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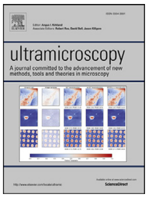

Correlative micro-diffraction and differential phase contrast study of mean inner potential and subtle beam-specimen interaction

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ABSTRACT

We present a correlative micro-diffraction and differential phase contrast (DPC) study within scanning transmission electron microscopy (STEM) on the determination of mean inner potential (MIP) and explain the origin of subtle beam-specimen interactions at the edge of wedge-shaped crystals using both experiment and simulation. Our measurement of MIP of Si and GaAs resulted in 12.48 ± 0.22 V and 14.15 ± 0.22 V, respectively, from directly evaluating beam refraction in micro-diffraction mode. DPC-STEM measurements gave very similar values. Fresnel fringes within the diffraction disk resulting from interaction of the highly coherent electron beam with the aperture are observed and a numerical simulation scheme is implemented to reproduce the effect of the specimen on the fringe pattern. Perfect agreement between experiment and simulation has been achieved in terms of subtle displacements of the fringes upon approaching the sample edge with the electron probe. The existence of the fringes has minor effect on the DPC-STEM signal, which is well below the noise level of our setup at practically reasonable acquisition times. We suggest the possibility to perform pseudo-contactless probing of weak potential differences in beam sensitive samples by evaluating the subtle displacements of Fresnel fringes quantitatively.

1. Introduction

Probing nanometer scale electrostatic (Coulomb) potential and/or the corresponding electric field, and establishing structure-properties relationships at this length scale in advanced functional materials are not only of fundamental interest but also of technological relevance and importance, especially for materials with application in the ever miniaturizing and integrating electronics. Prominent examples include the need to accurately locate and quantify junctions in MOSFETs [1], mapping the nanoscale charge distribution (which is linked to electrostatic potential via the Poisson's equation) in devices which include dielectric layers [2] or understanding the behavior of devices fabricated with materials involving piezoelectric effect in desired (e.g., piezoelectric nano-generators [3]) or undesired manner (e.g., quantum confined stark effect in InGaN-based light emitting doides [4]). Suitable techniques, especially those practically robust and compatible/extendable in combination with *in-situ* experimental setups, are on demand to unveil these challenges. Electrostatic potential, as pure phase object, can only be "seen" by the phase of probing radiation/particle wave. Transmission electron microscopy (TEM) based phase contrast and holography methods are the most suitable, if not the only, tools for this purpose, due to its high resolution and high sensitivity, which are fundamentally rooted in the short wavelength of high-energy electrons and the highly localized and strong electron-matter interaction. From the practical side, various techniques can be realized due to the versatility of modern electron microscopes that became even further extended by recent instrumental breakthroughs [5].

When nanoscale electrostatic potential is considered from the experimental point of view, mean inner potential (MIP) is the most important quantity [6]. The term "nanoscale" here refers to a length scale larger than the inter-atomic spacings where long-range and slowly-varying potentials, including the aforementioned examples, are considered and the continuum model still applies [7,8]. The MIP is, by definition, the local volume (usually considered at unit cell scale) average of the Coulomb potential of the sample, which can be theoretically calculated or measured from electrically neutral bulk samples with known thickness gradient (e.g., Refs. [9,10]). For a general sample assuming homogeneous potential in projection, the local average scattering potential (in absolute scale with vacuum level set to zero) being probed by the high-energy electrons, can be interpreted classically by superposition of MIP and the normally referred electrostatic potential (e.g.,

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potential induced by polarization, etc.), which is the deviation of local Coulomb potential from the MIP of the bulk.

Out of many forms electron holography [11], off-axis and in-line electron holography [12] in the TEM are the most popular and successfully implemented techniques for certain applications [13–15]. However, both methods show limitation for many of the above mentioned applications. In case of off-axis holography, interference fringes between electrons passing though the sample and vacuum are generated by the help of electron biprism(s). These fringes are recorded and then used to restore the local amplitude and phase of the electron wave leaving the sample (exit wave). Therefore, the available sample region being studied has to be located close to sample edge. In the case of in-line electron holography, a series of defocused images are recorded, and the phase information is reconstructed based on the (non-linear) transport of intensity equation [16,17]. Due to the large defocus, which is necessary to reveal Fresnel fringes at weak potential discontinuities [16], the retrievable resolution is limited. Alternative and/or complementary methods are needed to further expand the applicability to challenging studies.

Differential phase contrast (DPC) using electrons based on scanning TEM (STEM), which was originally proposed by Rose [18] and first realized experimentally by Dekkers and De Lang [19] in the 1970s, has seen a renaissance of interest recently. This is probably due to the recent demonstration of probing fields in real space at atomic scale [20] with aberration-corrected fine electron probe along with the progress in qualitative and quantitative interpretation [21–23]. It's worth to appreciate that DPC-STEM has been demonstrated robust and intuitive, and applied extensively in the studies of magnetic materials at micrometer to few tens of nanometer scales for decades, e.g. refs. [24,25] and references therein. Recently it also stepped into nano-scale with the probe spherical aberration correction in field-free mode [26,27] i.e., objective lens *off*, which is generally required for the studies of magnetic materials, but not necessary for study of electric fields.

In a typical DPC-STEM detection setup, the angular dependent intensities of the bright field beam disk are recorded by the intensities on the quadrant segments of (annular) bight field detector. The DPC-STEM signal composes the difference of intensity from opposite quadrants, which measures the shift of center of mass (COM) of the beam disk [21,27]. Only if the beam intensity can be assumed homogeneous (i.e., quasi-kinematic scattering condition, further discussed later), the DPC-STEM signal can be intuitively interpreted as a measure of the deflection of the beam disk as a whole. In this way, a two dimensional map of beam deflection while the probe raster over an arbitrary area of interest is recorded. Therefore, DPC-STEM is expected to show several advantages including 1) relaxed requirement of sample geometry and region of interest, 2) sharp features in focus, 3) flexible field of view and 4) simultaneous direct detection of phase and absorption contrast compared to alternative methods. Despite these arguments, the interpretation of DPC-STEM signal in recent literature had led to certain ambiguity for experimentalists, e.g. Refs. [20,21,28–30]. A recent work revealed [23] that the ambiguity arises due to the different length scales of electric fields under discussion and pointed out the importance of experimental conditions (most importantly, the interaction volume in physical words, or the optical settings practically) suitable for probing the respective scales of interest, i.e., atomic scale or nanometer scale.

At (inter-)atomic scale, sub-ångström electron probe, formed in practice with large convergence angle (> 20 mrad) and spherical aberration-correction, is required. Due to the probe spreading and strong dynamical scattering under these conditions, a full 2D diffraction patterns as function of probe position as well as dedicated simulations taking into account all necessary parameters describing electron-matter interaction are prerequisite to render a qualitative interpretation; and quantitative evaluation of experimental data is expected to expand from the studies of dedicated model systems [21–23]. At nanometer scale, in contrast, much smaller probe convergence angle is applied and correction of spherical aberration is not necessary in objective *field-on* mode (but critical in *field-free* mode [26]) to form nanometer-sized probes practically. This type of setup is sometimes referred to as *conventional* DPC-STEM, which is the focus of this paper. Under these conditions, much thicker samples can be studied and the classical view of simple and intuitive beam deflection is usually implied, where homogeneous transmission beam intensity is assumed [28]. This assumption is not necessarily justified for certain applications and thus leads to mis-interpretation, as demonstrated in MacLaren et al. [30]. In any case, investigation of the diffraction patterns under identical condition is indispensable to elucidate and/or evaluate the DPC-STEM results qualitatively and quantitatively.

On the practical side so far, there are only few applications, e.g., in Refs. [28–32], of DPC-STEM to the quantitative and systematic study of electrostatic potential (and electric fields) in crystals. MIP of a crystalline is a well defined quantity and has been extensively studied both experimentally and theoretically for many materials. Therefore, the measurement of MIP of a well-known material can be regarded not only as an additional independent measure of MIP of this particular material, but also as an approach to validate/calibrate the methods themselves for their further application in extracting electrostatic potential and electric field in samples quantitatively.

In this paper, we document our experimental studies on the measurements of MIP of Si and GaAs from cleaved crystal wedges by correlative micro-diffraction mapping and DPC-STEM and on the observation and simulation of phenomena resulting from the subtle interaction between the highly coherent electron beam and the specimen. We first summarize the theoretical essentials for interpreting the electron beam deflection/refraction at nanometer scale in Section 2. Then, our experimental setup, including optical settings and DPC signal calibration, will be detailed in Section 3. The results of measuring the MIP of Si and GaAs with micro-diffraction mapping and DPC-STEM, under various diffraction conditions, are presented in Section 4. Afterwards, in Section 5 the subtle interaction between highly coherent electron beam and specimen observed in micro-diffraction will be treated by numerical simulation resulting in perfect agreement with experiment. A conclusion will be drawn in the final section.

2. Theoretical essentials

We first summarize and derive, from a practical perspective, the essential equations for interpreting the results from micro-diffraction and DPC-STEM measurements in terms of electrostatic potential (and the corresponding electric field).

Under kinematic (i.e., single scattering) approximation, the phase difference of electrons passed through a non-magnetic specimen relative to that passed through vacuum is [33]:

$$\phi(\mathbf{r}) = C_E \int_{-\infty}^{+\infty} U(x,y,z)dz = C_E \int_0^t (U_i + U_e(x,y,z))dz, \qquad (1)$$

where

$$C_E = \frac{2\pi e}{\lambda} \cdot \frac{E_0 + E}{E(2E_0 + E)}$$

is the interaction constant, only dependent on the incident electron energy E (e elementary charge, λ electron wavelength and E_0 electron rest energy), $U(x, y, z)$ is the local Coulomb potential of the probed sample, written as superposition of the MIP U_i and the (additional) electrostatic potential $U_e(x, y, z)$ (cf. discussions in

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