach, see Yariv & Yeh (1984).] Clearly, the Bragg law of maximal diffraction efficiency is not generally true, at least not when the interaction between wave and medium is strong. This point has been demonstrated in numerical calculations (Liu & Cheng, 1986; Moharam, Gaylord, Sincerbox, Werlich & Yung, 1984).

5. Summary and discussions

A. Assumptions

This is an exact potential scattering theory based on a simplifying model. In this model, (i) spin effects are ignored entirely, (ii) electron motion is governed by a Schrödinger equation, (iii) the crystal slab is infinite in lateral extension, (iv) the interaction potential is terminated at the crystal surfaces, and (v) the incident wave is a monoenergetic plane wave. But, as a common practice, the effect of the presence of inelastic scattering on elastic scattering may be taken into account as electron 'absorption' by adopting a complex potential (Yoshioka, 1957). In such a lossy diffraction, the averaged permittivity $\varepsilon_{000}^{\rm II}$ in the definition of $\mathbf{K}^{\rm II}$ and hence in the Bragg condition (26) is to be replaced by its real part, $\operatorname{Re}(\varepsilon_{000}^{\rm II})$. Meanwhile, the exact electron-current conservation law (22) and, consequently, the approximate conservation law (23) will no longer hold.

Besides, relativistic kinematic corrections may also be incorporated (Fujiwara, 1961, 1962; Howie, 1962). In this theory we replace the non-relativistic total energy E in the definition of $\varepsilon(\mathbf{r})$ with $W - m_0c^2$ and replace the non-relativistic wavelength $\lambda = h(2m_0E)^{-1/2}$ with the relativistic wavelength $\lambda_r = ch(W^2 - m_0^2c^4)^{-1/2}$, where W is the relativistic total energy and c is the speed of light. These corrections can be justified in a fully relativistic theory for the same diffraction problem analyzed in a similar fashion (Liu, 1987). Clearly, when any one of the assumptions made in such a model is not approximately satisfied in a given diffraction problem, this theory may not be applicable.

B. Method of analysis

This diffraction theory is formulated in a differential-equation approach, known as the rigorous coupled-wave analysis in optical diffraction studies (Gaylord & Moharam, 1982). A rigorous coupled-wave analysis starts with a coupled-wave expansion of the wave field, and reduces the numerical calculation to a standard eigenvalue problem. The present author has also applied this method to formulate a fully relativistic theory based on the Dirac equation (Liu, 1987), although Gevers & David (1982) had independently used the same approach. Both theories start with a coupled-wave expansion of the spinor wave field, in which the spatial parts are nearly the same but the spin states are different. Since the Dirac equation with (Liu, 1987) or without (Gevers & David, 1982) magnetic dipole–dipole interaction is of first order, so also are the resulting coupled-amplitude equations. Therefore, unlike the present theory, the amplitude equations can be directly reduced to a standard eigenvalue problem without recourse to the state-variable technique.

In summary, the rigorous coupled-wave analysis was developed (i) for electron diffraction from a crystal based upon the (first-order) Dirac equation (Gevers & David, 1982), and (ii) for electromagnetic wave diffraction from gratings with a single grating vector based upon the (second-order) Helmholtz equation (Moharam & Gaylord, 1981, 1983*a*) and the (first-order) Maxwell equations (Moharam & Gaylord, 1983*b*; Rokushima & Yamakita, 1983). Transmittance and reflectance are defined in all these theories. The second line of development has been extended to gratings with two grating vectors based on the Helmholtz equation (Liu, 1985) and three grating vectors based on the Maxwell equations (Liu, 1986). The last case includes a rigorous coupled-wave theory for X-ray diffraction from planar ideal crystals as a special case. The theory presented in this paper is a rigorous coupled-wave theory of electron diffraction based on the (second-order) Schrödinger equation.

C. Relations with other exact theories based on the same model

The present theory and the exact theories of Colella (1972), van Dyck (1967), and Lynch & Moodie (1972) are all based on the same model but formulated in different differential-equation approaches. Their exact formal solutions should be equivalent. Their main differences and similarities have been pointed out in the *Introduction* and in the derivation of the present theory (§§ 2 and 3).

D. Numerical calculation scheme

In this theory, the exact solution of the diffraction-wave amplitudes of all orders is formally expressed in explicit infinite-matrix form. Numerical calculation may be easily implemented on digital computers by straightforward programming, when the matrices involved are truncated into a finite form. No special techniques or algorithms are required. Suppose only the waves from (0, 0, 0)th order to all orders up to $(\pm L, \pm L, \pm L)$th are retained. The number of both forwardly and backwardly diffracted waves is then $2N = 2(2L+1)^3$, which is equal to the dimension of the truncated finite matrices involved in numerical calculation.

For comparison, we note that in a fully relativistic and rigorous coupled-wave theory (Gevers & David, 1982; Liu, 1987), the corresponding matrix dimension is doubled [*i.e.* $4N = 4(2L+1)^3$] owing to the existence of two distinct polarization states, while in the approximate theories for spinless electrons (*e.g.* Bethe, 1928; Colella, 1972; Cowley & Moodie, 1957; Howie & Whelan, 1961; Tournarie, 1962; Lynch & Moodie, 1972; Sturkey, 1962), the number of waves retained is N since either the back scattering or the forward scattering is usually neglected. The computer memory requirement for calculation in these theories is roughly proportional to the squared value of the number of waves, except for the physical-optics approach of Cowley & Moodie (1957) in which it is proportional only to the number of waves (Goodman & Moodie, 1974).

APPENDIX *A*

Scattering matrix

An equivalent form of formula (18) is

$$\begin{bmatrix} \mathsf{a}^{\mathrm{I}-} \\ \mathsf{a}^{\mathrm{III}+} \end{bmatrix} = \mathsf{S} \begin{bmatrix} \mathsf{a}^{\mathrm{I}+} \\ \mathsf{a}^{\mathrm{III}-} \end{bmatrix} \tag{A1}$$

with

$$\mathsf{S} = \begin{bmatrix} \mathsf{M}_3\mathsf{M}_1^{-1} & \mathsf{M}_4 - \mathsf{M}_3\mathsf{M}_1^{-1}\mathsf{M}_2 \\ \mathsf{M}_1^{-1} & -\mathsf{M}_1^{-1}\mathsf{M}_2 \end{bmatrix}. \tag{A2}$$

Since the matrix S transforms the amplitudes of incident waves into those of diffracted waves, it is the scattering matrix in this rigorous coupled-wave theory of electron diffraction from a crystal with infinite lateral extension. But it is quite different from the scattering matrix defined in the usual scattering-theory formalism for a finite-ranged scattering potential [see, for example, Taylor (1972) or Rodberg & Thaler (1967)]. The scattering matrix S here is not unitary even for real scattering potentials, since the plane-wave functions on which S is defined are not complete orthogonal functions. In particular, this set of waves contains evanescent waves. It is not the complete set of free-particle wave functions used in the usual scattering theory. An exact conservation law can be readily derived from (*A*1):

$$\sum_{m,n,t} (|a_{mnt}^{\mathrm{I}-}/b|^2 + |a_{mnt}^{\mathrm{III}+}/b|^2) = (\mathsf{S}^{*\prime}\mathsf{S})_{00010001}. \tag{A3}$$

It appears, however, that the terms on the left-hand side cannot be interpreted as probabilities.

APPENDIX *B*

Approximate probability-current conservation law

The probability current density **j** contains the product terms between each pair of the plane-wave components of the stationary state. The surface integral of the product term between the (m, n, t)th and (u, v, w)th plane waves over the box surface described in the main text involves the factor

$$\operatorname{sinc}(L_x\{(m-u)b_{1x} + (n-v)b_{2x} + (t-w)b_{3x}\})$$

and/or

$$\operatorname{sinc}(L_y\{(m-u)b_{1y} + (n-v)b_{2y} + (t-w)b_{3y}\}),$$

where $\operatorname{sinc} x = (\sin \pi x)/\pi x$ and L_x, L_y are the lateral sizes of the box in the x and y directions. If L_x and L_y are chosen large enough such that $L_xb_{jx} \gg 1$ and $L_yb_{jy} \gg 1$ for $j = 1$, 2, and 3, the sinc functions will both approach $\delta_{mu}\delta_{nv}\delta_{tw}$. Consequently, all of the cross product (interference) terms involved in equation (22) will approximately vanish and the approximate conservation law (23) follows. The corresponding result for electromagnetic wave diffraction from gratings with a single grating vector was shown by Russell (1984).

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