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Transfer and reconstruction of the density matrix in off-axis electron holography

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ABSTRACT

The reduced density matrix completely describes the quantum state of an electron scattered by an object in transmission electron microscopy. However, the detection process restricts access to the diagonal elements only. The off-diagonal elements, determining the coherence of the scattered electron, may be obtained from electron holography. In order to extract the influence of the object from the off-diagonals, however, a rigorous consideration of the electron microscope influences like aberrations of the objective lens and the Möllenstedt biprism in the presence of partial coherence is required. Here, we derive a holographic transfer theory based on the generalization of the transmission cross-coefficient including all known holographic phenomena. We furthermore apply a particular simplification of the theory to the experimental analysis of aloof beam electrons scattered by plane silicon surfaces.

1. Introduction

Intensity distributions recorded by transmission electron microscopy (TEM) generally depend on the probability density as well as the spatial and temporal coherence within the scattered electron beam after interaction with an object. For instance, high-resolution images obtained from elastically scattered electrons are modulated by the combined influence of coherent lens aberrations and the partially coherent source summarized in the transmission cross-coefficient (TCC) [1,2]. Even more importantly, different inelastic scattering processes [3–9] and statistical fluctuations within the object [10], within the energy and momentum distribution of the electron beam or within the surrounding environment [11–13] also change the coherence properties of the beam. These modulations can be effectively described within the framework of density matrix transfer through the object and electron microscope.

Regarding the transfer through the object, van Hove showed in 1954 that the dynamic form factor relates the diffracted intensity and density-density correlation in the object [14]. However, since this concept only describes the diagonal elements of the density matrix (intensities) detectable at the back focal plane, it is insufficient for considering image formation of inelastically scattered electrons at the image plane. Therefore, the dynamic form factor was later generalized to the mixed dynamic form factor (MDFF) [15] incorporating also the off-diagonals (i.e. coherence). In first order Born approximation the MDFF is proportional to the cross-spectral density (known from classical optics) at the back focal plane [16]. The inverse temporal Fourier transformation of the cross-spectral density is called mutual coherence function in turn [17,18]. Equivalent to the cross-spectral density, the quantum mechanical density matrix description [19,20] was applied to describe inelastic scattering phenomena in first order and beyond [9]. The MDFF-concept provides powerful approximations describing image formation influenced by plasmon excitations [8,21–24], single electron excitations [25–30] and phonon excitations [25,26,24].

To obtain a comprehensive picture on the scattering process a measurement comprising both probability density and coherence is therefore indispensable. However, the quantum nature of the measurement process [31,32] prohibits direct access to this combined data in principle. This general information loss implies, e.g. the impossibility of exactly predicting the backward and forward evolution of intensity from a single intensity measurement (in the space below the object). This is the reason, why inelastic scattering cross-sections retrieved from measurements at the back focal plane of the objective lens have no or just very limited predictive power for the formation of intensities measured at the image plane.

Therefore we are interested in experimentally reconstructing the (reduced) density matrix of a probe electron after interaction with an object. Since information about the coherence is lost by intensity measurements, they have to be artificially encoded in the intensity distribution before measurement. This can be realized by relating different positions within the wave field with each other, i.e. by using electron holography [33,34]. In the off-axis type

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electron holography [35], the Möllenstedt biprism realizes a superposition of two partial waves in the image plane, which are separated by a certain distance in the object plane [36]. Thus, the resulting interference fringe pattern contains correlations of the wave field measurable in terms of local interference fringe contrast and phase shift with respect to a reference [37,35,38,39]. Especially, inelastically scattered electrons attenuate characteristically the fringe contrast, which was studied by means of inelastic electron holography in past [40–44]. Theoretical considerations [22,23] show that the coherence experimentally measured in this manner is related to distinguished off-diagonal elements of the electron beam density matrix selectable experimentally by changing the biprism voltage. Based on these results, we generally investigate the method off-axis electron holography for reconstructing the density matrix of inelastically scattered electrons. This requires a detailed transfer theory of the density matrix in a holographic TEM, which was partially considered already in Ref. [45]. For elastically scattered waves in conventional TEM, the transfer was described using the transmission cross-coefficient (TCC) [17, p. 530] applied to TEM imaging [1,2,46–50]. Similarly, the transfer of inelastically scattered electrons was described with the help of methods developed in classical optics [17, p. 537 using the propagation of mutual coherence [16,25,26,24]. But to our knowledge, only the *intensity* at the image or back focal plane was discussed. In this paper we develop in a first step a generalized transfer theory describing the intensity *and coherence* at various detection planes in the transmission electron microscope based on the concept of the TCC. In a second step, we incorporate the Möllenstedt biprism [36] and generally relate the recorded interference fringe pattern to the electron density matrix at the object exit plane. This is the prerequisite for the interpretation of further experimental results.

This paper is organized as follows. In Section 2, we formulate the transfer of the reduced density matrix of the probe electron ρ_s under partially coherent illumination. Here, we further generalize the TCC concept for the propagation of density matrices from the object to the image plane. This is the prerequisite for the discussion of the holographic transfer theory deduced in Section 3. There, we derive the holographic TCC describing in general the density matrix transfer in case of a general off-axis electron holographic experiment. We will interpret centre band and sideband contributions to the interference pattern in terms of modulated diagonal and off-diagonal elements of the density matrix at the object exit plane. We show that all phenomena in conventional off-axis electron holography are included in the presented theory. In Section 4, we introduce a procedure for density matrix reconstruction, which requires simplifications in the transfer theory. We deduce corresponding conditions and verify them numerically. In Section 5 we apply the developed procedure to a famous experiment as proposed in Ref. [51], which was intended to visualize the quantum-to-classical transition by decoherence: We reconstruct the density matrix from an interference fringe pattern of aloof beam electrons [52] inelastically scattered by a planar silicon surface.

2. Density matrix transfer in TEM

This section introduces the transfer of the density matrix using the concept of the TCC. We first relate the density matrix to the measurable current density. Subsequently we transfer the density matrix from the electron source plane downwards to the object plane, which is optically located in the far field of the electron source. Here, the electron beam is modulated by interaction with the object. Isoplanatic aberrations affecting the image plane are treated by energy dependent phase shifts at the back focal plane of the objective lens. By integration over the beam ensemble parameters we will identify the generalized TCC. Throughout this section we will employ twisted Seidel coordinates. That means we neglect image magnification and rotation in the following implying that coordinates at the image plane are the same as at the object plane for instance.

2.1. The density matrix of the probe electron

The time integrated current density (intensity) of the scattered electron beam is the main signal in transmission electron microscopy, which is detected, e.g. by CCD-cameras. The scattering of high energy electrons in the range of 20 keV–1 MeV leads to comparably small lateral momenta. Thus, the expectation value of the current density becomes proportional to the quantum mechanical probability density of the electron wave ψ_0

$$\mathbf{j} = \frac{\hbar}{2im}(\psi_0^* \nabla \psi_0 - \psi_0 \nabla \psi_0^*) \approx \frac{\hbar \mathbf{k}_0}{m} |\psi_0|^2. \quad (1)$$

Here, m denotes the relativistic mass of the beam electron and $\mathbf{k}_0$ is the initial mean wave vector. Due to interaction with the object the beam electron entangles with internal degrees of freedom of the object [31,32], i.e. the product of object ground state $\tau_0(\boldsymbol{\xi})$ and the impinging beam electron $\psi_0(\mathbf{r})$ transforms to

$$\Psi(\mathbf{r}, \boldsymbol{\xi}) = \psi_0(\mathbf{r})\tau_0(\boldsymbol{\xi}) \rightarrow \sum_i \psi_i(\mathbf{r})\tau_i(\boldsymbol{\xi}). \quad (2)$$

Here, the wave functions τ_i represent orthonormal eigenfunctions in the Hilbert space of the object and $\psi_i = \langle \tau_i | \Psi \rangle$ are expansion coefficients depending on the probe electron degree of freedom $\mathbf{r}$, which we interpret as partial waves of the scattered electron. Measuring the probability density of the electron beam then consists of taking the absolute square of these waves and additionally integrating over all not observed object degrees of freedom $\boldsymbol{\xi}$:

$$\rho_s(\mathbf{r}) = \sum_{ij} \psi_i(\mathbf{r})\psi_j^*(\mathbf{r})\langle \tau_j | \tau_i \rangle = \sum_i |\psi_i(\mathbf{r})|^2. \quad (3)$$

The measurement destroys the phase relation between these waves ψ_i leading to an incoherent summation of intensities. Consequently the state of the probe electron changes irreversibly from a pure to a mixed state, which cannot be described by a single wave function. This corresponds in general to a non-unitary evolution of the electron state from the object entrance plane to the object exit plane. It is then convenient to use a more comprehensive description of the quantum state comprising both pure and mixed states in one quantity, the density matrix defined as [19,20]:

$$\rho_s(\mathbf{r}, \mathbf{r}') := \sum_i \psi_i(\mathbf{r})\psi_i^*(\mathbf{r}'). \quad (4)$$

Here, ρ_s denotes the reduced density matrix describing the state of the electron at the object exit plane. The term "reduced" indicates the integration over the internal object degrees of freedom as conducted in (3). The high convenience of the density matrix description now lends from the following observation. One can show that the any quantum mechanical expectation value of operators solely acting in the beam electron Hilbert space may be obtained from the density matrix without any knowledge about the entangled object. For instance, the diagonal elements of the density matrix (4) are proportional to the intensity of a pure/mixed state in the sense of (1). Furthermore, the off-diagonal elements describe the coherence of the probe electron state. The mixed state properties of the single probe electron introduced by inelastic interaction, i.e. probe electron–object entanglement, will be referred to as *state coherence* in the following (subscript s). The state coherence is determined by electron–electron correlations