New aspects of coherent hard X-ray imaging

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Summary: Hard X-ray radiography and tomography are common techniques for medical and industrial imaging. They normally rely on absorption contrast. However, the refractive index for X-rays is slightly different from unity and an X-ray beam is modulated in its optical phase after passing through a sample. The coherence of third generation synchrotron radiation beams makes a simple form of phase-contrast imaging, based on simple propagation, possible. Phase imaging can be used either in a qualitative way, mainly useful for edge-detection, or in a quantitative way, involving numerical retrieval of the phase from images recorded at different distances from the sample. The combination with tomography allows to determine at the micron scale the distribution of the complex refractive index. In combination with Bragg diffraction or reflection a sensitivity to atomic displacements or surface positions in the Ångström range is reached.

1 Introduction

Imaging is performed with a variety of probes, including light, electrons and X-rays. Hard X-rays (energy > 6 keV) offer a modest spatial resolution (micron scale), but are suited to 2D and 3D imaging of thick (millimeter scale) samples. Hard X-rays are commonly used for imagery in two ways. Radiography, in which the basis for contrast is inhomogeneous absorption in simple transmission, is extremely widespread and includes sophisticated variants such as computed tomography. In X-ray diffraction imaging (or X-ray topography) a beam diffracted by a single crystal is used to form an image revealing deviations from homogeneous reflectivity related to crystal defects.

Phase contrast as opposed to attenuation contrast is a powerful method for the investigation of light materials but also to distinguish, in absorbing samples, phases with very similar X-ray attenuation but different electron densities. Phase contrast imaging was pioneered in the early seventies by Ando and Hosoya [1]. Presently three hard X-ray methods of phase sensitive imaging exist: the interferometric technique [2, 3], the Schlieren technique [4, 5] and the propagation technique [6, 7]. The Schlieren approach is qualitative. The interferometric technique developed into a quantitative three-dimensional imaging technique but it requires sophisticated and extremely sensitive instrumentation. Furthermore the spatial resolution is in this case intrinsically limited to about 15 µm. The propagation technique is based on Fresnel diffraction and could develop recently thanks to the coherence of the X-ray beams available at third generation synchrotrons. This method is completely analogous to the defocusing method for imaging phase objects in electron microscopy, but with the difference that in the X-ray case the only in-focus position is that of the object itself, because efficient focusing optics are not commonly available in hard X-ray work.

Experimentally the sample is set in a (partially) coherent beam and the transmitted beam is recorded at a given distance \( D \) with respect to the sample that corresponds to the defocusing distance.
in a microscope. The region of the object mainly contributing to the corresponding point of the image (the first Fresnel zone) has a radius equal to \( r_F = \sqrt{\lambda D} \). When it is small compared to the typical transverse dimension \( a \) of the features in the sample, a separate fringe pattern shows up for every border in the sample, and the images are characteristic of the ‘edge-detection regime’ \( (r_F \ll a) \). Three-dimensional reconstruction of the boundaries inside the volume is feasible with the algorithm for absorption tomography [8]. At larger distance \( (r_F \approx a) \) several interference fringes show up in the radiographs. The resulting images, corresponding to the ‘holographic regime’, give little direct information on the sample. However, combining several such images, recorded at different distances, with a suitable numerical algorithm gives access to the phase modulation. For the largest distances one reaches the Fraunhofer limit \( (r_F \gg a) \).

Figure 1: Experimental set-up (operating in air) for phase sensitive imaging using the propagation or defocusing technique. The sample is illuminated with an extended, monochromatised and spatially coherent X-ray beam and the transmitted (or diffracted) beam is recorded at a distance \( D \) from the sample. The distance is easily varied and the sample can be rotated around a vertical axis for the tomographic acquisition.

The most striking advantage of this method is the extreme simplicity of the set-up. It is essentially the same as for absorption radiography. However for best results one needs a (sophisticated) third generation synchrotron and the optical elements of the beamline have to be carefully prepared to avoid spurious phase images [9].

2 Edge-enhancement versus quantitative imaging

Most of the work performed until now does not use specifically developed algorithms. Nevertheless edge-enhancement allows to visualise, in 3D, density discontinuities, such as reinforcing SiC particles in an aluminium matrix composite [10]. Fresnel fringes signal the presence and the position of cracks with an opening small compared to the spatial resolution. In-situ volume observations of the microstructure and damage at different strain levels can be obtained.

Phase imaging was also successfully used to investigate quasicrystals [11]. Phase radiographs revealed the presence of internal cavities, with a shape featuring the icosahedral point symmetry of the matrix, and crystalline precipitates. The combination with Bragg diffraction imaging allowed in this case to study the relationship between the microstructure and quasicrystal defects.

It is possible to fully exploit the quantitative information entangled in the Fresnel diffraction patterns. Phase retrieval is based on images recorded at different distances (typically four) and uses algorithms similar to those developed for high resolution electron microscopy [12, 13, 14]. The absence in the set-up of magnifying X-ray optics avoids aberrations such as spherical aberration and simplifies the phase retrieval procedure. On the other hand the spatial resolution is limited by the X-ray detector to about 1 micrometer. Phase maps retrieved for several angular positions of the sample (about 1000 ranging from 0 to 180 degrees) constitute the correct input for a tomographic reconstruction. This combined technique is called ‘holotomography’ and allows to determine the distribution of the electron density [15].

Figure 2a is a tomographic slice of an aluminium-silicon alloy recorded at \( D = 7 \) mm, sensitive only to variations in absorption. This is a map of the imaginary part of the refractive index.
It is impossible to distinguish the two phases, some bright spots appear corresponding to iron-rich inclusions. Figure 2b is a tomographic slice obtained for a single distance $D = 0.6\,\text{m}$, revealing density jumps as dark / light fringes. Figure 2c is a reconstructed map of the refractive index decrement, the difference between unity and the real part of the refractive index, which is proportional to the electron density. It clearly shows the slightly different densities of two (metallurgical) phases ($\Delta \rho \approx 0.05\,\text{g/cm}^3$). The grey phase is an aluminium-silicon eutectic and the dark phase is essentially pure aluminium with substitutional silicon.

Phase imaging at X-ray energies where the beam attenuation is strongly reduced allows to image specimens with good image contrast and lower deposited dose. This is crucial for imaging thick (millimeter range) biological samples in their natural, wet environment. Holotomographic imaging with micrometer resolution was performed in-situ on Arabidopsis as a model plant at a X-ray energy of 21 keV while keeping the dose acceptable.

### 3 Mapping with Ångstrom sensitivity

The spatial resolution presently obtained may seem modest considering the wavelength of the radiation used. Even with better X-ray optics becoming available the detectability, in simple transmission, is expected to be limited due to the weak interaction with matter to typically 10 nm. However, in combination with diffraction or reflection, a much better sensitivity can be achieved.

The combination with Bragg diffraction allows to image ferroelectric domains in periodically poled lithium niobate crystals and other non-centrosymmetric materials. Images of the Bragg diffracted beam at different distances from the sample give access, through numerical analysis, to the phase difference between the structure factor of oppositely poled regions [16]. This phase difference can be typically 140 degrees. It does not exist in simple transmission and is related to an atomic displacement of the niobium ion by 0.5 Å. Such an approach yields information on the relative position of the atomic structure in one domain type with respect to the other.

A beam reflected by a surface may acquire shifts in its phase related to surface shape errors. In this geometry height variations of the order of the X-ray wavelength ($< 1\,\text{Å}$) can introduce contrast of a few percent. The surface quality is a limiting factor in the use of X-ray mirrors for critical focusing or imaging applications. Coherent imaging can be a metrology tool in a range of sensitivity or field of view not covered by other techniques such as optical interferometry or atomic force microscopy. Phase retrieval allows to spatially resolve and measure the shape errors of the reflecting surface.
The above examples show that phase imaging, easily performed with coherent synchrotron radiation, increases extensively the possibilities of hard X-ray imaging.

References