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► To cite this version:

I. Snigireva, A. Snigirev. High Resolution Higher Energy X-ray Microscope for Mesoscopic Materials. 11th International Conference on X-ray Microscopy (XRM), Aug 2012, Shanghai, China. 4 p., <10.1088/1742-6596/463/1/012044>. <hal-01572752>

HAL Id: hal-01572752

<https://hal.science/hal-01572752v1>

Submitted on 8 Aug 2017

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High Resolution Higher Energy X-ray Microscope for Mesoscopic Materials

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Abstract. We developed a novel X-ray microscopy technique to study mesoscopically structured materials, employing compound refractive lenses. The easily seen advantage of lens-based methodology is the possibility to retrieve high resolution diffraction pattern and real-space images in the same experimental setup. Methodologically the proposed approach is similar to the studies of crystals by high resolution transmission electron microscopy. The proposed microscope was applied for studying of mesoscopic materials such as natural and synthetic opals, inverted photonic crystals.

1. Introduction

Mesoscopic structures - photonic crystals have attracted much attention in the past decade for their ability in manipulation of the light. Photonic crystals are the structures in which the refractive index varies periodically in space on the length scale of light. They possess a photonic band analogue of the electron band gap of semiconductors. High quality photonic crystals have promising applications in the area of photonics. Colloidal crystals obtained by convective assembly technique can be used as a template for making inverted photonic crystals with a desired refractive index. In order to obtain crystals with a full photonic band gap the structure needs to be controlled since the photonic band gap is highly sensitive to defects like stacking faults, dislocations and other deformations. To achieve perfect crystals, detailed knowledge about the crystal growth mechanism and resulting defect structures is required.

A major obstacle in studying the defect structure is the lack of appropriate methods that allow seeing the internal structure of photonic crystal in three dimensions. The application of TEM in the study of these materials is limited since the technique's probing depth is a few hundred nanometers. SEM is restricted to obtaining structural information from the surface. The only possibility for studying internal crystal structure by SEM is through physically cutting crystals apart and studying their cut edges. This can modify the crystal structure in the cutting process. Different optical methods, such as laser diffraction, confocal optical microscopy and optical spectroscopy are widely used for the in-situ characterization of immersed, fluorescent, and refractive-index matched colloidal crystals with the particle diameters in the order of a micrometer. However numerous inverse opals, fabricated from highly-absorbing materials, including carbon, transition metal oxides and metals, are dry and in contact with air, which implies that the structures are not refractive-index matched. One way to overcome this is the infiltration of crystal with a refractive index-matching fluid before imaging the structure. But capillary forces acting on the crystal during this process may change the structure,

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resulting in unreliable characterization. Recently X-ray high resolution diffraction based on Fourier transform by compound refractive lenses allows accessing the internal structure of the colloidal crystals [1-4]. X-rays can be used for practically all materials and they do not require index matching or fluorescent labelling. Their shorter wavelength lift restrictions related to diffraction limitations. In present work we demonstrate a novel X-ray microscopy technique to study mesoscopically structured materials, employing compound refractive lenses, where we can get diffraction and imaging in the same experimental setup [5]. The proposed microscope was applied for studying of natural and synthetic opals, inverted photonic crystals and colloidal goethite board-like particles.

2. Concept

In transmission electron microscopy (TEM) switching from diffraction to imaging mode is achieved by changing the lenses strength in order to bring different parts of the wave front to the detector area. General consideration in both cases of X-rays and electrons is conveniently based on the Abbe formulations of the imaging process. The significant advantage of using lens-based methodology is the possibility to retrieve the diffraction pattern and real-space images in the same experimental setup. This concept, well-known in TEM, is the key ingredient of our approach. In the present work we demonstrate the transferability of the optical scheme of a TEM to x-ray scattering from mesoscopic structures, applying the concept of X-ray refractive optics as a Fourier transformer [4]. Similar to the studies of “traditional” crystals by high resolution transmission electron microscopy (HRTEM), the sample has to be tilted so that a low-index direction is exactly perpendicular to the X-ray beam. All lattice planes about parallel to the X-ray beam will be close enough to the Bragg position and will diffract the primary beam. The diffraction pattern is the Fourier transform of the periodic (complex) refractive index in two dimensions, and if in the objective lens diffracted beams and the primary beam are brought together again, their interference provides a back-transformation and leads to an enlarged picture of the periodic structure – in its 2D projection.

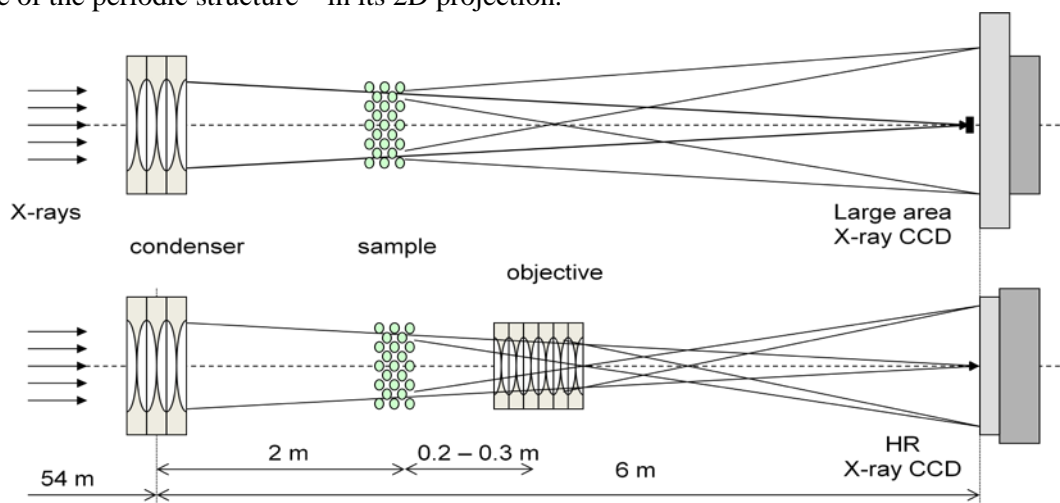


Fig. 1. Conceptual layout of the microscope. Diffraction (upper panel) and imaging (lower panel) modes

The microscope operates under a coherent illumination where a diffraction pattern of the specimen is formed in the back focal plane of the condenser and an inverted two-dimensional image of the object is formed by objective lens in the image plane [5]. The diffraction mode is used to investigate the crystal structure over the macroscopic distances and to orient the crystals parallel to the low index direction to perform high resolution imaging on the local scale. The structural image formation relies on phase contrast since it is made by interference of several diffracted beams. Only by using coherent illumination we can obtain contrast variations in the image of the specimen showing only variations in refractive index. The illumination coherence or spatial coherence length is determined by the effective source size and it is in the order of 100 - 200 μm . To ensure a reasonable contrast it should be a few orders of magnitude larger than the amount of fine detail to be imaged. The coherence in terms of the

angular source size determines the lens angular resolution and it is in the order of 10^{-6} rad. High angular resolution is needed for diffraction studies because for the mesoscopic materials the scattering angles are extremely small.

3. Experiment

The microscope was realized at the MOTB ID06 beamline using X-rays from 10 to 30 keV. It consists of condenser – used for coherent illumination of the sample in imaging mode and as Fourier transformer in diffraction mode, the objective lens and two CCD detectors. The condenser is comprised of 19 Be parabolic lenses with 300 micrometer radius of parabola apex producing focal length ~ 6 m. Condenser lenses were placed at a distance of 54 m from the undulator source. The collimating slits in front of the CRL limited the beam down to 200 μm in horizontal and vertical directions, thus fitting the effective aperture of the CRL. The objective lens assembly of 45-62 individual Be parabolic lenses with 50 micrometer radius of parabola apex was located at 2.3 m from the condenser. The sample was placed on the translation/rotation stage with the translation direction along the beam axis. Two X-ray 2D cameras were used: (i) large area Photonics science CCD detector with 18 μm resolution (9 μm pixel size) for diffraction experiments and (ii) high resolution Sensicam CCD detector with 1.3 μm resolution (0.645 μm pixel size). In the diffraction mode the direct focused beam was intercepted in front of the camera by a tungsten beamstop. Switching from the diffraction mode to the imaging mode was achieved by moving the objective lens into the beam, and the choice of proper camera (Fig. 1). Depending on the objective lens configuration the image distance (sample-to-objective lens distance) varied from 28 cm to 18 cm. Thus, the magnification factor between 10 and 30 was achieved. At maximum magnification a resolution of 100 nm was achieved.

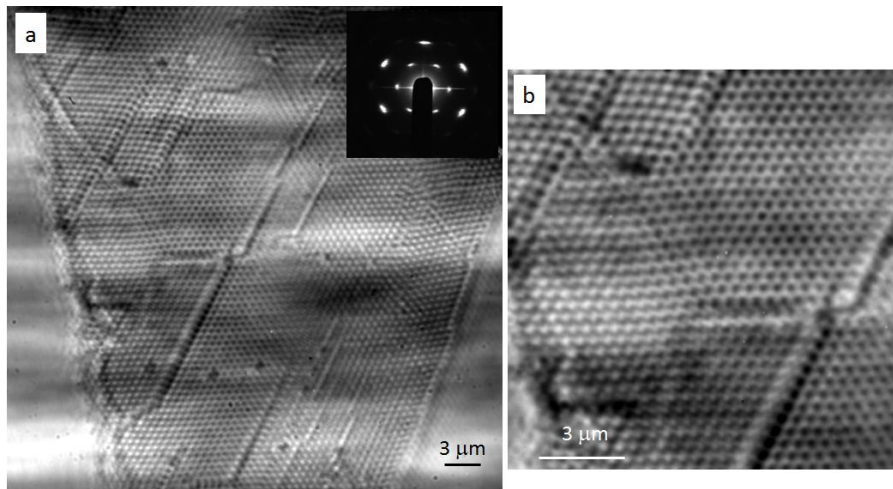


Fig.2 (a) X-ray image together with corresponding diffraction pattern of Co inverted photonic crystal; orientation is $\langle 001 \rangle$ zone perpendicular to the crystal surface. (b) Enlarged high resolution image of Co inverted crystal.

The microscope was applied for structural characterization of mesoscopic materials such as natural and synthetic opals, metal inverted photonic crystals. As an example we present here the microscopy study of cobalt inverted photonic crystals. Metallic inverse opal was prepared by electrodeposition of Co inside the voids of artificial opals. Artificial opals were fabricated using the novel synthetic approach based on electric-field –assisted vertical deposition of polystyrene microspheres onto conducting support. Monodisperse polystyrene microspheres with average diameter of 450 nm were synthesized by emulsifier-free emulsion of polymerization of styrene. The field of view in our experiments was varied from $50 \times 70 \mu\text{m}^2$ to $30 \times 50 \mu\text{m}^2$, depending of magnification. The sensitivity to the focal distance is quite high; 200 μm defocusing induces substantial blurring. Figure 2 shows an X-ray high resolution image of cobalt inverted photonic crystal recorded in $\langle 001 \rangle$ zone. A hexagonal type of arrangements is nicely visible.

The proposed microscope can be applied not only for photonic crystals, but for other mesoscopic materials, for example to study the behaviour of dispersion of colloidal goethite board-like particles under the magnetic field [6]. This system is opaque and hard to index match, therefore it unsuitable for studies with conventional microscopy methods. The particles possess a permanent magnetic moment along their long axis, together with an induced magnetic moment along the shortest particle axis. When a small magnetic field of about 200 mT is applied, the particles orient along the field and the smectic phase becomes highly oriented. The period between the vertical lines is about 380 nm, which is in agreement with the average smectic spacing obtained from diffraction (Fig. 3a). When the magnetic field was increased above 280 mT, the particles reoriented from parallel to perpendicular to the field. This is clearly seen in Fig. 3b, where the layers now possessed undulations as the long-range order was lost and the particles actually seem to have partly rearranged into a columnar phase.

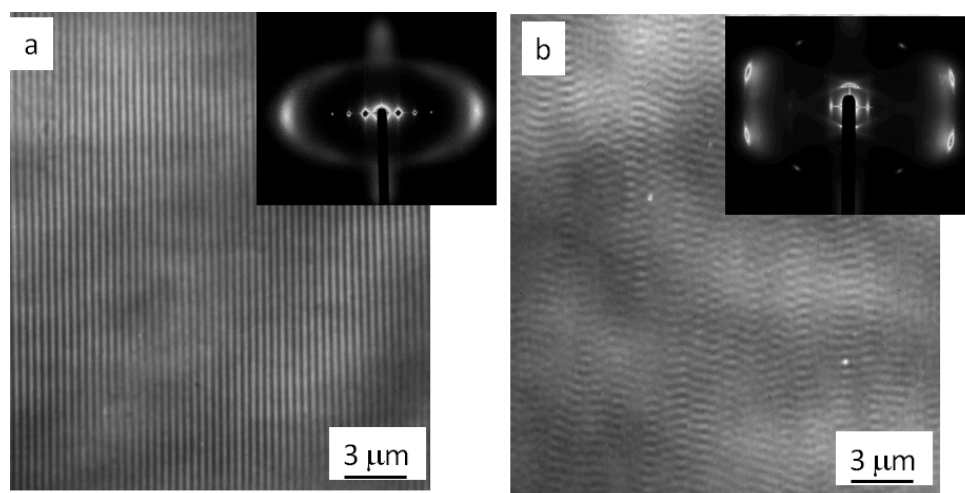


Fig.2 X-ray images together corresponding diffraction patterns of (a) highly ordered smectic phase of goethite rods in magnetic field of 200 mT and (b) reoriented smectic phase in the increased magnetic field 280 mT.

4. Conclusion.

The proposed approach provides the ultimate tool for the efficient studies of real structure of mesoscopic materials. Short acquisition times with modern area detectors allow the method to be extended to time-resolved studies and combined 3-D real/reciprocal space mapping. As immediate practical application of the microscope is the in-situ characterization of the real crystal structure during photonic crystal growth.

Developed X-ray microscope is conceptually similar to TEM; therefore a lot of approaches of TEM and HRTEM can be directly transferred to the domain of X-rays, including bright- and dark-field imaging, defocus series and exit-wave reconstruction for structure refinement, etc. Easy tunability in principle allows a multiscale investigation of materials.

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