Dynamic scattering theory for dark-field electron holography of 3D strain fields

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\textbf{Article info}

\textbf{Abstract}

Dark-field electron holography maps strain in crystal lattices into reconstructed phases over large fields of view. Here we investigate the details of the lattice strain–reconstructed phase relationship by applying dynamic scattering theory both analytically and numerically. We develop efficient analytic linear projection rules for 3D strain fields, facilitating a straightforward calculation of reconstructed phases from 3D strain rates. They are used in the following to quantify the influence of various experimental parameters like strain magnitude, specimen thickness, excitation error and surface relaxation.

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\section{1. Introduction}

Dark-field electron holography (DFEH) is a recently developed technique for measuring strain in nanostructures, in particular over wide fields of view \cite{1,2}. It has been applied to the study of strained-silicon transistors \cite{3-5} and epitaxial thin films \cite{6,7}. Different aspects of the technique itself have been investigated over this period. Precision has been studied as a function of experimental parameters such as exposure time, biprism voltage and sample thickness \cite{8,9}. The methodology has been extended to correct for thickness variations by taking conjugate bright-field electron holograms \cite{2}. The range of imaging conditions, notably magnification and spatial resolution, has been enlarged by adjusting lens configurations \cite{10,11}. However, one particular basic assumption remains unchallenged.

The current assumption when using DFEH is that either (a) the strain is uniform over the thickness of the foil, or that (b) the measured strain corresponds to the average strain over the thickness of the foil. Whilst the former poses no problems within the other assumptions of the method such as the column approximation, such specimens do not exist in practice. Any specimen that had this characteristic in the bulk (indeed, the vast majority of currently studied examples) will have lost it in the process of sample preparation. The two new free surfaces introduced by the thinning process will have relaxed some of the strain through the well-known thin-film effect \cite{12}. More importantly, the strain will now vary over the viewing direction, which we will define throughout as the z-axis. Furthermore, there is a tendency to look at specimens which have z-dependent strain, even in the “bulk.” Two cases in hand are quantum dot structures \cite{8} and modern 3D microelectronic devices such as FinFets \cite{13}. It is therefore vital to know what the measured strain corresponds to exactly.

The problem of z-dependent strain is not new and is inherent to any electron microscopy technique designed to measure strain. The difficulty is always how to evaluate, compensate and correct for it in the analysis. Convergent-beam electron diffraction (CBED), the first technique used to study strained-silicon devices \cite{14}, breaks down in the presence of significant column bending due to thin-film relaxation \cite{15}. The only solution is to model the relaxation with an assumed strain field, perform simulations and compare with the experimental data \cite{16}. To avoid brute-force atomistic multislice calculations \cite{17}, a Feynman diagram technique applied to dynamic theory was developed \cite{18}. In this approach, the strain is introduced as a perturbation to the full Bloch-wave calculation within the column approximation, and integrated numerically slice by slice through the specimen thickness. A more analytical theory does not currently exist.
The evaluation of $z$-dependent strain is perhaps even more difficult for zone-axis techniques such as high-resolution electron microscopy (HRTEM) [3] or nano-beam electron diffraction (NBED) [19]. On one hand, specimens tend to be thinner than for CBED, thus reducing dynamic effects, but on the other hand, the number of beams involved is prodigious. Beyond the woefully inadequate weak-phase object approximation, the only alternative is atomistic multislice simulations, coupled with image formation in the case of HRTEM [20]. Surprisingly, high-angle annular dark-field imaging (HAADF) has seen the most progress towards an analytical approach [21], following on the earlier analysis in terms of strain-induced inter-band scattering [22].

Indeed, we have to return to simpler scattering conditions, such as those prevalent in a DFEH experiment, to find an analytical theory which can incorporate a $z$-dependent strain field, exemplified by 2-beam dynamical theory [23,24]. Within this theory, analytical solutions were found for some special cases, such as Moiré contrast and stacking fault contrast. These represent a single step in lattice parameter (or strain) or displacement, respectively, within the foil thickness. Other cases have been implemented by a slice by slice approach with transmission matrices (see, e.g., the description in [25]). In the following we will show that the theory can be extended to include a varying $z$-dependent strain field in a more analytical way.

The organization of the paper follows closely the different levels of approximations used to incorporate strained lattices into scattering theory. After a short introduction to the optical setup of DFEH (Section 2) and high-energy electron scattering (Section 3.1), we discuss the notion of the geometric phase (Section 3.2) as an approximate way to describe weakly deformed lattices. The next step consists of contracting many-beam theory to the experimentally used approximate way to describe weakly deformed lattices. The next step proceeds towards an analytical approach with transmission matrices (see, e.g., the description in [25]). Indeed, we have to return to simpler scattering conditions, such as those prevalent in a DFEH experiment, to find an analytical theory which can incorporate a $z$-dependent strain field, exemplified by 2-beam dynamical theory [23,24]. Within this theory, analytical solutions were found for some special cases, such as Moiré contrast and stacking fault contrast. These represent a single step in lattice parameter (or strain) or displacement, respectively, within the foil thickness. Other cases have been implemented by a slice by slice approach with transmission matrices (see, e.g., the description in [25]). In the following we will show that the theory can be extended to include a varying $z$-dependent strain field in a more analytical way.

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2. Optical setup

To generate a dark field electron interference pattern, a strongly excited diffracted beam is generated by deliberately tilting the specimen into 2-beam conditions. Subsequently, the transmitted beam is blocked by an aperture and diffracted beams originating from an undisturbed and strained specimen region are superimposed with the help of a Möllenstedt biprism to form a hologram in the image plane. This optical setup is illustrated in Fig. 2. The slightly changing diffraction angle within the strained region translates into a

![Fig. 2. DFEH setup and coordinate system used in the text. Note that general vectors are denoted by small bold letters, e.g. $\mathbf{r}$, and vectors in $(x,y,z=\text{const})$-planes will be denoted by capital bold letters, e.g. $\mathbf{R}$.](image)

![Fig. 1. Strain field $e_{xx}(r)$ generated by H⁺-implantation in a Si matrix as calculated by finite element method as well as an infinitely thick specimen and will be denoted by $\varepsilon_{\text{bulk}}^{\text{inh}}$ in the text.](image)
They constitute a set of coupled first order differential equations, which can be integrated by various methods starting with the unperturbed electron wave at the entrance face of the crystal. A straightforward method consisting of a numerical integration with a predefined stepsize, the well-known Multislice algorithm [29], will be used in the following to provide numerical reference for less accurate analytical approximations describing the influence of strained lattices on electron scattering.

3.2. Geometric phase

One important approximation to describe the influence of strained crystal lattices on electron scattering is the so-called geometric phase. That approximation is based upon the presumption that an otherwise perfectly periodic crystal structure is modified by a displacement field, which changes slowly on the length scale of the lattice constant, i.e. $\delta u_i/\delta a=1$: then one can divide the total scattering volume into $n$ subvolumes centered around $r_n$ and much larger than the volume of one unit cell, i.e. $\Omega_0+\Omega_{uc}$, where the displacement $u_i$ is approximately constant. According to the shift property of the Fourier-transformation the scattering potential reads

$$V_n(r)=\sum_{\bar{R}}V_{uc}(g)e^{i2\pi x_r-\bar{r}_n-u_n).}$$

(5)

By noting that electron scattering within the high-energy regime also possesses a limited correlation length (i.e. the lateral distance over which an atomic potential influences the wave at the exit face), we can apply the so-called column approximation [30] to describe scattering within a cylindrical subvolume centered around $R_n$

$$\frac{\partial \phi(G, z; R_n)}{\partial z} = -i2\pi \frac{G^2 + 2K_0 \cdot \mathbf{G}}{2k_0} \phi(G, z; R_n) + iC_1 \mathbf{V} \mathbf{G} e^{-i2\pi u_z R_n} \phi(G, z; R_n).$$

(6)

Note that the only difference to HW consists of an additional multiplication of the Fourier components of the potential with the geometric phase term $\exp(-i2\pi G \mathbf{u} z/r_n)$. Exactly the same integration schemes applicable to HW remain valid due to the same mathematical structure. The complete exit wave of a strained lattice is obtained by patching together all column solutions, which is also computationally much less demanding than calculating the exit wave of the complete lattice, i.e. all columns, at once.

To illustrate the limitations of the geometric phase approximation, determined mainly by $\delta u_i/\delta a=1$, we compare the reconstructed strain obtained from DFEH imaging many-beam MS simulation, performed on 3 increasingly strong $e_{bulk}^{xx}$-uniaxially strained lattices (Fig. 3, for details of the simulation see Appendix C). As expected the geometric phase approximation becomes less valid with increasing strain. Since lattice strain usually stays well below 10%, however, the deviations can be neglected with respect to other factors discussed further below.

3.3. 2-Beam equation

It was mentioned above that in DFEH the crystal is oriented such that the intensity of the diffracted beam, carrying the information about the strained lattice, is maximized. This corresponds to a 2-beam condition, where the original beam and one diffracted beam are significantly stronger than all other beams. Note that the validity of the 2-beam approximation depends on various factors such as thickness, scattering potential, acceleration

$$\frac{d^2 \phi_c}{dz^2} = -i2\pi \left( \frac{(\mathbf{G} \cdot \mathbf{u}(z))}{\partial z} - S_c \right) \frac{d\phi_c}{dz} - \left( C_1 V \mathbf{G} V_{-c} + 4\pi^2 \frac{(\mathbf{G} \cdot \mathbf{u}(z))}{\partial z} S_c \right) \phi_c.$$

(10)

The reconstructed strain obtained from the [004]-beam simulated by means of many-beam MS on 3 increasingly strong $e_{bulk}^{xx}$-uniaxially strained lattices and the respective input lattice strain $e_{input}$

![Fig. 3. Comparison of reconstructed strain obtained from the [004]-beam simulated by means of many-beam MS on 3 increasingly strong $e_{bulk}^{xx}$-uniaxially strained lattices and the respective input lattice strain $e_{input}$](image)

3.4. Mathematical details

One major detail of the geometric phase approximation is the use of the 2-beam approximation:

$$\mathbf{V} = \mathbf{V}_0 + \mathbf{V}_1 + \mathbf{V}_2 + \mathbf{V}_3,$$

where $\mathbf{V}_0$ is the mean potential, $\mathbf{V}_1$ the first order correction, $\mathbf{V}_2$ the second order correction, and $\mathbf{V}_3$ the third order correction. The second order correction is given by

$$\mathbf{V}_2 = \frac{1}{2} \mathbf{V}_0 \cdot \mathbf{G} \mathbf{V}_0,$$

and the third order correction is given by

$$\mathbf{V}_3 = \frac{1}{6} \mathbf{V}_0 \cdot \mathbf{G} \mathbf{V}_0 \cdot \mathbf{G} \mathbf{V}_0.$$
introduces but a constant phase shift, which corresponds to a free choice of the reference area in DFEH.

3.4. Special (analytic) cases

In order to discuss some general aspects of the scattering, we deliberately choose a constant displacement (i.e. \( \mathbf{u}(z) = 0 \)) and analyze the well-known solutions of the damped harmonic oscillator (Eq. (10)). The two elementary solutions \( \phi^{(0)}(z) = \exp(2\pi i k_{1,2} z) \) read

\[
k_{1,2} = \frac{s_G}{2} \pm \kappa_G
\]

(11)

\[
\kappa_G = \pm \frac{1}{2\xi G} \sqrt{\pi^2 \kappa^2 + C^2_{\xi G} V_{\xi G} V_{C\xi G}}
\]

(12)

Note that the parameter \( \kappa_G \) is almost purely real due to the generally small absorptive part of the potential. By taking into account the boundary conditions

\[
\phi^{(0)}(0) = 0 \quad \text{and} \quad \frac{1}{iC_G V_{\xi G}} \frac{\partial \phi^{(0)}}{\partial z}(0) = \phi^{(0)}(0) = 1
\]

(13)

at the entrance face, the diffracted beam reads

\[
\phi^{(0)}(z) = \frac{C_G V_{\xi G}}{2\pi i (k_1 - k_2)} (\exp(2\pi i k_1 z) - \exp(2\pi i k_2 z))
\]

(14)

Similarly, with

\[
\phi_0^{(0)}(0) = 1, \quad \frac{1}{iC_G V_{\xi G}} \frac{\partial \phi_0^{(0)}}{\partial z}(0) = \phi_0^{(0)}(0) = 1
\]

(15)

the transmitted beam reads

\[
\phi_0^{(0)}(z) = \frac{1}{k_1 - k_2} (k_1 \exp(2\pi i k_2 z) - k_2 \exp(2\pi i k_1 z))
\]

(16)

Here, we observe the well-known Pendellösung with the effective extinction length

\[
\xi_G = \Re \left\{ \frac{\pi}{\sqrt{\pi^2 \kappa^2 + C^2_{\xi G} V_{\xi G} V_{C\xi G}}} \right\}
\]

(17)

defined as the propagation distance between two zeros in the diffracted beam.\(^2\) Note that the extinction length is increased by non-zero excitation errors \( s_G \). In the experiment it is therefore possible to influence the amplitude of the diffracted beam by deliberately changing \( s_G \). We furthermore observe a damping of the electron wave if absorption \( V_{\xi G} = 0 \) introduces imaginary components in \( k_{1,2} \). The phase of the diffracted beam increases linearly with \( z \) and the increase is proportional to the mean inner potential \( \bar{V}_0 \) and the excitation error \( s_G \). These phase terms have to be subtracted, when analyzing the reconstructed phase in terms of the geometric phase, which is most elegantly achieved experimentally by choosing a reference region with the same thickness and orientation like the strained region.

3.5. von-Neumann expansion

As it was mentioned above, in case of a general \( z \)-dependent displacement field no closed analytic form can be derived. However, one can exploit that usually \( \mathbf{u}(z) / \partial z \approx \mathbf{1} \), for applying a perturbation expansion. We use the well-known approximation scheme based on the von-Neumann series expansion of the differential equation (10) with respect to the smallness parameter \( \mathbf{u}(z) / \partial z \approx \mathbf{1} \). When stopping the expansion after the first order term, i.e. \( \phi_G = \phi^{(0)}_G + \phi^{(1)}_G + \mathcal{O}(\mathbf{u}(z) / \partial z)^2 \), we obtain (see Appendix B) for the diffracted beam (\( s_G = 0 \) for the moment)

\[
\phi_G = \frac{\pi C_G V_{\xi G}}{\kappa_G} \left( \frac{1}{4\pi^2} \left( e^{2\pi i c z} - e^{-2\pi i c z} \right) + \kappa_G \int_0^1 \left( e^{2\pi i c (t - 2z)} - e^{-2\pi i c (t - 2z)} \right) G(z) \cdot u(z) \, dz \right)
\]

(18)

If we now keep in mind that absorption is a second order effect yielding a comparatively small imaginary component of the diffracted wave vector \( \mathcal{N}(s_G) \approx \mathcal{N}(s_G) \) and therefore

\[
\sin(2s_G c \xi_G z) = e^{2\pi i c z} - e^{-2\pi i c z}
\]

and use again that the first order term is small compared to the zero order one, i.e.

\[
\phi_G = \phi^{(0)}_G + \phi^{(1)}_G + \phi^{(0)}_G \exp(\phi^{(1)}_G / \phi^{(0)}_G) = \phi^{(0)}_G \exp(i\phi_G).
\]

(20)

Eq. (18) could be further simplified to yield an expression for the reconstructed phase in DFEH

\[
\phi_G = -2\pi \int_0^1 f_G(z) G(z) \cdot u(z) \, dz
\]

(21)

with

\[
f_G(z, t) = \Re \left\{ \frac{2\pi C_G \cos(2\pi c z)}{\sin(2\pi c z t)} \right\}
\]

(22)

defining the weighting function \( f_G \) for the displacement projection. This integral expression constitutes the main result of our analytic analysis. We first note that in the leading order the strained lattice produces an effect in the phase and not the amplitude of the diffracted beam, which explains why phases reconstructed with DFEH can be used for strain measurements. The mathematical structure, i.e. a line integral of \( u \) with a weighting kernel, facilitates a straightforward interpretation of typical strain profiles observed experimentally without performing large scale scattering simulations. By comparison to “brute force” MS simulations, we will show below that the error of this first order approximation is small if strain fields are small. The weighted integral is a linear projection rule for the originally 3D geometric phase (or distortion) field (see Section 3.2) and therefore applies equally to all values linearly depending on the reconstructed phase, in particular the strain tensor.

We want to point out that one can alternatively derive the weighting function in a less formal way based on a \( z \)-dependent Master equation for the 2-beam case. Accordingly, one writes down the intensity of the diffracted beam in the exit plane as a sum of beams created at a certain depth in the crystal phase-shifted with the corresponding geometric phase. Both the original transmitted (Eq. (16)) and diffracted beams (Eq. (14)) contribute locally to the diffracted beam. The respective weight in the final result is determined by the value of the transmitted (diffracted) beam at \( z \) multiplied with the value in the exit plane \( t \) of the diffracted (transmitted) beam created at \( z \), i.e.

\[
f_{G,1} \sim \cos(-2\pi c z) \cos(-2\pi c_G(t - z))
\]

(23)

and

\[
f_{G,2} \sim \sin(-2\pi c z) \sin(-2\pi c_G(t - z)).
\]

(24)

Using

\[
\cos(-2\pi c_G(t - 2z)) = \cos(-2\pi c_G(t - z)) \cos(-2\pi c_G z) + \sin(-2\pi c_G(t - z)) \sin(-2\pi c_G z)
\]

(25)

and normalizing correctly, the sum of the two contributions yields the weighting function (21). Similar to the more stringent perturbation approach it was assumed that the original transmitted and
The corresponding result for the transmitted beam reads

\[
\phi_0 = -2\pi \int_0^t f_G^0(z) G \cdot u(z) \, dz
\]

(26)

with the weighting function

\[
f_G^0(z, t) = -\Re \left\{ \frac{2\kappa c \sin (2\kappa c (t-2z)) \cos (2\kappa c t)}{\cos (2\kappa c t)} \right\}.
\]

(27)

We want to highlight a connection between Fourier transformation and the weighting integrals (21) and (26). \(\kappa c\) is almost purely real, hence \(\phi_0\) is approximately proportional to a windowed cosine transform and \(\phi_0\) to a windowed sine transform within a \(z\)-interval between 0 and \(t\). It could therefore be possible to combine dark and bright field holography under 2-beam conditions to obtain information on both symmetric and antisymmetric parts of the displacement.

We will now discuss the general shape of the weighting function \(f_G^0\) of the diffracted beam and its influence on the measured phase in more detail. \(f_G^0\) depends on two variables, the thickness and the integration variable \(z\) (see Fig. 4). It is readily observed, that \(\int_0^t f_G^0(z) \, dz = 1\), i.e. in case of a constant displacement field in \(z\)-direction \((u(z) = u)\) the reconstructed geometric phase corresponds exactly to the 2D displacement field of the crystal lattice. i.e. \(\phi_G = -2\pi G \cdot u\). Thus, reconstructed strains from dark field EH are most easily obtained if strain fields are constant along \(z\), i.e. \(u(z) = u\).

To that end one has to suppress surface relaxations which is only partly possible by dedicated specimen preparation techniques. The evaluation of strains in the general 3D case is much more involved: the weighting kernel \(f_G^0(z, t)\) is \(G\)-dependent, hence different diffracted waves \(\hat{\phi}_G\) measure differently projected parts of the strain. For instance a \{004\}- and a \{008\}-beam in our test object are subject to very different weighting functions \((\kappa c\) is changing) resulting eventually in different reconstructed strains even though the diffraction direction is the same. Also general strain tensor components reconstructed from linearly independent diffraction directions belonging to different lattice plane families, e.g. \{004\} and \{220\}, have to be interpreted with caution since their weighting kernel is different. In Si that problem can be avoided if using \{111\}-diffracted beams for reconstructing the strain tensor.

It is now useful to rescale the \(z\)-coordinate in terms of the extinction length, i.e. \(z = z/\xi_G\) and \(t = t/\xi_G\), which facilitates a material and \(G\)-independent discussion of the weighting function. It is observed that \(f_G^0\) depends on \(t/\xi_G\) in the reconstructed phase. Experimentally, one exploits this property by using the bright field phase for measuring thickness profiles [2]. (ii) Reconstructed strains can have inverted signs if they are dominated by contributions stemming from depths, which are weighted by the negative part of the weighting kernel. For instance, the ubiquitous surface relaxation in thin TEM specimen is weighted negatively if \(t < \xi_G\) (see Fig. 4). More detailed investigations on this particular effect will be presented elsewhere. (iii) The reconstructed phase is “blind” with respect to abrupt displacement field changes, such as occurring at abrupt interfaces, if they occur at particular depths \(z_e = (n + 1/2)\xi_G\). In that case the changed displacement field is weighted by an antisymmetric function (see e.g. \(f_G^0(0.5\xi_G < z < \xi_G; t = 1.25\xi_G\) and \(1.5\xi_G\)) in Fig. 4), hence its integral vanishes. To illustrate this behavior and verify the accuracy of the weighting function formalism as promised above, we calculated the \{004\}-diffracted beam within a multiple-beam MS simulation at a sample containing a sharp change of \(\text{e}^{\text{bulk}}\)-uniaxial strain at different depths (see Appendix C for details of the simulation). Fig. 5 shows that the change at exactly \(0.5\xi_G\) is not visible in the diffracted wave which is in line with the analytic prediction of the perturbative 2-beam theory.\(^3\) Notably, such a change in lattice strain becomes visible in the phase of the transmitted beam, which can be understood by the following intuitive argument (or by directly evaluating Eq. (26)): starting with a maximal (minimal) diffracted (transmitted) beam at \(0.5\xi_G\) exchanges the usual role of transmitted and diffracted beams at the entrance face. Consequently the phase information about a strain starting at \(0.5\xi_G\) is now attached to the transmitted beam.

We finally note the weighting function for the reconstructed phase of the diffracted beam with non-vanishing

\(^3\) Small deviations close to \(\xi_G\) are caused by the influence of multiple beams and the limited sampling in the MS simulation.

Fig. 4. Weighting function \(f_G^0(z, t)\) (color) with 4 1D-cuts at special thicknesses \(t = 0.5, 0.75, 1.25, 1.5\xi_G\). Note that \(f_G^0(0 < z < 0.5\xi_G; 0.5\xi_G) = f_G^0(0 < z < 0.5\xi_G; 1.5\xi_G)\). The black dashed/dotted line indicates the point of symmetry at \(z = t/2\). (For interpretation of the references to color in this figure caption, the reader is referred to the web version of this article.)
length modulation proportional to the antisymmetric part of the distortion, hence it introduces an amplitude (and no phase) strain mean of many-beam MS scattering simulations at a specimen containing the model strain profile $\phi(t)$, sharply beginning at (a) the entrance face, (b) $z_c/2$ and (c) $z_c$.

excitation error $s_c$

$$f_c(z, t) = 2e^{\pi R} \left\{ \frac{\kappa_c \cos(2\pi\kappa_c(t-\tau)) - \phi(z)}{\sin(2\pi\kappa_c t)} \right\}. \quad (28)$$

Note that the additional antisymmetric weighting is predominantly imaginary, hence it introduces an amplitude (and no phase) modulation proportional to the antisymmetric part of the distortion. Consequently, the effect of a non-zero excitation error to the reconstructed phase mainly reduces to changing the extinction length $\xi_c$ (see Eq. (17)). One can therefore fine-tune the extinction length in order to maximize the diffraction amplitude and thus the signal-to-noise ratio in the reconstructed phase [32]. A positive side effect of maximizing the diffracted beam amplitude is that the weighting function assumes one of the particularly simple shapes at $f_c^{\text{phase}}(z, t_e(n + 0.5)\xi_c))$, i.e., the weighting becomes independent from the real thickness of the specimen. Indeed maximizing the diffraction amplitude by adjusting the excitation error $s_c$ is common practice, when performing DFEH. Unfortunately, the precision of the goniometer and beamtilt provided by the instrument is limited which puts some restrictions on the latter method.

4.Summary and outlook

Based on dynamical scattering and perturbation theory we derived closed and easy to use analytical expressions for phases reconstructed by means of dark field EH. Accordingly, generally $z$-dependent strain fields are weighted with a $z$-dependent weighting function, when projected by the electron beam. This weighting function is predominantly symmetric with respect to the middle plane of the specimen and depends on the diffracted beam, the crystal potential, the specimen thickness and the deviation of the crystal orientation from ideal Bragg conditions. As a consequence, depending on the particular shape of the $z$-dependent strain fields and the diffracted beam, reconstructed phases might be more sensitive to surface strain or bulk strain. This is of particular importance since surface relaxation is difficult to avoid when preparing thin TEM specimen. We furthermore point out that this formalism, i.e. linear projection of strain or displacement fields with a weighting function, constitutes an important prerequisite towards the tomography of 3D strain fields, since it provides a link to the commonly used projection transformations (Radon transformation). In future publications we will present experimental evidence for the above presented theory.

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Appendix A. Detailed derivation of second order differential equation

In order to shorten notation and facilitate a better understanding of the transformations given below we redefined all reciprocal lattice vectors, i.e. $\mathbf{G}$ and $s_c$, to include the factor $2\pi n$. Only in the final result, we have drawn out the factor again to be coherent with the results shown in the main text. The transformation steps leading to a second order differential equation for the diffracted beam read

$$\frac{\partial^2 \phi_c}{\partial z^2} = i e^{\pi R} \left( C_\xi V_0 \left( \frac{\partial \phi_c}{\partial z} - i \frac{\partial \phi_c}{\partial \xi_c} \right) + C_\xi V_c \left( \frac{\partial \phi_c}{\partial \xi_c} - i \frac{\partial \phi_c}{\partial \xi_c} \right) \right) + i s_c \frac{\partial \phi_c}{\partial \xi_c}$$

(A.1)
Using the reciprocal lattice vector definition without the prefactor $2\pi$ one obtains Eq. \(10\)

$$\frac{\partial^2 \hat{\phi}_C}{\partial z^2} = \left(-i2\pi \left(\frac{\partial (\mathbf{G} \cdot \mathbf{u}(z))}{\partial z} - s_C \right) \frac{\partial}{\partial z} C^2_\mathbf{G} \mathcal{V}_C \mathcal{V}_C' - 4\pi^2 \frac{\partial (\mathbf{G} \cdot \mathbf{u}(z))}{\partial z} s_C \right) \hat{\phi}_C.$$  \hspace{1cm} (A.2)

The same procedure carried out for the transmitted beam yields a slightly modified expression

$$\frac{\partial^2 \hat{\phi}_C}{\partial z^2} = i C_\mathbf{G} \mathcal{V}_C \mathcal{V}_C' \frac{\partial}{\partial z} \left( i \frac{\partial (\mathbf{G} \cdot \mathbf{u}(z))}{\partial z} \frac{\partial \hat{\phi}_C}{\partial z} + s_C \frac{\partial \hat{\phi}_C}{\partial z} \right) \hat{\phi}_C.$$  \hspace{1cm} (Eq. \(17\))

$$\hat{\phi}_C = i \frac{\partial (\mathbf{G} \cdot \mathbf{u}(z))}{\partial z} \frac{\partial \hat{\phi}_C}{\partial z} + s_C \frac{\partial \hat{\phi}_C}{\partial z} \hat{\phi}_C.$$  \hspace{1cm} (A.3)

Again we note the result using the reciprocal lattice vector definition without the prefactor $2\pi$

$$\frac{\partial^2 \hat{\phi}_C}{\partial z^2} = \left(-i2\pi \left(\frac{\partial (\mathbf{G} \cdot \mathbf{u}(z))}{\partial z} - s_C \right) \frac{\partial}{\partial z} C^2_\mathbf{G} \mathcal{V}_C \mathcal{V}_C' \hat{\phi}_C \right).$$ \hspace{1cm} (A.4)

In case of $\mathbf{a}_u/\partial z = 0$ the two elementary solutions $\hat{\phi}_{1,2} = \exp(k(2\pi x_i z_2))$ are equivalent to those of the diffracted beam, i.e.

$$k_{1,2} = \frac{s_C}{2} \pm \frac{1}{2\pi} \sqrt{2^2 s_C^2 + C^2_\mathbf{G} \mathcal{V}_C \mathcal{V}_C'}. \hspace{1cm} (A.5)$$

With $\hat{\phi}_C(0) = 1$, \((1/iC_\mathbf{G} \mathcal{V}_C \mathcal{V}_C')(\partial \hat{\phi}_C/\partial z)(0) = 0\) the transmitted beam reads

$$\hat{\phi}_C = \left( k_{1,2} e^{ik_2 z} - k_{1,2} e^{ik_1 z} \right). \hspace{1cm} (A.6)$$

### Appendix B. Detailed derivation for first-order von-Neumann approximation

In order to shorten notation and facilitate a better understanding of the transformations given below we redefined all reciprocal lattice and wave vectors, i.e. $\mathbf{G}$, $s_C$, $k_{1,2}$ and $\kappa$, to include the factor $2\pi$. Only in the final result, we have drawn out the factor again to be coherent with the results shown in the main text. The integral formulation of the second order differential Eqs. (10) and (A.4) reads

$$\hat{\phi}_C = \hat{\phi}_C^{(0)} - \int_0^t \Gamma(t-z) \frac{\partial (\mathbf{G} \cdot \mathbf{u}(z))}{\partial z} \left( \frac{\partial \hat{\phi}_C}{\partial z} + s_C \hat{\phi}_C \right) \frac{dz}{dz} \hspace{1cm} (B.1)$$

and

$$\hat{\phi}_0 = 0 + \int_0^t \Gamma(t-z) \frac{\partial (\mathbf{G} \cdot \mathbf{u}(z))}{\partial z} \left( \frac{\partial \hat{\phi}_0}{\partial z} + s_C \hat{\phi}_0 \right) \frac{dz}{dz} \hspace{1cm} (B.2)$$

respectively, where

$$\Gamma(t-z) = \frac{1}{k_2 - k_1} \left( \exp(k_1 z) - \exp(k_2 z) \right) \hspace{1cm} (B.3)$$

denotes the Greens function of the dammed harmonic oscillator. The first-order approximation in a von-Neumann type series expansion is now obtained by replacing all expressions on the right hand side containing $\hat{\phi}$ with the corresponding zero-order expression $\hat{\phi}^{(0)}$, i.e.

$$\hat{\phi}_C^{(1)} = \hat{\phi}_C^{(0)} - \int_0^t \Gamma(t-z) \frac{\partial (\mathbf{G} \cdot \mathbf{u}(z))}{\partial z} \left( \frac{\partial \hat{\phi}_C^{(0)}}{\partial z} + s_C \hat{\phi}_C^{(0)} \right) \frac{dz}{dz} \hspace{1cm} (B.4)$$

and

$$\hat{\phi}_C^{(1)} = \hat{\phi}_C^{(0)} + \int_0^t \Gamma(t-z) \frac{\partial (\mathbf{G} \cdot \mathbf{u}(z))}{\partial z} \left( \frac{\partial \hat{\phi}_C^{(0)}}{\partial z} + s_C \hat{\phi}_C^{(0)} \right) \frac{dz}{dz}.$$ \hspace{1cm} (B.5)

Repeating this procedure iteratively yields higher-order terms and eventually the complete von-Neumann series, which converges under the well-known convergence conditions [33]. Inserting the zero order solutions (Eqs. (14) and (16)) and omitting the perturbation order index, one obtains

$$\hat{\phi}_C = \frac{C_\mathbf{G} \mathcal{V}_C}{k_1 - k_2} \left( e^{ik_1 z} - e^{ik_2 z} \right) + \frac{i}{k_1 - k_2} \int \left( e^{ik_1 (t-z)} - e^{ik_2 (t-z)} \right) \frac{\partial (\mathbf{G} \cdot \mathbf{u}(z))}{\partial z} \frac{dz}{dz} \hspace{1cm} (B.6)$$

$$\hat{\phi}_0 = \frac{C_\mathbf{G} \mathcal{V}_C}{k_1 - k_2} \left( e^{ik_1 z} - e^{ik_2 z} \right) + \frac{i}{k_1 - k_2} \int \left( e^{ik_1 (t-z)} - e^{ik_2 (t-z)} \right) \frac{\partial (\mathbf{G} \cdot \mathbf{u}(z))}{\partial z} \frac{dz}{dz}$$

Reinserting the prefactor $2\pi$ one obtains Eq. (18) if $s_C = 0$ or the general Eq. (28) when applying the transformations leading from Eqs. (18)–(21).

The corresponding transformations for the transmitted beam read

$$\frac{\partial^2 \hat{\phi}_C}{\partial z^2} = \left( \frac{\partial (\mathbf{G} \cdot \mathbf{u}(z))}{\partial z} - s_C \right) \frac{\partial}{\partial z} \left( \frac{\partial \hat{\phi}_C}{\partial z} + s_C \hat{\phi}_C \right) \hspace{1cm} (B.7)$$

Setting $s_C = 0$ and performing a partial integration yields

$$\hat{\phi}_0 = \frac{1}{2} \left( e^{-ik_2 z} + e^{-ik_1 z} \right) + \frac{1}{2} \left( e^{ik_2 z} - e^{ik_1 z} \right) \mathbf{G} \cdot \mathbf{u}(z)$$

$$- \int \left( e^{ik_2 (t-z)} - e^{ik_1 (t-z)} \right) \frac{\partial (\mathbf{G} \cdot \mathbf{u}(z))}{\partial z} \frac{dz}{dz} \hspace{1cm} (B.8)$$

After applying the transformations leading from Eq. (18) to Eq. (21) and reinserting the prefactor $2\pi$ one obtains Eq. (26) including the weighting function $f_\mathbf{u}^0$ for the transmitted beam phase.
Appendix C. Multislise simulation parameters

We used the home-grown (S)TEM simulation software SEMI, implementing a numerical integration of the paraxial approximation of the approximated Klein–Gordon wave equation with predefined stepsize referred to as Multislise in the literature [29] to accurately propagate the electron probe through uniaxially strained Si lattices in Sections 3.2 and 3.5. Simulation parameters separated into TEM (Å), crystallographic (B) and numerical (C) parameters are summarized in the following: (A) \( U_s = 200 \, \text{kV}, \) beam tilt (parallel dark-field conditions for \([004]-\text{beam} = \arcsin (G)/q_0 = 9.2 \, \text{radm}, \) diameter of dark-field aperture = 4.6 radm; (B) \( a_B = 5.431 \, \text{Å}, \) \( S_G = Fd = 3 \, \text{m}, \) \( t = 250 \, \text{nm}; \) (C) \( \Delta T_{\text{slice}} = 1 \, \text{Å}, \) sampling = 0.008 nm\(^{-1}\); supercell = \( 128 \times 1 \times 695.17 \times 5.43 \, \text{Å}, \) atomic scattering potentials from Ref. [34]. The Si-atoms within the supercell are displaced according to the strain fields mentioned in the main text.

References


