Ultramicroscopy 139 (2014) 29–37

Contents lists available at ScienceDirect

Ultramicroscopy

journal homepage: www.elsevier.com/locate/ultramic

A nanocrystalline Hilbert phase-plate for phase-contrast transmission electron microscopy

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ARTICLE INFO

Article history:
Received 12 September 2013
Received in revised form
30 December 2013
Accepted 17 January 2014
Available online 25 January 2014

Keywords:
Transmission electron microscopy
Phase-plate
Phase-contrast
Crystalline film
Metal film

ABSTRACT

Thin-film-based phase-plates are applied to enhance the contrast of weak-phase objects in transmission electron microscopy. In this work, metal-film-based phase-plates are considered to reduce contamination and electrostatic charging, which up to now limit the application of phase-plates fabricated from amorphous C-films. Their crystalline structure requires a model for the simulation of the effect of crystallinity on the phase-plate properties and the image formation process. The model established in this work is verified by experimental results obtained by the application of a textured nanocrystalline Au-film-based Hilbert phase-plate. Based on the model, it is shown that monocrystalline and textured nanocrystalline phase-plate microstructures of appropriate thickness and crystalline orientation can be a promising approach for phase-contrast transmission electron microscopy.

1. Introduction

Recent advances in the application of physical phase-plates (PP) have opened up novel imaging possibilities in transmission electron microscopy (TEM). Thin-film-based PPs take advantage of the mean inner potential V_0 of amorphous C-films, which yields an acceleration of the electrons with respect to vacuum propagation. A linear dependence of the relative phase shift φ and the film thickness z is given for amorphous materials and crystalline structures under kinematic diffraction conditions [1]

$$\varphi(z) = -\frac{\pi}{\lambda U}\frac{1+2\alpha U}{1+\alpha U}V_0 z \quad \text{with } \alpha = 0.9785\times 10^{-6}\frac{1}{V} \tag{1}$$

with the electron wavelength λ and the acceleration voltage U. Located in the back focal-plane (BFP) of the objective lens, centrosymmetric Zernike PPs induce a relative phase shift of $0.5\,\pi$ between the electrons scattered by the sample and the unscattered beam [2,3]. Non-centrosymmetric Hilbert PPs impose a relative phase shift of π on electrons in one half of the diffraction pattern, while the zero-order beam and the second half of the diffraction pattern remain unaffected [4–6]. In weak-phase object approximation, both approaches provide contrast enhancement at low and intermediate spatial frequencies.

However, PPs fabricated from amorphous C-films are affected by contamination. Subsequent electrostatic charging of the PP microstructure induces artefacts and aberrations, which are difficult to correct. Post-processing amorphous C-coating and heating under improved vacuum conditions reduce contamination [7]. However, the lifetime of PPs still seems to be limited by a steady irreversible degeneration of the amorphous C-film.

Simultaneously, a variety of electrostatic PPs emerged, which apply metal shielded electrodes to reduce electrostatic charging [8–12]. However, contamination remains of fundamental concern and further limitations arise from insulator faces exposed to the zero-order beam. Moreover, fabrication and implementation of the electrostatic PPs remain challenging compared to their thin-film-based counterparts.

Limitations arising from amorphous C-films suggest the fabrication of metal-film-based PPs with a higher electrical conductivity and material stability. However, up to now, the application of metal films was avoided and, if applied, little consideration was given to the effect of their nanocrystalline structure [13]. Dynamic electron diffraction by grains in random crystalline orientation causes a random distribution of the relative phase shift, which prevents an application in phase-contrast TEM.

This work, for the first time, relates the properties of PPs fabricated from Au-films to their crystalline structure. A model for the simulation of crystalline PP microstructures is established. Simulations of monocrystalline and textured nanocrystalline PP microstructures illustrate the effect of crystallinity in phase-contrast TEM. Dynamic electron

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http://dx.doi.org/10.1016/j.ultramic.2014.01.002

diffraction results in a dependence of the relative phase shift on the film thickness and the crystalline orientation. Moreover, beams, which are diffracted by the crystalline PP microstructure, cause a shift of image information. The model is verified by experimental results obtained by the application of a textured nanocrystalline Au-film-based Hilbert PP.

2. Materials and methods

Metal-film-based Hilbert PPs were fabricated from nanocrystalline Au-films deposited by electron-beam evaporation (*Kurt J. Lesker – PVD 75*) on mica substrates (*PELCO – Mica Sheets*). A surfactant treatment of the mica substrates prior to the deposition reduces the adhesion of the Au-films and facilitates their removal in the floating process. The Au-films were floated off the mica substrates on a distilled water surface and deposited on Cu-grids. Subsequent plasma cleaning (*Binder Labortechnik – TPS 216*) eliminates surfactant residua. Using a focused ion-beam system (*FEI Company – Strata 400 STEM*), rectangular windows were structured into the Au-film, providing a Hilbert PP in the individual meshes. The Cu-grid was mounted on a customised objective aperture stripe (*Frey Precision*) and implemented in the BFP of the transmission electron microscope (*Philips – CM 200 FEG/ST*), which is operated at an acceleration voltage of 200 kV.

Plan-view TEM samples revealed the nanocrystalline structure of the Au-film. The film thickness was determined by a simultaneous deposition on a Si-substrate. Cross-section TEM samples were prepared using a focused ion-beam system. Assuming a mean inner potential of 25 V, Eq. (1) predicts a relative phase shift of 1.2 π induced by the deposited Au-film of 21 nm thickness [14].

Electron backscatter diffraction (EBSD) measurements were performed in a scanning electron microscope (*LEO – Gemini 1530*) equipped with an EBSD detection system (*HKL Technology – Channel 4*) to determine the crystalline orientation of individual grains of the Au-film.

The model established for the simulation of crystalline PP microstructures was implemented in a software package written for MATLAB (*The MathWorks*). Bloch-wave calculations were performed by BLOCH4TEM [15], while multislice calculations were carried out by STEMsim [16]. Powder-patterns were simulated by JEMS [17].

3. Results

3.1. Simulation of the effect of crystalline PP microstructures in phase-contrast TEM

This section presents the model, which is established to simulate the effect of crystallinity on the PP properties and the image formation process. Based on the idealised case of a monocrystalline PP microstructure, the considerations are extended to textured nanocrystalline PP microstructures with different percentages of grains in a preferred crystalline orientation, i.e. a variable degree of texture.

The effect of a crystalline Hilbert PP in phase-contrast TEM is schematically illustrated in Fig. 1. The illuminated sample area is uniformly separated into patches due to main memory limitations of common desktop computers. Individual patches are conveniently processed in reasonable computing time. Separation yields a central patch (■ in the object plane) on the optical axis and distant patches (▲) off the optical axis. Considering the image intensity of the central patch, the "*primary*" image intensity is given by the beams originating from the central patch and passing the BFP without being diffracted as indicated by the black optical path.

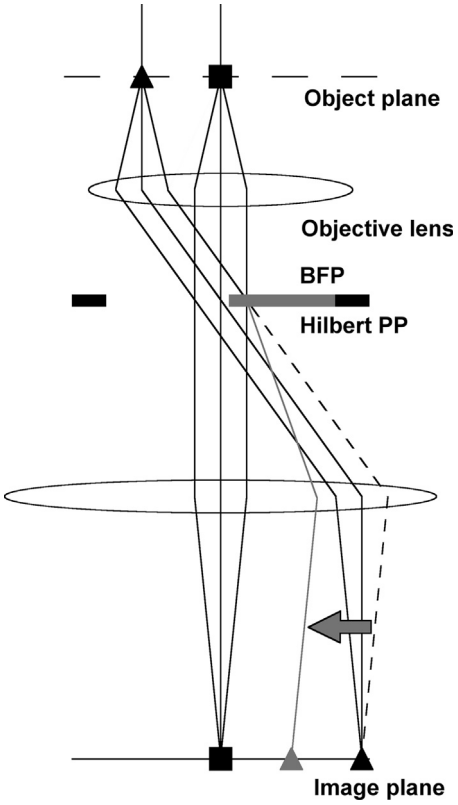

Fig. 1. Schematic illustration of the effect of a crystalline Hilbert PP in phase-contrast TEM. Diffraction of the outer right beam of the distant patch (▲ in the object plane) by the crystalline PP microstructure results in a shift of image information as indicated by the grey optical path and marked by the grey arrow. Hence, "*shadow*" images (▲ in the image plane) are superimposed on the "*primary*" image intensity, which is given by the electrons passing the BFP without being diffracted.

Moreover, electrons incident on distant patches, which are diffracted by the crystalline PP microstructure, cause a shift of image information $\mathbf{d}$ in the image plane

$$\mathbf{d} = \lambda f \mathbf{g} \tag{2}$$

which is determined by the focal length f and the reciprocal lattice vector $\mathbf{g}$ related to Bragg-diffraction. Diffraction of the outer right beam of the distant patch is indicated by the grey optical path and marked by the grey arrow. Hence, the image intensity is given by a superposition of the "*primary*" image intensity of the central patch and the shifted "*shadow*" images of distant patches.

Bloch-wave calculations quantify the effect of the crystalline PP microstructure [18]

$$\Psi^{\mathbf{O}}(\mathbf{r}) = \sum_{\mathbf{g}} \Psi^{\mathbf{O}}_{\mathbf{g}}(z) e^{2\pi i(\mathbf{u}_0 + \mathbf{g})\mathbf{r}} \tag{3}$$

with the lateral coordinate $\mathbf{r}$ and the refraction corrected incident wave vector $\mathbf{u}_0$. The fraction and the relative phase shift of the electrons whether or not diffracted by the crystalline PP microstructure are given by the complex factors $\Psi^{\mathbf{O}}_{\mathbf{g}}(z)$, which depend on the film thickness and the crystalline orientation $\mathbf{O}$. Diffraction is related to $\Psi^{\mathbf{O}}_{\mathbf{g}\neq\mathbf{0}}$, while $\Psi^{\mathbf{O}}_{\mathbf{0}}$ represents beams, which are not diffracted and not shifted by the crystalline PP microstructure.

An instructive application is the simulation of the effect of a monocrystalline Hilbert PP. Material- and instrumentation-specific data is listed in Table 1. The parameters are characteristic of the transmission electron microscope (*Philips – CM 200 FEG/ST*) used in this work and the simulations are performed under in-focus conditions. The illuminated sample area is deliberately increased for illustrative purposes and chosen to be square-shaped. An area