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Thin-film-based phase plates for transmission electron microscopy fabricated from metallic glasses

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Introduction

- Thin-film-based phase plates (PPs) enhance the contrast of weak-phase objects in transmission electron microscopy (TEM):

  Hilbert PP: Half of the diffraction pattern is covered by a thin film excluding the zero-order beam[1] (Fig. 1a)

  Zernike PP: Whole diffraction pattern is covered by a thin film with a small-diameter hole passed by the zero-order beam[2] (Fig. 1b)
- Conventional Hilbert and Zernike PPs are fabricated from amorphous C (aC) films
- ➡ Application limited by electron-beam induced degeneration and electrostatic charging
- ➡ **Search for alternative materials**[3]
- Metallic glass alloys $Zr_{65.0}Al_{7.5}Cu_{27.5}$ (ZAC) and $Pd_{77.5}Cu_{6.0}Si_{16.5}$ (PCS) are promising PP materials with beneficial characteristics:
  - Amorphous structure for temperatures up to the crystallization temperature
  - High electrical conductivity (three orders of magnitude higher than aC) [4,5,6]
- ➡ **Investigation of metallic glass alloys regarding their application as a PP material**

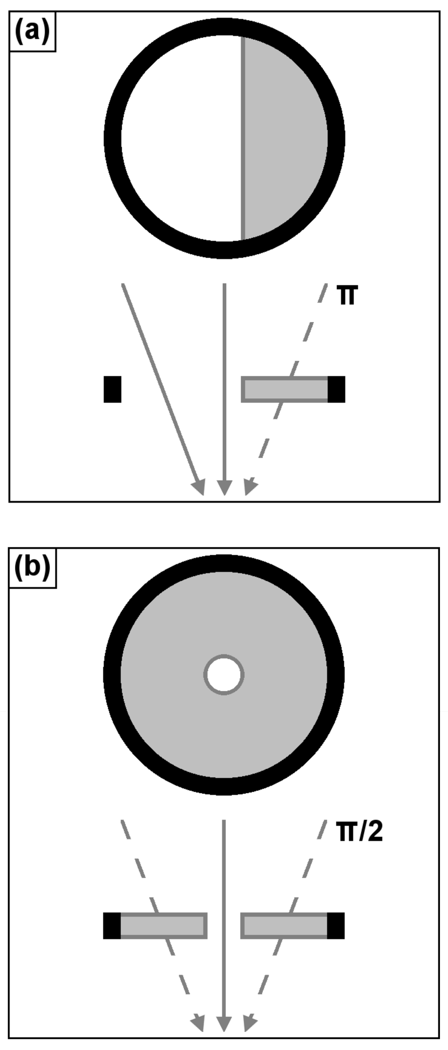

Fig. 1: Schematic illustration of a (a) Hilbert PP and (b) Zernike PP located in the back focal plane of the objective lens.

Fabrication of ZAC-film-based Hilbert/ PCS-film-based Zernike PPs

- Sputter deposition of the ZAC/PCS-film on mica/NaCl-substrates
- Floating process on grids
- Focused Ga^{+}-ion-beam structuring of rectangular windows (Fig. 2a)/small-diameter holes (Fig. 2b)
- Electron-beam evaporation of a thin aC-coating to avoid electrostatic charging
- Implementation in the back focal plane of a Philips CM200 FEG/ST

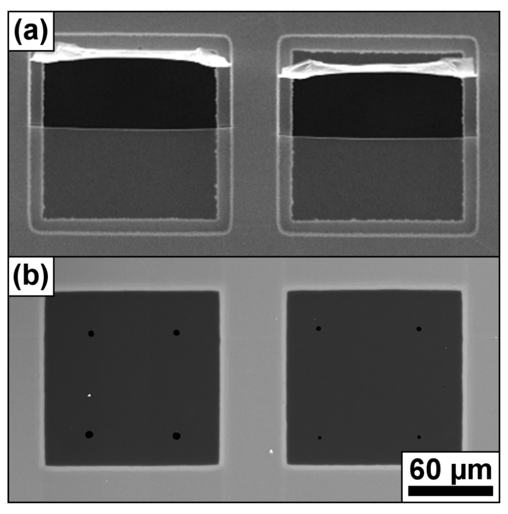

Fig. 2: Scanning electron microscopy image of a 1x2 array of (a) Hilbert PPs and (b) Zernike PPs.

Outstanding properties of metallic glass alloys

1. Inelastic scattering in the ZAC-alloy

- Inelastic plasmon scattering investigated by electron energy loss spectroscopy (EELS) in a ZEISS 912Ω:
  - Comparison of the plasmon intensity in low-loss EELS spectra of a 24 nm ZAC- and a 49 nm aC-film (Fig. 3)
  - Both films induce a phase shift of π at 200 keV
- ➡ Inelastic mean free path at 120 keV larger in ZAC (224 nm) than in aC (150 nm)
- ➡ Low probability for inelastic plasmon scattering in the ZAC-alloy

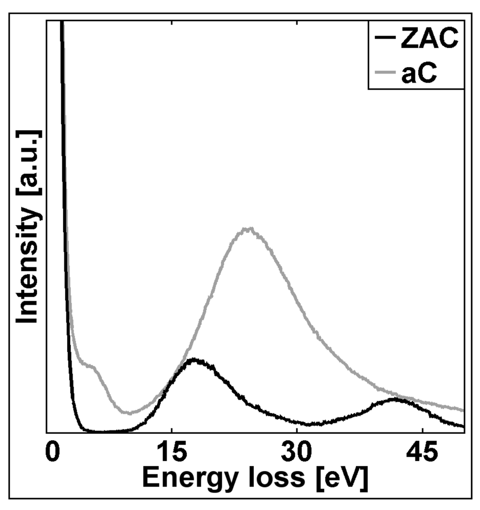

Fig. 3: EELS spectra of a 24 nm ZAC- and a 49 nm aC-film taken at 120 kV.

2. Oxidation resistance of the PCS-alloy

- Chemical composition of the PCS-alloy analyzed by energy-dispersive X-ray spectroscopy (EDXS) in a FEI Titan³ 80-300 using a cross section TEM sample of a PCS-film deposited on a Si-substrate
- Slight fluctuations of the chemical composition across the film thickness but the amorphous structure is preserved
- No oxide formation at the surface (blue line, Fig.4)
- ➡ Electrostatic charging by electrically insulating oxide layers avoided

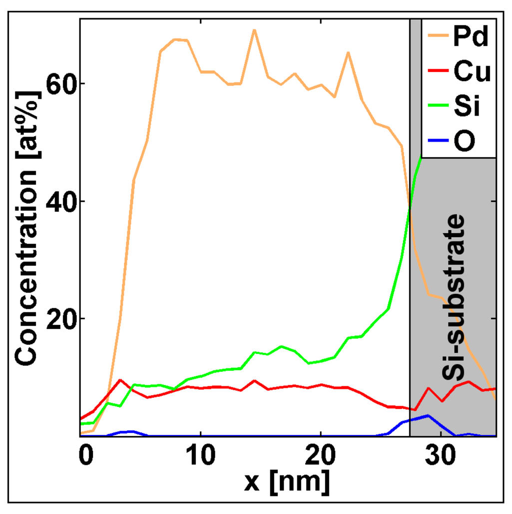

Fig. 4: Chemical composition of the PCS-alloy derived from an EDXS line profile.

Phase-shifting properties of a ZAC-film-based Hilbert PP

- Phase-shifting properties of the ZAC-film-based Hilbert PP revealed in the diffractogram of a phase-contrast image of an aC test object (Fig. 5a)
- Complementary behavior of the Thon rings in regions below (red) and above (green) the cut-on frequency (Fig. 5b)
- ➡ Phase shift of π induced by the ZAC-film-based Hilbert PP
- Negligible damping of the Thon rings above the cut-on frequency (Fig. 5b)
- ➡ **Information limit not impaired by inelastic scattering by the ZAC-alloy**
- Circular Thon rings indicate the absence of electrostatic charging

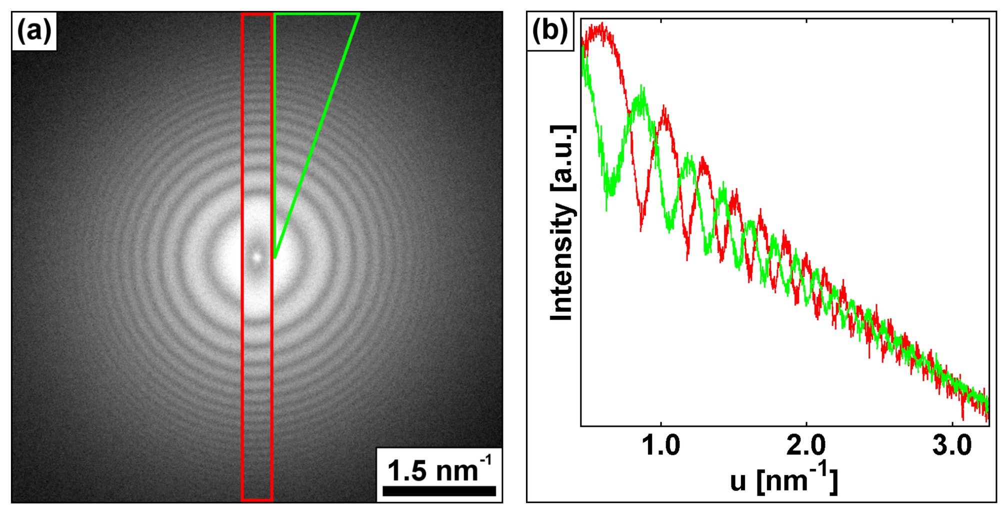

Fig. 5: Application of a ZAC-film-based Hilbert PP on an aC test object.
(a) Diffractogram of a phase-contrast image of an aC test object. Vertical red lines mark the cut-on frequency given by the PP edge.
(b) Azimuthally averaged intensity profiles taken in the red and green regions of (a).

Phase-shifting properties of a PCS-film-based Zernike PP

- Phase-shifting properties of the PCS-film-based Zernike PP revealed in the diffractogram of a phase-contrast image of an aC test object (Fig. 6a)
- Determination of the phase shift by a fit of the phase-contrast transfer function in regions below (red) and above (green) the cut-on frequency (Fig. 6b)
- ➡ Phase shift close to π/2 induced by the PCS-film-based Zernike PP
- Deviation of the measured intensity profile (solid green line) and the calculated fit (dashed green line) at the cut-on frequency indicates slight electrostatic charging close to the PP hole

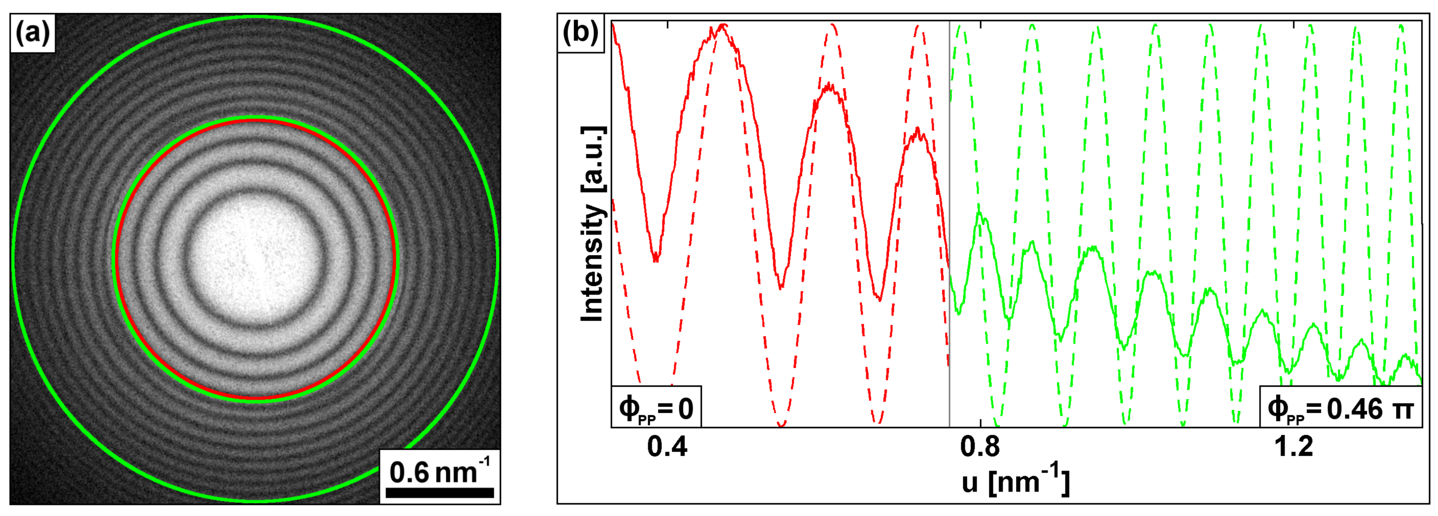

Fig. 6: Application of a PCS-film-based Zernike PP on an aC test object.
(a) Diffractogram of a phase-contrast image of an aC test object. The red circle marks the cut-on frequency given by the hole diameter. Spatial frequencies can be separated in regions below (red) and above (green) the cut-on frequency.
(b) Azimuthally averaged intensity profiles (solid lines) and fit of the phase-contrast transfer function (dashed lines) in the red and green regions of (a).

Summary

- Thin-film-based PPs fabricated from metallic glasses show the expected phase shifting properties
- Metallic glass alloys are promising PP materials with outstanding properties:
  - ZAC: Low probability for inelastic plasmon scattering
  - PCS: Absence of electrically insulating surface-oxide layer

References

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