Grazing incidence X-ray ptychography for in situ studies of thin sub-monolayer films of nanoparticles.

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Summary: Grazing incidence X-ray ptychography is a promising technique for investigation of thin sub-monolayer films of nanoparticles. Preliminary results on a reconstruction of a test sample are presented. A potential impact for characterization of surface energy, particle size and growth due to Ostwald ripening is expected.

1. INTRODUCTION

The performance of a number of systems in material and biological sciences is determined by their structural properties at the micro and nanoscale. The ability to image and analyze the surface structure at such length scales is crucial for correlating it with optical, electrical and other properties of the system. X-ray microscopy is a non-destructive and high-resolution technique well suited for *in situ* or *operando* studies of such systems without sophisticated sample preparation. Coherent diffraction imaging (CDI) is an X-ray microscopy technique with a high degree of penetration that does not rely on imaging optics such as lenses. It is thus not limited by aberrations of lenses or their numerical apertures and the resolution is determined by the largest scattering angle collected with sufficient signal to noise ratio [1]. In addition, CDI techniques are able to recover quantitative information of the sample's complex-valued refractive index with high accuracy that is not accessible by conventional X-ray transmission methods.

X-ray ptychography [2] is a scanning CDI technique that provides unlimited fields-of-view for the sample reconstruction and enables reconstruction of the generally unknown illumination function [3]. In a ptychography experiment, the incoming beam interacts with the sample experiencing attenuation and phase shift. The exit-wave is then propagated to a detector that collects intensity of diffraction patterns from a series of adjacent positions of the sample, overlapped by the illumination function. The phase that is lost at the detector needs to be recovered giving rise to the so-called phase-retrieval problem. Ptychography phase-retrieval algorithms play the role of an image-forming lens by recovering the unknown phase numerically, using iterative algorithms [4, 5].

CDI under grazing incidence [6, 7] is more suitable for investigation of surface properties of thin films. In such a configuration, coherent X-ray scattering from substrate-supported nanostructures is measured below the critical angle of the sample substrate. This shallow angle provides a high interaction cross-section with the sample because of the large footprint and the total external reflection of the incident beam from the substrate. The benefit of the grazing-incidence CDI over high-angle transmission-mode CDI [8] due to the larger imaged area along the spanned beam footprint is however somewhat offset by the reduced resolution along the beam propagation.

2. EXPERIMENTAL METHOD

The experiment was performed at the cSAXS beamline of the Swiss Light Source (SLS) facility in Switzerland. X-ray ptychography requires high-brightness coherent X-ray beam provided only at large scale facilities, i.e. synchrotrons and free electron X-ray lasers. The experiment was carried out at an X-ray energy of 6.2 keV, which provides the highest coherence of the beam at the cSAXS beamline. The X-rays are attenuated and phase-shifted when reflected and transmitted by and through the sample and substrate. The exit wave front is then propagating onto a far-field detector (Pilatus 2M) placed 7 meters downstream. After data acquisition, a phase-retrieval algorithm [9] is used to solve the problem of missing phase and reconstructs the complex refractive index of the sample and the illumination profile at the sample plane.

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3. RESULTS

We performed grazing incidence X-ray ptychography of Copper Zinc Tin Sulfide (CZTS) grains in a thin film solar cell absorber layer under optimized annealing conditions. Preliminary simulations show that reconstruction of the sample can be achieved by the proposed method using modified propagation of the exit wave front from the sample plane to the detector [7]. Figure 1 shows part of a Siemens star phantom that corresponded to the imaged area used for ptychographic reconstruction along with a complex-valued reflection function of the reconstructed Siemens star (under a grazing incidence angle of 0.27 degrees, both corrected with respect to the aspect ratio of the reconstruction pixel size).

The proposed method has a potential for *in situ* studies of particle-substrate interactions in a gaseous environment, under elevated temperatures and will allow describing time-evolution of an inhomogeneous sample structure. The grazing incidence configuration is of special interest for the study of sparse monolayers of nanoparticles that yield weak scattering signal in a conventional transmission configuration. Future improvements to the method will include grazing incidence ptychographic tomography for achieving isotropic resolution in object reconstruction. This requires better alignment of the measured projections and higher precision in the sample motion.
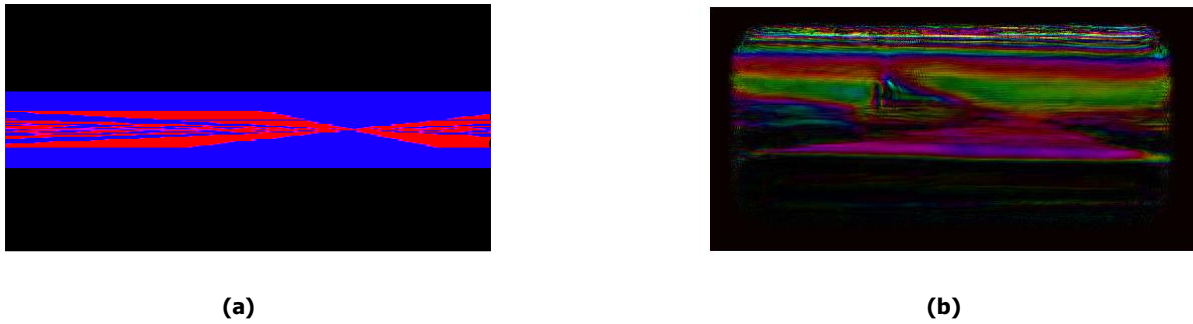

(a)   (b)

Figure 1: (a) Scanned part of the Siemens star phantom, the blue region is showing entire size of the supporting wafer, red pattern is the Siemens star with highly anisotropic resolution. (b) Reconstruction of complex-valued reflection function of the region simulated in (a).

References

[1] C. G. Schroer, et al. "Coherent x-ray diffraction imaging with nanofocused illumination," Phys. Rev. Lett. 101, 090801 (2008).

[2] J. M. Rodenburg, et al. "Hard-X-ray lensless imaging of extended objects," Phys. Rev. Lett., vol. 98, no. 3, pp. 1–4, 2007.

[3] Thibault, Pierre, et al. "Probe retrieval in ptychographic coherent diffractive imaging." Ultramicroscopy 109.4 (2009): 338-343.

[4] J. R. Fienup, "Reconstruction of an object from the modulus of its Fourier transform," Opt. Lett., vol. 3, no. 1, p. 27, 1978.

P. Gilbert, "Iterative methods for the three-dimensional reconstruction of an object from projections," J. Theor. Biol., vol. 36, no. 1, pp. 105–117, 1972.

[5] Vartanyants, I. A., D. Grigoriev, and A. V. Zozulya. "Coherent X-ray imaging of individual islands in GISAXS geometry." Thin Solid Films 515.14 (2007): 5546-5552.

[6] Sun, Tao, et al. "Three-dimensional coherent X-ray surface scattering imaging near total external reflection." Nature Photonics 6.9 (2012): 586.

[7] Levine, Joanne R., et al. "Grazing-incidence small-angle X-ray scattering: new tool for studying thin film growth." *Journal of Applied Crystallography* 22.6 (1989): 528-532.

[8] M. Odstrčil, A. Menzel, and M. Guizar-Sicairos, "Iterative least-squares solver for generalized maximum-likelihood ptychography," Opt. Express 26, 3108-3123 (2018)

Zhang, Bosheng, et al. "Coherent Diffractive Imaging With Arbitrary Angle of Incidence." U.S. Patent Application No. 14/839,738.