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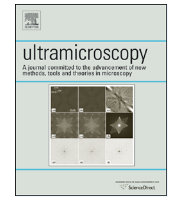

No surprise in the first Born approximation for electron scattering

M. Lentzen *

Ernst Ruska Centre, Research Centre Jülich, 52425 Jülich, Germany

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ABSTRACT

In a recent article it is argued that the far-field expansion of electron scattering, a pillar of electron diffraction theory, is wrong (Treacy and Van Dyck, 2012 [1]). It is further argued that in the first Born approximation of electron scattering the intensity of the electron wave is not conserved to first order in the scattering potential. Thus a “mystery of the missing phase” is investigated, and the supposed flaw in scattering theory is seeked to be resolved by postulating a standing spherical electron wave (Treacy and Van Dyck, 2012 [1]). In this work we show, however, that these theses are wrong. A review of the essential parts of scattering theory with careful checks of the underlying assumptions and limitations for high-energy electron scattering yields: (1) the traditional form of the far-field expansion, comprising a propagating spherical wave, is correct; (2) there is no room for a missing phase; (3) in the first Born approximation the intensity of the scattered wave is conserved to first order in the scattering potential. The various features of high-energy electron scattering are illustrated by wave-mechanical calculations for an explicit target model, a Gaussian phase object, and for a Si atom, considering the geometric conditions in high-resolution transmission electron microscopy.

1. Introduction

In a recent article, “*A surprise in the first Born approximation for electron scattering*” [1], it is argued that the far-field expansion of electron scattering, a pillar of electron diffraction theory, is wrong. It is further argued that in the first Born approximation of electron scattering the intensity of the electron wave is not conserved to first order in the scattering potential. By comparing the near-field and far-field expansions of electron scattering a missing phase of a quarter wavelength is detected, and this “mystery of the missing phase” is seeked to be resolved by an improved far-field expansion. With this improvement the interference of an outgoing and an incoming spherical wave is postulated, which would result in a standing scattered electron wave with the desired additional phase shift of a quarter wavelength.

These theses are wrong, however, and the notion of a missing phase is a mere misconception in the comparison of the near-field and far-field expansions. We will show that the far-field expansion of electron scattering is correct, and that no extra phase shift of a quarter wavelength has to be introduced for the scattered wave. We will further show that in the first Born approximation the intensity is indeed conserved to first order in the scattering potential.

In order to elucidate the arguments we will review the essential parts of scattering theory, focussing on high-energy electron scattering, with all underlying assumptions and limitations clearly presented. We have chosen as basis for the review classic textbooks on optics [2], wave mechanics [3,4], and of transmission electron microscopy [5,6], where rich sources of further material can be found. The various mathematical expressions describing features of electron scattering are illustrated by wave-mechanical calculations for an explicit target model, a phase object with a Gaussian phase distribution, considering the geometric conditions in high-resolution transmission electron microscopy. In order to keep the review, the illustration, and the discussion as concise as possible, explanatory material, that can be found in the above textbooks as well, has been organised in a number of Appendices.

2. Near-field and far-field expansion of electron scattering

In [1] the far-field expansion of electron scattering is derived through the Green function formalism, in which the wave equation is transformed into an integral equation. The boundary conditions of the scattering problem are then expressed via the proper Green functions, in this case an outgoing or an incoming spherical wave, or linear combinations thereof. Here lies at first a certain arbitrariness, as both spherical waves fulfil the mathematical requirement of decreasing rapidly enough for large distances from the scattering centre. The arbitrariness can be resolved by additionally judging the physical situation in the scattering problem.

We will resort to Kirchhoff's diffraction theory [2], which is free of mathematical arbitrariness and well-suited to derive, within

* Tel.: +49 2461616644.
E-mail address: m.lentzen@fz-juelich.de

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one framework, not only near-field and far-field expansion but also the propagation formula of electron scattering. The latter is important for all calculations in the intermediate range of distances from the scattering centre, and it can also be used to demonstrate the near-field and far-field limits.

Starting from the Fresnel–Kirchhoff diffraction integral [2] and defining a point source S, an object plane Σ, and a detector plane D parallel to Σ, the disturbance at the detector plane is

$$\psi(\boldsymbol{R}) = \psi_S(\boldsymbol{R}) - \frac{i}{\lambda z}\ \psi_S(\boldsymbol{R}) \iint_\Sigma (T(\boldsymbol{r})-1)\ \exp\left(\pi i\ \frac{|\boldsymbol{r}-\boldsymbol{R}|^2}{\lambda z}\right)\ d\Sigma, \quad (1)$$

where $\boldsymbol{r}$ is a vector in the object plane, $\boldsymbol{R}$ is a vector in the detector plane, z is the distance between these planes, λ is the wavelength, $T(\boldsymbol{r})$ is a transmission function defined on the object plane,

$$\psi_S(\boldsymbol{R}) = A_S\ \frac{\exp(2\pi iks)}{s}\ \exp(2\pi ikz) \quad (2)$$

the disturbance at the detector plane from the spherical source wave alone, A_S is the source amplitude, s is the distance from the source to the object plane, and $k=1/\lambda$. The origins for the vectors $\boldsymbol{r}$ and $\boldsymbol{R}$ be the points of intersection of object and detector plane with the optical axis running perpendicular to these planes through the point source. The above forms for $\psi(\boldsymbol{r})$ and $\psi_S(\boldsymbol{R})$ rely on the assumption of s being much larger than z and on the parabolic expansion of the distances from the source point to an object point, denoted by $\boldsymbol{r}$, and from this object point to a detector point, denoted by $\boldsymbol{R}$. Compared to the standard form of the Fresnel–Kirchhoff diffraction integral a separate term $\psi_S(\boldsymbol{R})$ has been isolated, being properly compensated by "-1" in the integral. This form is particularly useful if $T(\boldsymbol{r})$ differs from 1, representing the undisturbed transmission in vacuo, only for a small target area around the optical axis. Then $T(\boldsymbol{r})-1$ is zero outside the target area, and the integral extends only over that small area.

The near-field expansion of the Fresnel–Kirchhoff diffraction integral is

$$\psi_N(\boldsymbol{R}) = \psi_P\ \exp(2\pi ikz)T(\boldsymbol{R}), \quad (3)$$

with ψ_P the amplitude of the plane source wave, see Appendix A.

The far-field expansion is

$$\psi_F(\boldsymbol{R}) = \psi_P\ \left(\exp(2\pi ikz) + f(\mathbf{g})\frac{\exp(2\pi ikd)}{d}\right), \quad (4)$$

with the scattering factor

$$f(\mathbf{g}) = -\frac{\mathrm{i}}{\lambda}\ \iint_\Sigma (T(\boldsymbol{r})-1)\exp(-2\pi \mathrm{i}\mathbf{g}\cdot\boldsymbol{r})d\Sigma, \quad (5)$$

the diffraction vector $\mathbf{g}=\boldsymbol{R}/\lambda z$, and setting $d=z+R^2/(2z)$ in the phase of the scattered wave. See Appendix B on the convention for the symbol $f(\mathbf{g})$. A careful consideration of all the geometric approximations used so far shows that these are justified for the small scattering angles of high-energy electron scattering, and that the phase errors introduced are small enough to derive the far-field limit from the Fresnel–Kirchhoff diffraction integral. The conditions are shown in Appendix C.

The near-field and far-field expansions can now be investigated more closely by assuming a phase change $\phi(\boldsymbol{r})$ of the electron wave over the target area, and $\phi(\boldsymbol{r})=0$ outside, so that

$$T(\boldsymbol{r}) = \exp(i\phi(\boldsymbol{r})) \quad (6)$$

over the target area, and $T(\boldsymbol{r})=1$ outside. This model, together with the near-field expansion (3), is the phase grating approximation of electron scattering, the eikonal approximation of wave optics, and the Wentzel–Kramers–Brillouin approximation of quantum mechanics. In high-energy electron diffraction the phase change $\phi(\boldsymbol{r})$ is derived from the integral of the electric potential along straight trajectories through the target, parallel to the direction from source to target, see e.g., [5,6] or Appendix D.

Expanding $T(\boldsymbol{r})$ in orders of $\phi(\boldsymbol{r})$ yields the near-field expansion

$$\psi_N(\boldsymbol{R}) = \psi_P\ \exp(2\pi ikz)\left(1 + i\phi(\boldsymbol{R}) - \frac{1}{2}\phi^2(\boldsymbol{R}) + O(\phi^3(\boldsymbol{R}))\right), \quad (7)$$

and the scattering factor of the far-field expansion

$$f(\mathbf{g}) = \frac{1}{\lambda}\iint_\Sigma \phi(\boldsymbol{r})\exp(-2\pi i\mathbf{g}\cdot\boldsymbol{r})d\Sigma + \frac{i}{2\lambda}\iint_\Sigma \phi^2(\boldsymbol{r})\exp(-2\pi i\mathbf{g}\cdot\boldsymbol{r})\ d\Sigma + O(\phi^3(\boldsymbol{r})). \quad (8)$$

The leading term, linear in ϕ, of the scattered near-field wave has a constant phase of $\pi/2$ relative to the plane source wave, whereas the leading term of the scattered far-field wave (4) has a phase of $\pi R^2/(\lambda z)$ varying over the detector plane, if the real function $\phi(\boldsymbol{r})$ is at least centro-symmetric, so that the first term of $f(\mathbf{g})$ is real. For $R=0$ this phase is zero, and thus the phases of the leading linear terms differ by $\pi/2$ in the forward direction. This consideration can be extended to any order in ϕ, where the same difference in phase of $\pi/2$ appears.

3. A journey from the near-field to the far-field

In high-energy electron scattering both the near-field and far-field expansion derive from the Fresnel–Kirchhoff diffraction integral, as shown above, and it is thus possible to investigate the development of the phase of the scattered wave from the near-field to the far-field by Fresnel propagation.

The expansion of the transmission function $T(\boldsymbol{r})$ in orders of $\phi(\boldsymbol{r})$ in the Fresnel–Kirchhoff diffraction integral (1),

$$\psi(\boldsymbol{R}) = \psi_P\exp(2\pi ikz)\left(1 + \sum_{m=1}^{\infty}\frac{i^m}{m!}\iint_\Sigma \phi^m(\boldsymbol{r})\left(-\frac{i}{\lambda z}\right)\ \exp\left(\pi i\frac{|\boldsymbol{r}-\boldsymbol{R}|^2}{\lambda z}\right)d\Sigma\right), \quad (9)$$

is now solved for an explicit model of the phase object,

$$\phi(\boldsymbol{r}) = \phi_0\ \exp\left(-\frac{r^2}{2b^2}\right), \quad (10)$$

yielding

$$\psi(\boldsymbol{R}) = \psi_P\ \exp(2\pi ikz)\left(1 + \sum_{m=1}^{\infty}\frac{i^m}{m!}\phi_0^m\frac{2\pi b^2}{2\pi b^2 + im\lambda z}\ \exp\left(-m\pi\frac{R^2}{2\pi b^2 + im\lambda z}\right)\right), \quad (11)$$

in which the coordinate on the detector, $\boldsymbol{R}$, and the propagation distance, z, can be expressed in dimensionless quantities $\zeta=\lambda z/(2\pi b^2)$ and $\boldsymbol{\rho}=\boldsymbol{R}/(\sqrt{2\pi}\ b)$ so that the scattered part, the sum in (11), becomes

$$S(\rho) = \sum_{m=1}^{\infty}\frac{i^m}{m!}\frac{\phi_0^m}{1+im\zeta}\ \exp\left(-m\pi\frac{\rho^2}{1+im\zeta}\right), \quad (12)$$

where the strength of the scattering and the rate of convergence of the sum depend on ϕ_0, the peak of the phase change (10). The scattering factor (8) becomes

$$f(\mathbf{g}) = \frac{2\pi b^2}{\lambda}\sum_{m=1}^{\infty}\frac{i^{m-1}\phi_0^m}{m!\ \ m}\ \exp\left(-\frac{2\pi^2 b^2}{m}g^2\right), \quad (13)$$

which can be rewritten as $f(\theta)$ or $f(\rho)$, see Appendix E.

A map of the intensity at the detector plane, displayed in Fig. 1, the modulus squared

$$|\psi(\boldsymbol{\rho})|^2 = |\psi_P|^2|1+S(\boldsymbol{\rho})|^2 \quad (14)$$

as a function of the propagation distance, ζ, exhibits a number of characteristic features, shown here for the case of weak scattering, $\phi_0=0.1$. A set of Fresnel fringes radiates from the target area, $\rho<1$ at $\zeta=0$; the area covered by fringes is bounded by a cone $\rho=\zeta$ around the ζ axis, which corresponds to the upper limit, θ_{max}, of