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Quantitative determination of elastic and inelastic attenuation coefficients by off-axis electron holography

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ABSTRACT

Off-axis electron holography is a well-established transmission electron microscopy technique, typically employed to investigate electric and magnetic fields in and around nanoscale materials, which modify the phase of the reconstructed electron wave function. Here, we elaborate on a detailed analysis of the two characteristic intensity terms that are completing the electron hologram, the conventional image intensity and the interference fringe intensity. We show how both are related to elastic and inelastic scattering absorption at the sample and how they may be separated to analyze the chemical composition of the sample. Since scattering absorption is aperture dependent, a quantitative determination of the corresponding attenuation coefficients (reciprocal mean free path lengths) requires the use of holographic image modi with well-defined objective aperture stops in the back-focal plane of the objective lens. The proposed method extends quantitative electron holography to a correlated three-in-one characterization of electric and magnetic fields, *Z*-contrast and dielectric losses in materials.

1. Introduction

In the past, off-axis electron holography (EH) has been established as a valuable transmission electron microscopy (TEM) characterization method mainly because of the simple proportionality between the reconstructed phase of the electron wave and static electric and magnetic potentials in and around the sample valid under medium (nanometer) resolution imaging conditions (out-of-zone-axis crystal orientation) [1]. Here, the phase shift is proportional to the projected electrostatic potential and the magnetic vector potential of the specimen projected along the beam propagation direction, which has been used in a multitude of studies of electric and magnetic solid state physics phenomena (see [2–8] and references therein).

In addition to the phase, a medium resolution off-axis hologram contains the conventional image intensity and the amplitude of the interference terms as valuable information, which is, however, only occasionally exploited to remove the thickness dependence of the measured projected potentials [9–12]. It is well-known that the conventional image contrast is generated by the so-called scattering absorption due to the interception of those electrons, which have been scattered into angles larger then the aperture stop of the TEM [13,14]. For typical aperture semi-angles larger then several milliradians, this contrast mechanism is dominated by elastic scattering on the screened Coulomb potentials of the atoms in the material, hence may be used to discern different chemical compositions [15]. For medium resolution imaging conditions, the image intensity damping is of Lambert–Beer type. This approximation breaks down at a certain specimen thickness, because multiple scattering eventually increases the probability to scatter into angles larger than the objective aperture in a non-linear manner. The thickness range for Lambert–Beer type scattering increases towards materials with low scattering power, large acceleration voltages and large aperture angles [14,16,17].

In their pioneering work McCartney and Gajdardziska-Josifovska [9] showed that the interference amplitude is dampened also in a Lambert–Beer fashion by inelastic scattering events. This result was subsequently used to remove the thickness dependency from the measured projected potentials as well as to determine inelastic mean free path lengths (MFPL) [9,10], which, however, were occasionally at odds [11,12,18] with theoretical computations or other experimental findings, notably from electron energy loss spectroscopy (EELS). Later, various experiments [19–21] widened this perspective by revealing that a non-vanishing interference contrast may be obtained also from inelastic scattering events (plasmons in that case). In an effort to explain this intriguing result Schattschneider and Lichte [22] showed that the interference amplitude relates to off-diagonal elements of the density matrix pertaining to the inelastically scattered electron beam. Moreover, Verbeeck et al. [23] pointed out that the intensity of these inelastic interference terms may be neglected for the interference distances above 100 nanometers, which are typically used in medium

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resolution off-axis EH.

In a recent tomographic study on GaAs–$Al_{0.33}Ga_{0.67}As$ core–shell nanowires [15], we showed that the interference amplitude of the electron wave is affected by elastic scattering absorption in addition to the inelastic one and discussed under which conditions elastic and inelastic attenuation may be separately reconstructed by means of off-axis holography. This opened new prospects to the precise determination of sample thicknesses, the separation and correlation of chemical composition and electric potential changes and the study of very low electron-energy losses by means of the coherence based energy-filter property of off-axis EH. In the following we elaborate on this result in order to establish a more quantitative interpretation of the observed attenuation in terms of physical properties of the material. To this end we illuminate the theoretical background and perform several experimental studies on a variety of materials. Here, we particularly pay attention to the correct adjustment and gauging of the scattering angle limiting objective aperture, which plays a crucial role for the measured attenuation coefficients. Finally, we reinterpret various previous MFPL measurements reported in literature thereby reducing (and partially removing) eventual discrepancies in comparison to other techniques or theory.

2. Theory

This section contains a detailed derivation of the elastic and inelastic contrast mechanisms valid under medium resolution holographic imaging conditions. A short version of which, omitting some details pertaining to the inelastic scattering mainly, has been previously given in Ref. [15]. Therefore, the reader, familiar with this theory or interested more into the experimental details and results, may skip the following lines and jump directly to the final expressions for centerband and sideband attenuation given by Eqs. (19), (22) and (24), (25), respectively.

Within the scope of the axial scattering approximation, mostly valid for medium resolution out-of-zone-axis imaging conditions, any lateral kinetic energy within the electron beam may be neglected [1]. Accordingly, the von Neuman (or kinetic [24]) equation governing the propagation of the electron beam's density matrix

$$\rho\left(\mathbf{r}, \mathbf{r}', z\right) = \sum_m \Psi_m\left(\mathbf{r}, z\right)\Psi_m^*\left(\mathbf{r}', z\right) \tag{1}$$

may be approximated as

$$\frac{\partial \rho}{\partial z} = -i\left[\hat{H}, \rho\right] = -i\sum_m \left(\Psi_m^*(\mathbf{r}', z)H_m(\mathbf{r}, z)\Psi_m(\mathbf{r}, z)\right. \tag{2}$$

$$\left. -\Psi_m(\mathbf{r}, z)H_m(\mathbf{r}', z)\Psi_m^*(\mathbf{r}', z)\right), \tag{3}$$

where the Hamiltonians H_m are scalar valued in the position representation because the kinetic energy term is neglected. Moreover, the Hamiltonian functions generally depend on the index m to allow for a varying composition of the quantum state, e.g., due to inelastic scattering.

In the case of purely elastic scattering of a pure wave function $\left(\rho(\mathbf{r}, \mathbf{r}', z) = \Psi(\mathbf{r}, z)\Psi^*(\mathbf{r}', z)\right)$ the axial Hamiltonian [25]

$$H_{\mathrm{ax}} = C_E\Phi(\mathbf{r}, z) + \frac{i}{2}\mu_{\mathrm{el}}^{(\alpha)}(\mathbf{r}, z) \tag{4}$$

contains the electrostatic potential Φ as a real (interaction constant $C_E = 7.3\frac{\mathrm{rad}}{\mathrm{V\mu m}}$ at 200 kV), phase shifting, part and the elastic attenuation coefficient μ_{el} as imaginary part, dampening the axial wave field. This phenomenological incorporation of scattering absorption is based on the observation that electrons, which have been scattered into angles larger then the semi-angle $\alpha = O\,(\mathrm{mrad})$ of a typical aperture stop, possess a small probability to be backscattered into forward direction again. Consequently, they may be incrementally removed from the axial wave field to a good approximation and we can write

$$\frac{\partial \rho(\mathbf{r}, \mathbf{r}', z)}{\partial z} = -i\left(H_{\mathrm{ax}}(\mathbf{r}, z) - H_{\mathrm{ax}}(\mathbf{r}', z)\right)\rho(\mathbf{r}, \mathbf{r}', z), \tag{5}$$

which has the solution

$$\rho(\mathbf{r}, \mathbf{r}', z) = T_{\mathrm{el}}(\mathbf{r}, \mathbf{r}', z)\rho(\mathbf{r}, \mathbf{r}', 0), \tag{6}$$

where function T, depending on two spatial coordinates, is referred to as mutual object transparency in the following. In case of purely elastic axial scattering, the transparency

$$\mathrm{T}_{\mathrm{el}}(\mathbf{r}, \mathbf{r}') = T_{\mathrm{w}}(\mathbf{r}) \otimes T_{\mathrm{w}}^*(\mathbf{r}') \tag{7}$$

is separable into two wave transmission functions

$$T_{\mathrm{w}}(\mathbf{r}) = \exp\left(i\int_0^{t(\mathbf{r})} C_E\Phi(\mathbf{r}, z)\mathrm{d}z\right) \times \exp\left(-\frac{1}{2}\int_0^{t(\mathbf{r})} \mu_{\mathrm{el}}^{(\alpha)}(\mathbf{r}, z)\mathrm{d}z\right) \tag{8}$$

containing the electrostatic potential Φ, projected over the thickness t, as phase and the projected elastic damping coefficient as amplitude argument. Accordingly, the elastic damping, caused by the absorption of electrons elastically scattered into angles larger than the aperture stop with semi-angle α, takes on a Lambert–Beer form in the axial scattering approximation.

Similar to the phenomenological treatment of scattering absorption within axial elastic scattering, we may now approximate the redistribution of the electrons among the various incoherent channels m due to inelastic (including thermal diffuse) scattering by incorporating a mutual inelastic mixing coefficient μ_{inel} into the axial von Neumann equation [26,27]

$$\frac{\partial \rho(\mathbf{r}, \mathbf{r}', z)}{\partial z} = -i\left(H_{\mathrm{ax}}(\mathbf{r}, z) - H_{\mathrm{ax}}(\mathbf{r}', z)\right)\rho(\mathbf{r}, \mathbf{r}', z) - \mu_{\mathrm{inel}}(\mathbf{r}, \mathbf{r}', z)\rho(\mathbf{r}, \mathbf{r}', z). \tag{9}$$

By comparison with the exact expression (2) we see that this mixing term is an approximation to the sum over m-dependent Hamiltonians (stripped from the m-independent elastic part), which may not be separated into a sum of two terms depending solely on $\mathbf{r}$ or $\mathbf{r}'$ anymore. Taking into account that phase shifts due to inelastic interactions are small compared to elastic ones, the mixing coefficient is to a good approximation real valued.

It follows from (9) that the mutual object transparency may be written as a product of the elastic and inelastic one, with the latter affecting the total absorption (main diagonal of $\mathrm{T}_{\mathrm{inel}}$) and coherence (off-diagonal of $\mathrm{T}_{\mathrm{inel}}$) following a Lambert–Beer law

$$\mathrm{T}(\mathbf{r}, \mathbf{r}') = \mathrm{T}_{\mathrm{el}}(\mathbf{r}, \mathbf{r}')\underbrace{\exp\left(-\int_0^{t(\mathbf{r})} \mu_{\mathrm{inel}}(\mathbf{r}, \mathbf{r}', z)\mathrm{d}z\right)}_{\mathrm{T}_{\mathrm{inel}}(\mathbf{r}, \mathbf{r}')}. \tag{10}$$

Note that linear approximations to the above transparencies, e.g., $\mathrm{T}_{\mathrm{inel}}(\mathbf{r}, \mathbf{r}') \approx 1 - \int_0^{t(\mathbf{r})} \mu_{\mathrm{inel}}(\mathbf{r}, \mathbf{r}', z)\mathrm{d}z$, closely related to the notion of the mixed dynamic form factor in Fourier space, have been thoroughly discussed by various authors to describe inelastic scattering, e.g., at plasmons, within the first order Born approximation [22,28–31]. Moreover, one readily observes that the "elastic" separability is lost under inelastic interaction because the beam electrons become entangled to the object's degrees of freedom [32,33], i.e.,