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2003 J. Phys. A: Math. Gen. 36 803

(http://iopscience.iop.org/0305-4470/36/3/314)

INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS A: MATHEMATICAL AND GENERAL

J. Phys. A: Math. Gen. **36** (2003) 803–816 PII: S0305-4470(03)34209-X

A useful expansion of the exponential of the sum of two non-commuting matrices, one of which is diagonal

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Received 25 February, in final form 24 July 2002
Published 7 January 2003
Online at stacks.iop.org/JPhysA/36/803

Abstract

The matrix exponential plays an important role in solving systems of linear differential equations. We will give a general expansion of the matrix exponential $S = \exp[\lambda(A + B)]$ as $S_{n,m} = e^{\lambda b_n}\delta_{n,m} + \sum_{q=1}^{\infty} \sum_{l_1=0}^{N} \cdots \sum_{l_{q-1}=0}^{N} a_{n,l_1} \cdots a_{l_{q-1},m} C^{(q)}_{n,l_1,\ldots,l_{q-1},m}(B, \lambda)$ with $C^{(q)}_{n,l_1,\ldots,l_{q-1},m}(B, \lambda)$ being an analytical expression in $b_n, b_{l_1}, b_{l_2}, \ldots b_{l_{q-1}}, b_m$, and the scalar coefficient λ. A is a general $N \times N$ matrix with elements $a_{n,m}$ and B a diagonal matrix with elements $b_{n,m} = b_n\delta_{n,m}$ along its diagonal. The convergence of this expansion is shown to be superior to the Taylor expansion in terms of $(\lambda[A + B])$, especially if elements of B are larger than the elements of A. The convergence and possibility of solving the phase problem through multiple scattering is demonstrated by using this expansion for the computation of large-angle convergent beam electron diffraction pattern intensities.

PACS numbers: 03.65.Nk, 02.30.−f

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Mathematical models of many problems in biology, physics and economics involve systems of linear, constant coefficient equations

$$\frac{\delta x}{\delta \lambda}(\lambda) = Ax(\lambda).$$

Such systems of equations have the general solution

$$x(\lambda) = e^{\lambda A}x(0).$$

As an example the solution of probably the most fundamental equation in modern physics, the Schrödinger equation,

$$\mathrm{i}\frac{\delta\Psi(\vec{r},t)}{\delta t} = H\Psi(\vec{r},t)$$

falls into this class, where the Hamiltonian $H = \hbar/(2m|e|)\nabla^2 + V$ is a square matrix that can be split into a potential energy term, the matrix V and a diagonal matrix containing the kinetic energy terms. Assuming that the wavefunction $\Psi(\vec{r},t) = \Phi(\vec{r})\exp(2\pi\mathrm{i}[k_z + Et/h])$ has the form of a modulated plane wave and the second-order derivative of $\Phi(\vec{r})$ in the direction of its propagation (z) is negligible, the time-independent Schrödinger equation can also be written as a system of linear differential equations in z

$$\frac{\delta\Phi(\vec{r})}{\delta z} = \left[\left(\frac{\mathrm{i}\nabla_{xy}^2}{4\pi k_z} - \frac{\vec{k}_{xy}}{k_z}\cdot\vec{\nabla}_{xy}\right) + \frac{\mathrm{i}}{4\pi k_z}V(\vec{r})\right]\Phi(\vec{r}) = 0 \tag{1}$$

where we have the general matrix of potential energy terms $\mathrm{i}V/(4\pi k_z)$ and a diagonal matrix for the kinetic energy. In reciprocal space the differential operators ∇_{xy}^2 and ∇_{xy} turn into linear operators, making this also a problem of the class treated in this paper. A prominent example would be multiple scattering of high energy particles in matter, or rather its inversion, where the problem is that of determining the (periodic) scattering potential $V(\vec{r})$ from the scattered radiation, which is equivalent to finding A from the moduli of the entries of a single column of the matrix $S = \exp(\lambda[A+B])$, where B is a diagonal matrix with known elements. Varying the elements in B lets us set up many of these systems of equations, but unless we can find some linear approximation for them, they will still remain hard to solve directly. The purpose of this paper is to provide a general expansion which allows us to express the matrix exponential $S = \exp(\lambda[A+B])$ as

$$S_{n,m} = \mathrm{e}^{\lambda b_n}\delta_{n,m} + \sum_{q=1}^{\infty}\sum_{l_1=1}^{N}\cdots\sum_{l_{q-1}=1}^{N} a_{n,l_1}\cdots a_{l_{q-1},m}C^{(q)}_{n,l_1,\ldots,l_{q-1},m}(B,\lambda)$$

where $a_{n,m}$ are elements of A, and $C^{(q)}_{n,l_1,\ldots,l_{q-1},m}(B,\lambda)$ is an expression in elements of B and λ.

The convergence of this expansion will be discussed and is shown to be superior to the Taylor expansion in terms of $(\lambda[A+B])$, especially if the largest elements of B are larger than the largest element of A.

This expansion provides a disentanglement of the matrix exponential into a sum of terms containing different length products of elements of A with known coefficients. In the field of high energy electron diffraction (HEED) in the transmission geometry, the direct inversion of convergent beam (CBED) diffraction patterns to structure factors is a problem of exactly this type [1]. CBED patterns, whose scattering intensity, expressed in the matrix exponential Bloch wave formalism, provide experimental data for the moduli of certain elements in the matrix S for fixed structure factors in the matrix A, but many different matrices B. The application of this expansion to the inversion of CBED patterns will therefore be discussed in section 3.

2. Expansion of the matrix exponential

There exist many different ways to compute the matrix exponential $S = \exp(M)$. Nineteen of them are listed in [2]. Computing the matrix exponential from its Taylor expansion is probably one of the least efficient ways. However, this method involves only integer powers of matrix elements, allowing every element of S to be expressed as a polynomial of elements in M in a straightforward way. The matrix exponential is defined as

$$S = \mathrm{e}^{\lambda(A+B)} = \lim_{p\to\infty} \sum_{j=0}^{p} \frac{\lambda^j}{j!}(A+B)^j \tag{2}$$

where

$$[(A+B)^j]_{n,m} = \sum_{l_1=1}^{N}\sum_{l_2=1}^{N}\cdots\sum_{l_{j-1}=1}^{N}\left(a_{n,l_1}+b_{n,l_1}\right)\left(a_{l_1,l_2}+b_{l_1,l_2}\right)\cdots\left(a_{l_{j-1},m}+b_{l_{j-1},m}\right)$$

$$= b_n^j\delta_{n,m} + \sum_{q=1}^{j}\underbrace{\sum_{l_1=1}^{N}\sum_{l_2=1}^{N}\cdots\sum_{l_{q-1}=1}^{N}}_{q-1} a_{n,l_1}a_{l_1,l_2}\cdots a_{l_{q-1},m}$$

$$\times \sum_{j_0=0}^{j-q}\sum_{j_1=0}^{j-q-j_1}\cdots\sum_{j_{q-1}=0}^{j-q-\sum_{i=0}^{q-2}j_i} b_n^{j_0}b_{l_1}^{j_1}\cdots b_{l_{q-1}}^{j_{q-1}}b_m^{j-q-\sum_{i=0}^{q-1}j_i}$$

$$= b_n^j\delta_{n,m} + \sum_{q=1}^{j}\sum_{l_1=1}^{N}\sum_{l_2=1}^{N}\cdots\sum_{l_{q-1}=1}^{N} a_{n,l_1}a_{l_1,l_2}\cdots a_{l_{q-1},m}C^{(j-q,q)}_{n,l_1,\ldots,l_{q-1},m} \tag{3}$$

and (setting $l_0 = n, l_q = m$)

$$C^{(j,q)}_{l_0,l_1,\ldots,l_{q-1},l_q} = b_{l_q}^j\underbrace{\sum_{j_0=0}^{j}\left(\frac{b_{l_0}}{b_{l_q}}\right)^{j_0}\sum_{j_1=0}^{j-j_0}\left(\frac{b_{l_1}}{b_{l_q}}\right)^{j_1}\cdots\sum_{j_{q-1}=0}^{j-\sum_{i=0}^{q-2}j_i}\left(\frac{b_{l_{q-1}}}{b_{l_q}}\right)^{j_{q-1}}}_{q}.$$

If $b_{l_{q-1}} = b_{l_q}$ then the last sum is just $j+1-\sum_{i=0}^{q-2} j_i$ which gives

$$C^{(j,q)}_{l_0,l_1,\ldots,l_{q-1},l_q} = \frac{\delta}{\delta b_{l_{q-1}}}\left(b_{l_{q-1}}^{j+1}\sum_{j_0=0}^{j}\left(\frac{b_{l_0}}{b_{l_{q-1}}}\right)^{j_0}\cdots\sum_{j_{q-2}=0}^{j-\sum_{i=0}^{q-3}j_i}\left(\frac{b_{l_{q-2}}}{b_{l_{q-1}}}\right)^{j_{q-2}}\right).$$

In general we may have $b_{l_{q-d}} = \cdots = b_{l_{q-2}} = b_{l_{q-1}} = b_{l_q}, 0 \leqslant d \leqslant q$. Note, that we will also encounter such degeneracies if all the elements of B are distinct since elements of B will also be multiplied by themselves in the course of the matrix multiplication. Using

$$\sum_{j_1=0}^{N}\sum_{j_2=0}^{N-j_1}\cdots\sum_{j_d=0}^{N-\sum_{i=1}^{d}j_i} 1 = \frac{(N+1)(N+2)\cdots(N+d)}{d!} = \binom{N+d}{d} \tag{4}$$

we get

$$C^{(j,q)}_{l_0,l_1,\ldots,l_{q-1},l_q} = \frac{1}{d!}\left(\frac{\delta}{\delta b_{l_{q-d}}}\right)^d\left(b_{l_{q-d}}^{j+d}\sum_{j_0=0}^{j}\left(\frac{b_{l_0}}{b_{l_{q-d}}}\right)^{j_0}\cdots\sum_{j_{q-d-1}=0}^{j-\sum_{i=0}^{q-d-2}j_i}\left(\frac{b_{l_{q-d-1}}}{b_{l_{q-d}}}\right)^{j_{q-d-1}}\right)$$

$$= \frac{1}{d!}\left(\frac{\delta}{\delta b_{l_{q-d}}}\right)^d b_{l_{q-d}}^d C^{(j,q-d)}_{l_0,l_1,\ldots,l_{q-d}}. \tag{5}$$

An equivalent expression is given in [3]. Using the method of perturbation and passage to the limits we will treat the degenerate case as $\lim_{\epsilon_1,\ldots,\epsilon_d\to 0} C^{(j,q)}_{l_0,l_1,\ldots,l_{q-1},l_q}$, where $b_{l_{q-1}} = b_{l_q}(1+\epsilon_1)\cdots b_{l_{q-d}} = b_{l_q}(1+\epsilon_d)$. We will therefore now first concentrate on the non-degenerate case.

If $b_{l_{q-1}} \neq b_{l_q}$ the last sum is a finite geometric series:

$$C^{(j,q)}_{l_0,l_1,\ldots,l_{q-1},l_q} = b^j_{l_q} \sum_{j_0=0}^{j} \left(\frac{b_{l_0}}{b_{l_q}}\right)^{j_0} \cdots \sum_{j_{q-2}=0}^{j-\sum_{i=0}^{q-3} j_i} \left(\frac{b_{l_{q-2}}}{b_{l_q}}\right)^{j_{q-2}} \frac{1-\left(\frac{b_{l_{q-1}}}{b_{l_q}}\right)^{j+1-\sum_{i=0}^{q-2} j_i}}{1-\left(\frac{b_{l_{q-1}}}{b_{l_q}}\right)}$$

$$= \frac{1}{1-\left(\frac{b_{l_{q-1}}}{b_{l_q}}\right)} b^j_{l_q} \sum_{j_0=0}^{j} \left(\frac{b_{l_0}}{b_{l_q}}\right)^{j_0} \cdots \sum_{j_{q-2}=0}^{j-\sum_{i=0}^{q-3} j_i} \left(\frac{b_{l_{q-2}}}{b_{l_q}}\right)^{j_{q-2}}$$

$$- \frac{\left(\frac{b_{l_{q-1}}}{b_{l_q}}\right)}{1-\left(\frac{b_{l_{q-1}}}{b_{l_q}}\right)} b^j_{l_{q-1}} \sum_{j_0=0}^{j} \left(\frac{b_{l_0}}{b_{l_{q-1}}}\right)^{j_0} \cdots \sum_{j_{q-2}=0}^{j-\sum_{i=0}^{q-3} j_i} \left(\frac{b_{l_{q-2}}}{b_{l_{q-1}}}\right)^{j_{q-2}}$$

$$= \frac{b_{l_{q-1}}}{b_{l_{q-1}} - b_{l_q}} C^{(j,q-1)}_{l_0,l_1,\ldots,l_{q-1}} + \frac{b_{l_q}}{b_{l_q} - b_{l_{q-1}}} C^{(j,q-1)}_{l_0,l_1,\ldots,l_{q-2},l_q} \tag{6}$$

and in particular for $q = 0, 1$:

$$C^{(j,0)}_{l_0} = b^j_{l_0}$$

$$C^{(j,1)}_{l_0,l_1} = \begin{cases} \frac{b^{j+1}_{l_0}}{b_{l_0}-b_{l_1}} + \frac{b^{j+1}_{l_1}}{b_{l_1}-b_{l_0}} & \text{if} \quad b_{l_0} \neq b_{l_1} \\ \frac{\delta}{\delta b_0} b^{j+1}_{l_0} = \binom{j+1}{1} \frac{b^{j+1}_{l_0}}{b_{l_0}} & \text{if} \quad b_{l_0} = b_{l_1}. \end{cases}$$

In general, for non-degenerate b_{l_k} $(k = 0, \ldots, q)$ we get the following result:

$$C^{(j,q)}_{l_0,l_1,\ldots,l_q} = \sum_{k=0}^{q} b^{j+q}_{l_k} \prod_{\substack{r=0 \\ r\neq k}}^{q} \left(b_{l_k} - b_{l_r}\right)^{-1}. \tag{7}$$

We will give the proof by induction

$$C^{(j,q+1)}_{l_0,\ldots,l_q,l_{q+1}} = \frac{b_{l_q}}{b_{l_q} - b_{l_{q+1}}} C^{(j,q)}_{l_0,\ldots,l_q} + \frac{b_{l_{q+1}}}{b_{l_{q+1}} - b_{l_q}} C^{(j,q)}_{l_0,\ldots,l_{q-1},l_{q+1}} = \sum_{k=0}^{q-1} b^{j+q}_{l_k} \prod_{\substack{r=0 \\ r\neq k}}^{q-1} \left(b_{l_k} - b_{l_r}\right)^{-1}$$

$$\times \left(\frac{b_{l_q}}{\left(b_{l_q} - b_{l_{q+1}}\right)\left(b_{l_k} - b_{l_q}\right)} + \frac{b_{l_{q+1}}}{\left(b_{l_{q+1}} - b_{l_q}\right)\left(b_{l_k} - b_{l_{q+1}}\right)} \right)$$

$$+ \frac{b^{j+q+1}_{l_q}}{b_{l_q} - b_{l_{q+1}}} \prod_{r=0}^{q-1} \left(b_{l_q} - b_{l_r}\right)^{-1} + \frac{b^{j+q+1}_{l_{q+1}}}{b_{l_{q+1}} - b_{l_q}} \prod_{r=0}^{q-1} \left(b_{l_{q+1}} - b_{l_r}\right)^{-1}$$

$$= \sum_{k=0}^{q-1} b^{j+q}_{l_k} \prod_{\substack{r=0 \\ r\neq k}}^{q-1} \left(b_{l_k} - b_{l_r}\right)^{-1} \frac{b_{l_k}}{\left(b_{l_k} - b_{l_{q+1}}\right)\left(b_{l_k} - b_{l_q}\right)}$$

$$+ b^{j+q+1}_{l_q} \prod_{\substack{r=0 \\ r\neq q}}^{q+1} \left(b_{l_q} - b_{l_r}\right)^{-1} + b^{j+q+1}_{l_{q+1}} \prod_{\substack{r=0 \\ r\neq q+1}}^{q+1} \left(b_{l_{q+1}} - b_{l_r}\right)^{-1}$$

$$= \sum_{k=0}^{q+1} b^{j+q+1}_{l_k} \prod_{\substack{r=0 \\ r\neq k}}^{q+1} \left(b_{l_k} - b_{l_r}\right)^{-1} = C^{(j,q+1)}_{l_0,\ldots,l_q,l_{q+1}}.$$

Let us now consider the degenerate case. As already stated above, the degenerate case is unavoidable for $q > 2$, even if the diagonal elements in B are distinct. Let us assume that the values $b_{l_0}, \ldots, b_{l_d}$ $(d \geqslant 0)$ are almost degenerate, i.e. $b_{l_0} = b'_{l_0} + \epsilon_0, \ldots, b_{l_d} = b'_{l_0} + \epsilon_d$. Then the degenerate case is defined by the limit $\epsilon_0, \ldots, \epsilon_d \to 0$.

$$C^{(j,q)}_{l_0,\ldots,l_q} = \lim_{\epsilon_1,\ldots,\epsilon_d \to 0} \left[\sum_{k=0}^{d} \left(b'_{l_0} + \epsilon_k\right)^{j+q} \prod_{\substack{r=0 \\ r\neq k}}^{d} \frac{1}{\epsilon_k - \epsilon_r} \prod_{r=d+1}^{q} \frac{1}{\left(\left(b'_{l_0} + \epsilon_k\right) - b_{l_r}\right)} \right.$$
$$\left. + \sum_{k=d+1}^{q} b_{l_k}^{j+q} \prod_{r=0}^{d} \frac{1}{b_{l_k} - \left(b'_{l_0} + \epsilon_k\right)} \prod_{\substack{r=d+1 \\ r\neq k}}^{q} \left(b_{l_k} - b_{l_r}\right)^{-1} \right].$$

Since the limit is independent of the direction from which we approach it, we choose $\epsilon_k = \epsilon\, \mathrm{e}^{\mathrm{i}k\phi_0}$, with $\phi_0 = 2\pi/(d+1)$, i.e. $\epsilon_0 = \epsilon, \epsilon_1 = \epsilon\, \mathrm{e}^{\mathrm{i}\phi_0}$, etc. With this choice we can make use of the following equalities:

$$\sum_{k=0}^{d} \mathrm{e}^{\mathrm{i}k\phi_0} = \sum_{k=0}^{d} (\mathrm{e}^{\mathrm{i}k\phi_0})^s = \left(\sum_{k=0}^{d} \mathrm{e}^{\mathrm{i}k\phi_0} \right)^s = 0 \qquad s = 1, 2, \ldots \tag{8}$$

$$\sum_{\substack{k_1=0 \\ k_1\neq k}}^{d} (\mathrm{e}^{\mathrm{i}k_1\phi_0})^s = -\mathrm{e}^{\mathrm{i}sk\phi_0} \tag{9}$$

$$\sum_{\substack{k_1=0 \\ k_1\neq k}}^{d} \sum_{\substack{k_2=0 \\ k_2\neq k,k_1}}^{d} \cdots \sum_{\substack{k_s=0 \\ k_s\neq k,k_1,\ldots,k_{s-1}}}^{d} \mathrm{e}^{\mathrm{i}k_1\phi_0}\, \mathrm{e}^{\mathrm{i}k_2\phi_0} \cdots \mathrm{e}^{\mathrm{i}k_s\phi_0} = (-1)^s s!\, \mathrm{e}^{\mathrm{i}sk\phi_0} \tag{10}$$

$$\prod_{\substack{k'=0 \\ k'\neq k}}^{d} \frac{1}{\mathrm{e}^{\mathrm{i}k\phi_0} - \mathrm{e}^{\mathrm{i}k'\phi_0}} = \frac{\mathrm{e}^{\mathrm{i}k\phi_0}}{d+1}. \tag{11}$$

We will also define a new variable name: $b_{0,r} = b'_{l_0} - b_{l_r}$.

$$C^{(j,q)}_{l_0,\ldots,l_q} = \lim_{\epsilon\to 0} \left[\sum_{k=0}^{d} \left\{ \left(\sum_{j'=0}^{j+q} \binom{j+q}{j'} b_{l_0}^{j+q-j'} \epsilon^{j'}\, \mathrm{e}^{\mathrm{i}j'k\phi_0} \right) \epsilon^{-d} \prod_{\substack{r=0 \\ r\neq k}}^{d} \frac{1}{\mathrm{e}^{\mathrm{i}k\phi_0} - \mathrm{e}^{\mathrm{i}r\phi_0}} \right.\right.$$
$$\left.\left. \times \prod_{\substack{r=d+1 \\ r\neq k}}^{q} \frac{1}{(b_{0,r} + \epsilon\, \mathrm{e}^{\mathrm{i}k\phi_0})} \right\} \right] + \sum_{k=d+1}^{q} \frac{b_{l_k}^{j+q}}{\left(b_{l_k} - b_{l_0}\right)^{d+1}} \prod_{\substack{r=d+1 \\ r\neq k}}^{q} \left(b_{l_k} - b_{l_r}\right)^{-1}$$
$$= \sum_{j'=0}^{j+q} \binom{j+q}{j'} b_{l_0}^{j+q-j'} \frac{1}{(d+1)}$$
$$\times \lim_{\epsilon\to 0} \left[\frac{\sum_{k=0}^{d} \mathrm{e}^{\mathrm{i}k\phi_0}\, \mathrm{e}^{\mathrm{i}j'k\phi_0} \epsilon^{j'} \prod_{\substack{k'=0 \\ k'\neq k}}^{d} \prod_{r=d+1}^{q} (b_{0,r} + \epsilon\, \mathrm{e}^{\mathrm{i}k'\phi_0})}{\epsilon^d \prod_{k'=0}^{d} \prod_{r=d+1}^{q} (b_{0,r} + \epsilon\, \mathrm{e}^{\mathrm{i}k'\phi_0})} \right]$$
$$+ \sum_{k=d+1}^{q} \frac{b_{l_k}^{j+q}}{\left(b_{l_k} - b_{l_0}\right)^{d+1}} \prod_{\substack{r=d+1 \\ r\neq k}}^{q} \left(b_{l_k} - b_{l_r}\right)^{-1} \tag{12}$$

using (11). For $i > d$ the limit of the expression in square brackets is zero. For $i \leqslant d$ we need to evaluate that expression explicitly. Using (8) and some thought we get for the denominator

$$\epsilon^d \prod_{k'=0}^{d} \prod_{r=d+1}^{q} (b_{0,r} + \mathrm{e}^{\mathrm{i}k'\phi_0}\epsilon) = \epsilon^d \prod_{k'=0}^{d} \prod_{r=d+1}^{q} b_{0,r} = \epsilon^d \left(\prod_{r=d+1}^{q} b_{0,r} \right)^{d+1}. \tag{13}$$

The numerator needs to be expanded in powers of ϵ. The product

$$\prod_{\substack{k'=0 \\ k'\neq k}}^{d} \prod_{r=d+1}^{q} (b_{0,r} + \epsilon\, \mathrm{e}^{\mathrm{i}k'\phi_0}) = \sum_{s=0}^{d(q-d)} \epsilon^s P^{(s)}(k, d, q, \phi_0)$$

over all possible combinations of r and $k' \neq k$ can be expressed in a sum of terms, each involving a certain power of ϵ. The first one, of course is just the product over all the first terms in the sum $(b_{0,r} + \epsilon\, \mathrm{e}^{\mathrm{i}k'\phi_0})$, i.e.

$$P^{(0)}(d, q) = \prod_{r=d+1}^{q} b_{0,r}^d \tag{14}$$

where the power d comes from the fact that there are d different values of $k' = 0, \ldots, d$, $k' \neq k$. The next few terms are

$$\epsilon P^{(1)}(k, d, q, \phi_0) = \epsilon \prod_{r=d+1}^{q} b_{0,r}^d \left[\sum_{r=d+1}^{q} \frac{\sum_{\substack{k_1=0 \\ k_1\neq k}}^{d} \mathrm{e}^{\mathrm{i}k_1\phi_0}}{b_{0,r}} \right] = -\epsilon\, \mathrm{e}^{\mathrm{i}k\phi_0} \prod_{r=d+1}^{q} b_{0,r}^d \left[\sum_{r=d+1}^{q} \frac{1}{b_{0,r}} \right] \tag{15}$$

$$\begin{aligned} \epsilon^2 P^{(2)}(k, d, q, \phi_0) = \frac{1}{2!}\epsilon^2 \prod_{r=d+1}^{q} b_{0,r}^d \Bigg[& \sum_{r_1=d+1}^{q} \sum_{\substack{r_2=d+1 \\ r_2\neq r_1}}^{q} \frac{\left(\sum_{\substack{k_1=0 \\ k_1\neq k}}^{d} \mathrm{e}^{\mathrm{i}k_1\phi_0} \right)^2}{b_{0,r_1} b_{0,r_2}} \\ & + \sum_{r=d+1}^{q} \frac{\sum_{\substack{k_1=0 \\ k_1\neq k}}^{d} \sum_{\substack{k_2=0 \\ k_2\neq k, k_1}}^{d} \mathrm{e}^{\mathrm{i}k_1\phi_0}\, \mathrm{e}^{\mathrm{i}k_2\phi_0}}{b_{0,r}^2} \Bigg] \\ = \frac{1}{2!}\epsilon^2 2!\, \mathrm{e}^{\mathrm{i}2k\phi_0} \prod_{r=d+1}^{q} b_{0,r}^d & \left[\sum_{r_1=d+2}^{q} \sum_{r_2=d+1}^{r_1-1} \frac{1}{b_{0,r_1} b_{0,r_2}} + \sum_{r=d+1}^{q} \frac{1}{b_{0,r}^2} \right] \end{aligned} \tag{16}$$

where we used (9) in (15) and (9) and (10) in (16). The $1/2!$ term accounts for the times we double counted terms when performing the double sum. The left-hand side of equation (16) demonstrates that we have to carefully separate products containing the same index r twice from those that do not. If the same index r appears twice we need to make sure that we do not also have the same k_1 index twice in the same term. This is because we are trying to find all possible combinations of sums $(b_{0,r} + \epsilon\, \mathrm{e}^{\mathrm{i}k'\phi_0})$ out of a given pool without multiplying one with itself. This method can be extended to any power $s > 0$ of ϵ:

$$P^{(s)}(k,d,q,\phi_0) = \frac{1}{s!}\prod_{r=d+1}^{q} b_{0,r}^{d}\left[s!\sum_{r_1=d+s}^{q}\cdots\sum_{r_s=d+1}^{r_{s-1}-1}\frac{(-\mathrm{e}^{\mathrm{i}k\phi_0})^s}{b_{0,r_1}\cdots b_{0,r_s}}\right.$$
$$+\binom{s}{2}\sum_{r=d+1}^{q}\frac{(-1)^2 2!\,\mathrm{e}^{\mathrm{i}2k\phi_0}}{b_{0,r}^2}\left((s-2)!\sum_{\substack{r_1=d+s-2\\ r_1\neq r}}^{q}\cdots\sum_{\substack{r_{s-2}=d+1\\ r_{s-2}\neq r}}^{r_{s-3}-1}\frac{(-\mathrm{e}^{\mathrm{i}k\phi_0})^{s-2}}{b_{0,r_1}\cdots b_{0,r_{s-2}}}\right.$$
$$+\binom{s-2}{2}\sum_{r_1=d+1}^{r}-1\frac{(-1)^2 2!\,\mathrm{e}^{\mathrm{i}2k\phi_0}}{b_{0,r_1}^2}(s-4)!\sum_{\substack{r_2=d+s-2\\ r_2\neq r,r_1}}^{q}\cdots$$
$$\left.\left.\left.\times\sum_{\substack{r_{s-3}=d+1\\ r_{s-4}\neq r,r_1}}^{r_{s-4}-1}\frac{(-\mathrm{e}^{\mathrm{i}k\phi_0})^{s-2}}{b_{0,r_2}\cdots b_{0,r_{s-3}}}+\cdots\right)+\cdots+\binom{s}{s}\sum_{r=d+1}^{q}\frac{(-1)^s s!\,\mathrm{e}^{\mathrm{i}sk\phi_0}}{b_{0,r}^s}(s-s)!\right].$$

Writing down a few of those terms one will soon realize that the binomial coefficients due to the terms with products in which the same $b_{0,r}$ occurs repeatedly always cancel with the factorials that stem from the products with different $b_{0,r}$. We can therefore combine all those different sums into a single s-dimensional sum:

$$P^{(s)}(k,d,q,\phi_0) = \prod_{r=d+1}^{q} b_{0,r}^{d}(-1)^s\,\mathrm{e}^{\mathrm{i}sk\phi_0}\left[\sum_{r_1=d+1}^{q}\frac{1}{b_{0,r_1}}\sum_{r_2=d+1}^{r_1}\frac{1}{b_{0,r_2}}\cdots\sum_{r_s=d+1}^{r_{s-1}}\frac{1}{b_{0,r_s}}\right]. \tag{17}$$

Applying equation (8) we get

$$\sum_{k=0}^{d}\mathrm{e}^{\mathrm{i}k\phi_0}\,\mathrm{e}^{\mathrm{i}j'k\phi_0}\,\mathrm{e}^{\mathrm{i}sk\phi_0} = \sum_{k=0}^{d}\mathrm{e}^{2\pi\mathrm{i}k(j'+1+s)/(d+1)}$$
$$= \begin{cases}(d+1) & \text{if}\quad (j'+1+s)/(d+1) = 1,2,\ldots\\ 0 & \text{else.}\end{cases}$$

Since we have the upper limit of $s \leqslant d - j'$, the only non-vanishing term is the one with $s = d - j'$, for which also the powers of ϵ cancel. This allows us to write down the limit of the expression in square brackets in (12)

$$\lim_{\epsilon\to 0}\left[\frac{\sum_{k=0}^{d}\mathrm{e}^{\mathrm{i}k\phi_0}\,\mathrm{e}^{\mathrm{i}j'k\phi_0}\epsilon^{j'}\prod_{\substack{k'=0\\ k'\neq k}}^{d}\prod_{r=d+1}^{q}(b_{0,r}+\epsilon\,\mathrm{e}^{\mathrm{i}k'\phi_0})}{\epsilon^d\prod_{k'=0}^{d}\prod_{r=d+1}^{q}(b_{0,r}+\epsilon\,\mathrm{e}^{\mathrm{i}k'\phi_0})}\right]$$
$$= \lim_{\epsilon\to 0}\left[\sum_{k=0}^{d}\epsilon^{j'-d}\,\mathrm{e}^{\mathrm{i}(j'+1)k\phi_0}\prod_{\substack{k'=0\\ k'\neq k}}^{d}\prod_{r=d+1}^{q}(b_{0,r}+\epsilon\,\mathrm{e}^{\mathrm{i}k'\phi_0})\right]$$
$$= (d+1)(-1)^{d-j'}\prod_{r=d+1}^{q}b_{0,r}^{d}\left[\sum_{r_1=d+1}^{q}\frac{1}{b_{0,r_1}}\sum_{r_2=d+1}^{r_1}\frac{1}{b_{0,r_2}}\cdots\sum_{r_{d-j'}=d+1}^{r_{d-j'-1}}\frac{1}{b_{0,r_{d-j'}}}\right].$$

If we define $\{b'_{l_0},\ldots,b'_{l_u}\}$ as the set of unique b_{l_k} from the whole set $\{b_{l_0},\ldots,b_{l_q}\}$, each associated with a degeneracy d_k, we can write (12) in a more general form ($d_k = 0$, if b'_{l_k} is

unique, $d_k = 1$, if b'_{l_k} is singly degenerate, i.e. $b'_{l_k} = b_{l_{k1}} = b_{l_{k2}}$, etc).

$$C^{(j,q)}_{l_0,l_1,\ldots,l_q} = \sum_{k=0}^{u}\sum_{j'=0}^{d_k}\binom{j+q}{j'} b'^{j+q-j'}_{l_k}(-1)^{d_k-j'}\frac{\prod_{\substack{r=0\\r\neq k}}^{u}\left(b'_{l_k}-b'_{l_r}\right)^{d_k(d_r+1)}}{\prod_{\substack{r=0\\r\neq k}}^{u}\left(b'_{l_k}-b'_{l_r}\right)^{(d_k+1)(d_r+1)}}$$
$$\times\left[\sum_{\substack{r_1=0\\b_{l_{r_1}}\neq b'_{l_k}}}^{q}\frac{1}{\left(b'_{l_k}-b_{l_{r_1}}\right)}\sum_{\substack{r_2=0\\b_{l_{r_2}}\neq b'_{l_k}}}^{r_1}\frac{1}{\left(b'_{l_k}-b_{l_{r_2}}\right)}\cdots\sum_{\substack{r_{d_k-j'}=0\\b_{l_{r_{d_k-j'}}}\neq b'_{l_k}}}^{r_{d_k-j'-1}}\frac{1}{\left(b'_{l_k}-b_{l_{r_{d_k-j'}}}\right)}\right]$$
$$=\sum_{k=0}^{u}\sum_{j'=0}^{d_k}\binom{j+q}{j'}b'^{j+q-j'}_{l_k}D^{(j',q)}_{l_0,l_1,\ldots,l_q} \tag{18}$$

where

$$D^{(j',q)}_{l_0,l_1,\ldots,l_q} = (-1)^{d_k-j'}\prod_{\substack{r=0\\r\neq k}}^{u}\frac{1}{\left(b'_{l_k}-b'_{l_r}\right)^{(d_r+1)}}$$
$$\times\left[\underbrace{\sum_{\substack{r_1=0\\b_{l_{r_1}}\neq b'_{l_k}}}^{q}\frac{1}{\left(b'_{l_k}-b_{l_{r_1}}\right)}\sum_{\substack{r_2=0\\b_{l_{r_2}}\neq b'_{l_k}}}^{r_1}\frac{1}{\left(b'_{l_k}-b_{l_{r_2}}\right)}\cdots\sum_{\substack{r_{d_k-j'}=0\\b_{l_{r_{d_k-j'}}}\neq b'_{l_k}}}^{r_{d_k-j'-1}}\frac{1}{\left(b'_{l_k}-b_{l_{r_{d_k-j'}}}\right)}}_{d_k-j'}\right].$$

If $d_k = q$, which means that $b_{l_0} = b_{l_1} = \cdots = b_{l_q} = b'_{l_0}$, then $D^{(j',q)}_{l_0,l_1,\ldots,l_q} = \delta_{d_k,j'} = \delta_{q,j'}$.

This result can now be used in (3)

$$S_{n,m} = \lim_{p\to\infty}\sum_{j=0}^{p}\frac{\lambda^j}{j!}\left(b_n^j\delta_{n,m}+\sum_{q=1}^{j}\sum_{l_1=1}^{N}\sum_{l_2=1}^{N}\cdots\sum_{l_{q-1}=1}^{N}a_{n,l_1}\cdots a_{l_{q-1},m}\right.$$
$$\left.\times\sum_{k=0}^{u}\sum_{j'=0}^{d_k}\binom{j}{j'}b'^{j-j'}_{l_k}D^{(j',q)}_{l_0,l_1,\ldots,l_q}\right)$$
$$=\mathrm{e}^{\lambda b_n}\delta_{n,m}+\lim_{p\to\infty}\sum_{q=1}^{p}\sum_{l_1=1}^{N}\sum_{l_2=1}^{N}\cdots\sum_{l_{q-1}=1}^{N}a_{n,l_1}\cdots a_{l_{q-1},m}$$
$$\times\sum_{j=q}^{p}\frac{\lambda^j}{j!}\sum_{k=0}^{u}\sum_{j'=0}^{d_k}\binom{j}{j'}b'^{j-j'}_{l_k}D^{(j',q)}_{l_0,l_1,\ldots,l_q}$$
$$\overset{s=j-q}{=}\mathrm{e}^{\lambda b_n}\delta_{n,m}+\sum_{q\geqslant 1}\sum_{l_1=1}^{N}\sum_{l_2=1}^{N}\cdots\sum_{l_{q-1}=1}^{N}a_{n,l_1}\cdots a_{l_{q-1},m}$$
$$\times\sum_{k=0}^{u}\sum_{j'=0}^{d_k}\frac{\lambda^{j'}}{j'!}D^{(j',q)}_{l_0,l_1,\ldots,l_q}\lim_{p\to\infty}\sum_{s=0}^{p-q}\frac{\lambda^{s+q-j'}}{(s+q-j')!}b'^{s+q-j'}_{l_k}$$

$$= \mathrm{e}^{\lambda b_n}\delta_{n,m} + \sum_{q\geqslant 1}\sum_{l_1=1}^{N}\sum_{l_2=1}^{N}\cdots\sum_{l_{q-1}=1}^{N} a_{n,l_1}\cdots a_{l_{q-1},m}$$
$$\times \sum_{k=0}^{u}\sum_{j'=0}^{d_k}\frac{\lambda^{j'}}{j'!}D^{(j',q)}_{l_0,l_1,\ldots,l_q}\left[\mathrm{e}^{\lambda b'_{l_k}} - \sum_{r=0}^{q-j'-1}\frac{\left(\lambda b'_{l_k}\right)^r}{r!}\right]. \tag{19}$$

For a finite maximum value of q this expression has a finite number of terms. For the case that all $b_{l_k} = b_n$ are equal, because of $D^{(j',q)}_{l_0,l_1,\ldots,l_q} = \delta_{q,j'}$ we get

$$S_{n,m} = \mathrm{e}^{\lambda b_n}\left(\delta_{n,m} + \sum_{q\geqslant 1}\frac{\lambda^q}{q!}\sum_{l_1=1}^{N}\sum_{l_2=1}^{N}\cdots\sum_{l_{q-1}=1}^{N} a_{n,l_1}\cdots a_{l_{q-1},m}\right). \tag{20}$$

If $B_{n,m} = b\delta_{n,m}$, i.e. a diagonal matrix with the same entry for every element along the diagonal, the exponential term is independent of the combination of $l_1, \ldots, l_{q-1}$ and we get

$$S_{n,m} = \mathrm{e}^{\lambda b}\sum_{i=0}^{\infty}\frac{\lambda^i}{i!}(A^i)_{n,m} \quad \Rightarrow \quad S = \mathrm{e}^{\lambda B}\,\mathrm{e}^{\lambda A}.$$

This is simply the result for two commuting matrices A and B, where $\mathrm{e}^{\lambda(B+A)} = \mathrm{e}^{\lambda B}\,\mathrm{e}^{\lambda A}$, as one would expect, because A commutes with any diagonal matrix that has a constant value along the diagonal.

3. Example: approximation and inversion of LACBED patterns

The convergence of expression (19) strongly depends on the form of the matrices A and B and the value of the parameter λ and must therefore be examined for the particular application of interest.

The S-matrix notation is a fundamental mathematical tool in quantum scattering theory. For a review of inverse problems in quantum scattering the reader is referred to [6], where its theory, application and inverse problems are discussed in detail. In high energy electron scattering the condition that large elements of B are larger than those of A is usually satisfied, making this problem a practical application of this expansion [3–5].

Based on the Bloch wave solution to the relativistic multiple scattering Schrödinger equation of high energy scattering of electrons by the crystal potential [7, 8] the intensity of any point in an electron diffraction pattern defined by the reciprocal lattice vector $\vec{g}_n$ of the particular diffraction spot and the tangential component of the incident electron wavevector $\vec{k}_t$ is given by the modulus squared of the element of the scattering matrix $|S(\vec{k}_t)_{n,m}|^2$, where $\vec{g}_m = 0$

$$|S(\vec{k}_t)_{n,m}|^2 = [\exp\{\mathrm{i}T(A + B(\vec{k}_t))\}]_{n,m}\cdot[\exp\{-\mathrm{i}T(A^* + B(\vec{k}_t))\}]_{n,m} \tag{21}$$

where A is a square matrix with the reciprocal space potential energy terms $A_{n,m} = U_{\vec{g}_n-\vec{g}_m}$ as its off-diagonal elements and zeros along its diagonal. Here $U_{\vec{g}}$ are scaled Fourier coefficients of the crystal potential (structure factors). B is a diagonal matrix with terms $B_{n,n} = -(|\vec{g}_n|^2 + \vec{g}_n\cdot\vec{k}_t)$ related to the kinetic energy part of the Schrödinger equation along its diagonal, and $T = \pi\lambda t$, where λ is the electron wavelength, and t the thickness of the sample. The 17-beam A matrix of GaAs(011) for the reciprocal lattice vectors $\vec{g}_{[\bar{1}\bar{3}\bar{3}]}$, $\vec{g}_{[1\bar{3}\bar{3}]}$, $\vec{g}_{[\bar{4}00]}$, $\vec{g}_{[\bar{3}\bar{1}1]}$, $\vec{g}_{[3\bar{1}1]}$, $\vec{g}_{[0\bar{2}\bar{2}]}$, $\vec{g}_{[\bar{1}\bar{1}1]}$, $\vec{g}_{[1\bar{1}1]}$, $\vec{g}_{[000]}$, $\vec{g}_{[\bar{1}1\bar{1}]}$, $\vec{g}_{[11\bar{1}]}$, $\vec{g}_{[02\bar{2}]}$, $\vec{g}_{[\bar{3}1\bar{1}]}$, $\vec{g}_{[31\bar{1}]}$, $\vec{g}_{[400]}$, $\vec{g}_{[\bar{1}3\bar{3}]}$ and $\vec{g}_{[13\bar{3}]}$ ($|\vec{g}_{[\bar{1}\bar{3}\bar{3}]}| = 0.7709\,\text{Å}^{-1}$, $|\vec{g}_{[\bar{4}00]}| = 0.7075\,\text{Å}^{-1}$,

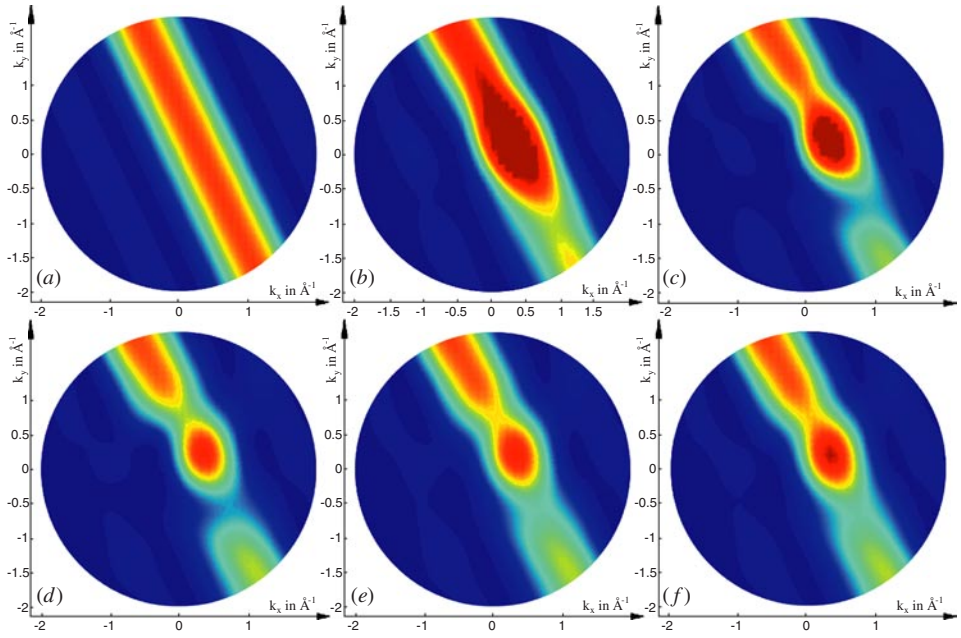

Figure 1. Successive approximation of the intensity in a GaAs(110) LACBED disc at $\vec{g} = [\bar{3}\bar{1}1]$ for an electron energy of 200 keV ($\lambda = 0.0251$ Å) and a thickness of 70 Å. The intensity has been calculated using expression (22) including terms up to orders (*a*) $q = 2$, (*b*) $q = 3$, (*c*) $q = 4$, (*d*) $q = 5$, and (*e*) $q = 6$ in the approximation of $|S(\vec{k}_t)_{n,m}|^2$. (*f*) shows the correct result including all orders for comparison. The colour scale is shown in figure 2.

$|\vec{g}_{[\bar{3}\bar{1}1]}| = 0.5866\,\text{Å}^{-1}$, $|\vec{g}_{[0\bar{2}2]}| = 0.5003\,\text{Å}^{-1}$, $|\vec{g}_{[1\bar{1}1]}| = 0.3063\,\text{Å}^{-1}$ $|\vec{g}_{[000]}| = 0\,\text{Å}^{-1}$) has 38 distinct structure factors (if absorption is included in the calculation)

$$A_{17} = \begin{pmatrix} 0 & U_1 & U_2 & U_3 & U_4 & \cdots & U_{10} & U_{11} & U_{12} & U_{13} & U_{14} \\ U_{15} & 0 & U_{16} & U_{17} & U_{18} & \cdots & U_{24} & U_{25} & U_{26} & U_{27} & U_{13} \\ U_{18} & U_4 & 0 & U_{28} & U_6 & \cdots & U_{32} & U_5 & U_{33} & U_{26} & U_{12} \\ \vdots & \vdots & \vdots & \vdots & \vdots & \ddots & \vdots & \vdots & \vdots & \vdots & \vdots \\ U_{24} & U_{10} & U_{38} & U_{23} & U_{32} & \cdots & U_{21} & U_{34} & 0 & U_{16} & U_2 \\ U_{13} & U_{14} & U_{10} & U_{11} & U_{12} & \cdots & U_2 & U_3 & U_4 & 0 & U_1 \\ U_{27} & U_{13} & U_{24} & U_{25} & U_{26} & \cdots & U_{16} & U_{17} & U_{18} & U_{15} & 0 \end{pmatrix}.$$

In order to give an idea of the size of the elements in A, we will show some of their moduli here:

$$\begin{pmatrix} 0 & 0.0012 & 0.0238 & \cdots & 0.0177 & 0.0168 & 0.0012 \\ 0.0012 & 0 & 0.0177 & \cdots & 0.0238 & 0.0012 & 0.0168 \\ 0.0238 & 0.0177 & 0 & \cdots & 0.0186 & 0.0238 & 0.0177 \\ \vdots & \vdots & \vdots & \ddots & \vdots & \vdots & \vdots \\ 0.0177 & 0.0238 & 0.0186 & \cdots & 0 & 0.0177 & 0.0238 \\ 0.0168 & 0.0012 & 0.0238 & \cdots & 0.0177 & 0 & 0.0012 \\ 0.0012 & 0.0168 & 0.0177 & \cdots & 0.0238 & 0.0012 & 0 \end{pmatrix} \text{Å}^{-2}.$$

This also exhibits the symmetries in A for the case of a non-centrosymmetric crystal without absorption in which case A is Hermitian and also has symmetry across the anti-diagonal,

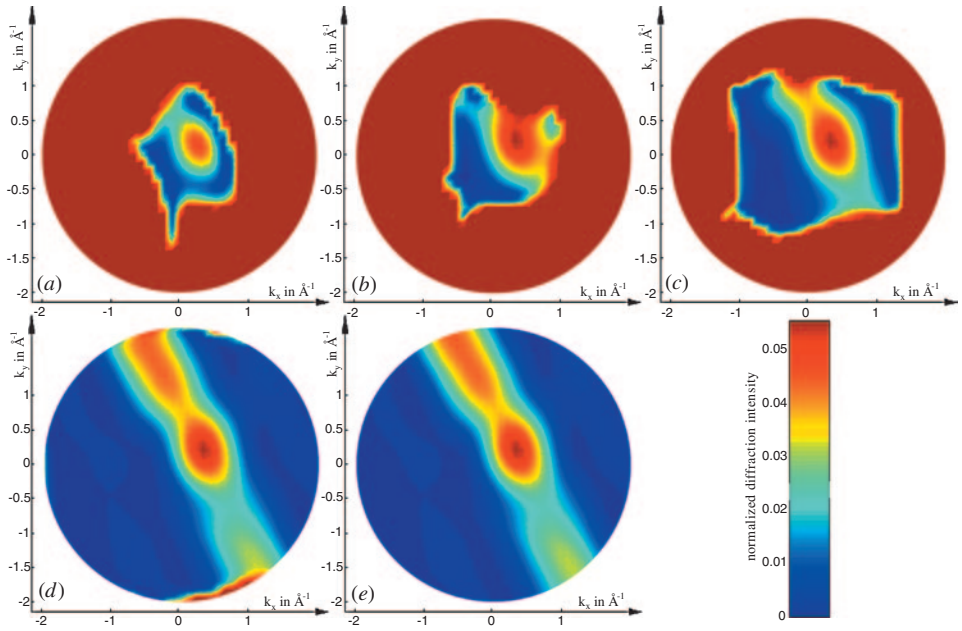

Figure 2. Successive approximation of the intensity in a GaAs(110) LACBED disc at $\vec{g} = [\bar{3}\bar{1}1]$ for an electron energy of 200 keV ($\lambda = 0.0251$ Å) and a thickness of 70 Å using the Taylor expansion of the matrix exponential. The complex amplitude has been calculated using expression (2) including terms up to order (*a*) $p = 5$, (*b*) $p = 10$, (*c*) $p = 20$, (*d*) $p = 40$ and (*e*) $p = 80$, whose modulus has then been squared to give the intensity. The colour scheme used in this figure, as well as figure 1, is shown on the bottom right. For large values of $|\vec{k}_t|$ the approximated results exceeded the colour scale (up to values of 10^{12}) in plots (*a*) through (*d*) and had to be plotted in the colour representing the upper cut-off value.

a property of equation (1). The largest structure factor modulus is $|U_5| = 0.0642$ Å^{-2}. There is no restriction on the phases of the $U_{\vec{g}}$. At 200 keV the electron wavelength is 0.0251 Å, so that for a thickness of $t = 70$ Å $T = \pi\lambda t \approx 5.515$ Å^2.

Since in this example we can only measure the diffraction intensity directly, i.e. $|S(\vec{k}_t)_{n,m}|^2$ for a certain n, the modulus squared of (19) has to be taken as

$$\begin{aligned}\left|S_{n,m}\right|^2 = \delta_{n,m} + \sum_{q=2}^{\infty} \sum_{l_1,\ldots,l_{q-2}=1}^{N} \sum_{p=1}^{q-1} & U_{\vec{g}_n-\vec{g}_{l_1}} \ldots U_{\vec{g}_{l_{p-1}}-\vec{g}_m} U^*_{\vec{g}_n-\vec{g}_{l_p}} \cdots \\ & \times U^*_{\vec{g}_{l_{q-2}}-\vec{g}_m} C^p_{n,l,\ldots,l_{p-1},m} \left(C^{q-p}_{n,l_p,\ldots,l_{q-2},m}\right)^* \\ & + 2\,\mathrm{Re}\left(\sum_{q=1}^{\infty} \sum_{l,l_2,\ldots,l_{q-1}=1}^{N} U_{\vec{g}_n-\vec{g}_l} U_{\vec{g}_l-\vec{g}_{l_2}} \cdots U_{\vec{g}_{l_{q-1}}-\vec{g}_m} C^q_{n,l,\ldots,l_{q-1},m}\right) \delta_{n,m}. \end{aligned} \tag{22}$$

For diffraction discs other than the central beam $n \neq m$, so that the terms including $\delta_{n,m}$ drop out.

Equation (22) achieves the separation of the multiple scattering series into a set of linear equations involving a product of terms consisting only of structure factors $U_{\vec{g}}$, and terms in $C^q_{n,l_1,\ldots}$ involving only known structural constants. These linear equations may therefore be inverted.

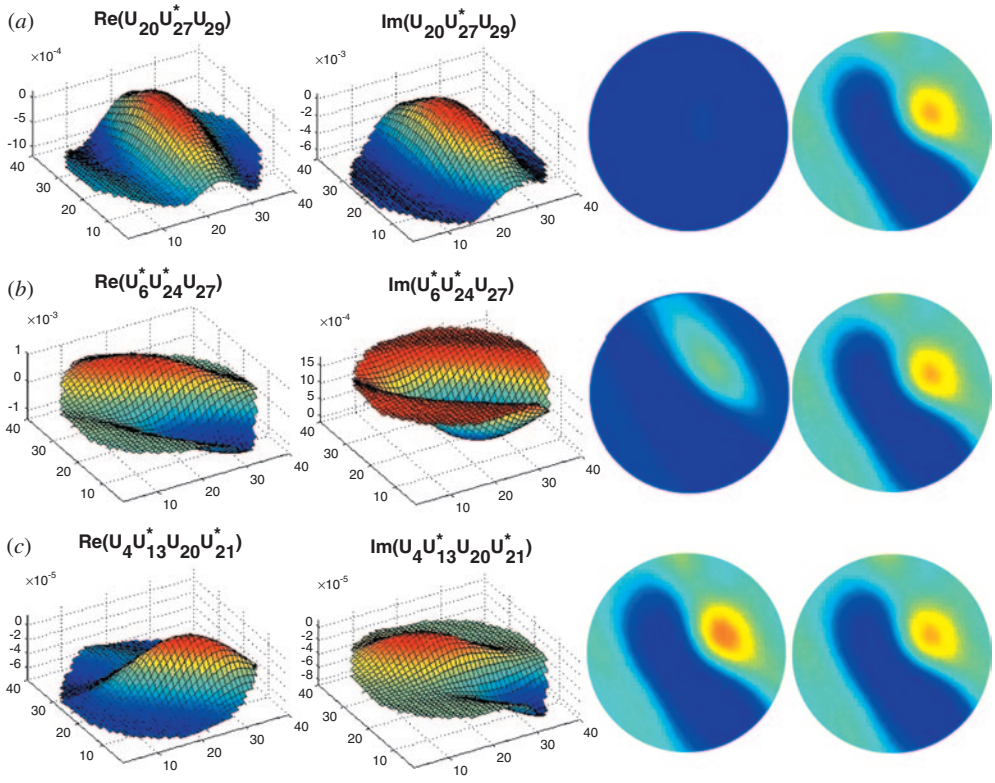

Figure 3. Approximation of LACBED data. The difference of simulated data for reflection $[\bar{3}\bar{1}1]$ and the sum of 'phase-less' terms (see text) is approximated by an increasing number of terms: (*a*) one term, (*b*) five terms and (*c*) 80 terms. The real and imaginary part of the first, fifth and eightieth term along with the expression for the $U_{\vec{g}}$ polynomials are shown in the two plots on the left of each figure. The third plot shows the accumulated intensity of all the terms up to the current one (i.e. (*a*) the first term only, (*b*) the first five terms and (*c*) the first 80 terms). The exact result is plotted on the far right of each row for comparison using the same colour scheme. The intensity of every expansion term, as well as the exact data has been offset by its value at $\vec{k}_t = 0$. The convergence angle is 50 mrad, thickness 40 Å, and the accelerating voltage 200 kV. The 17×17 A matrix contains 38 unique structure factors.

Figure 1 demonstrates the convergence of expression (22) for the case of the 17-beam GaAs(110) matrix and $n = 4$, i.e. $\vec{g} = [\bar{3}\bar{1}1]$. Every data point within the large-angle convergent beam electron diffraction (LACBED) discs (convergence angle of 50 mrad) is the modulus squared of a single element in $S(\vec{k}_t)_{n,m}$ (in this case $|S(\vec{k}_t)_{4,9}|^2$) as function of the x- and y-components of the incident electron beam wavevector $\vec{k}$. The first nonzero terms for $n \neq m$ are those with $q = 2$, involving products of elements of A of length 2, i.e. $\sum_{n_1,n_2} U_{n_1} U^*_{n_2} f(\vec{k}_t)$, where $f(\vec{k}_t)$ is the well-defined product of two C-coefficients. Figure 1(*b*) includes these $q = 2$ terms as well as those with $q = 3$ (containing products of elements of A of length 3). At $q = 6$ (figure 1(*e*)) the approximation shows already very good agreement with the exact result (figure 1(*f*)), which has been calculated using matrix diagonalization.

In contrast, figure 2 illustrates the use of the Taylor expansion of the matrix exponential for the same test case. Expression (2) has been used to approximate the matrix exponential for each point in the two-dimensional $\vec{k}_t$ vector space. The modulus squared of element $S(\vec{k}_t)_{4,9}$

has then been plotted. Note, that by squaring a polynomial of order p, we also include terms of order $2p$, i.e. also products of structure factors of length $2p$, whereas the value of q in expression (22) defines the true degree of polynomials of elements of A used in the expansion. Plots (a) through (d) in figure 2 show that the Taylor expansion converges first for small values of $|\vec{k}_t|$, which makes sense, because the elements of B ($B_{n,n} = -[|\vec{g}_n|^2 + \vec{g}_n \cdot \vec{k}]$) are small, if $|\vec{k}_t|$ is small. Since polynomials of elements of B associated with a certain polynomial of A-elements are included to all orders in expressions (19) and (22), this 'selective convergence behaviour' cannot be observed there.

We now consider electron diffraction experiments in which data from very thin crystals have been used to measure the moduli of the structure factors, and it remains to determine their phases, that is, to solve the phase problem. This can be done up to an arbitrary origin-dependent phase factor as follows. Since certain products of $U_{\vec{g}}$ (e.g. $U_1 U_1^*$, or $U_2 U_3 U_2^* U_3^*$) can be determined without knowledge of the phase of any of their members, because their phases cancel, we can calculate the contribution of all those terms and then subtract it from the experimental data. Figure 3 shows the approximation of this difference between simulated LACBED data and the sum of all the 'phase-less' terms in the expansion. Terms up to $q = 4$ have been included. Other than the thickness the computational parameters are the same as for figures 1 and 2, the thickness is $t = 40$ Å. The first two plots in each row show the function $f(\vec{k}_t)$ (product of two C-coefficients, as shown in expression (22)) and the corresponding $U_{\vec{g}}$-polynomial, split into real and imaginary parts. The third plot shows the sum of all those terms for a total of 1(2), 5(10) and 80(160) terms (number in parentheses, if real and imaginary parts are counted separately). Since for a large enough range in $\vec{k}_t$ vector space all the $f(\vec{k}_t)$ are linearly independent, the polynomials $\mathrm{Re/Im}\left(U_{n_1} \ldots U_{n_q}^*\right)$ can be determined individually by solving the set of linear equations, and therefore also their phases.

4. Conclusion

A new expansion for the general matrix exponential of two non-commuting matrices, one of which is diagonal, has been derived, which works up to any order. Many problems involving the solution of the time-dependent or time-independent Schrödinger equation can be expressed in the form of such matrix exponentials by writing the potential and kinetic energy terms in separate matrices. The convergence behaviour of this expansion has been illustrated by applying it to the approximation of multiple scattering of high energy electrons in a crystal. It has been shown how this expansion can be applied for solving the phase problem in multiple scattering experiments by expanding the set of nonlinear equations with a finite system of linear equations which can be solved by matrix inversion or other standard linear equation solvers. In particular, the inversion from two-dimensional rocking curves, which can be recorded as (LA)CBED patterns with a single exposure in most electron microscopes to the projected crystal potential has been analysed.

Acknowledgment

This work has been supported by NSF award DMR9814055.

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