

HARD X-RAY HOLOGRAPHY AT THE ESRF

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Hard X-ray holography permits the visualisation of the 3-D arrangement of atoms at the angstrom level. The development and future prospects of this new technique are described.

I NTRODUCTION

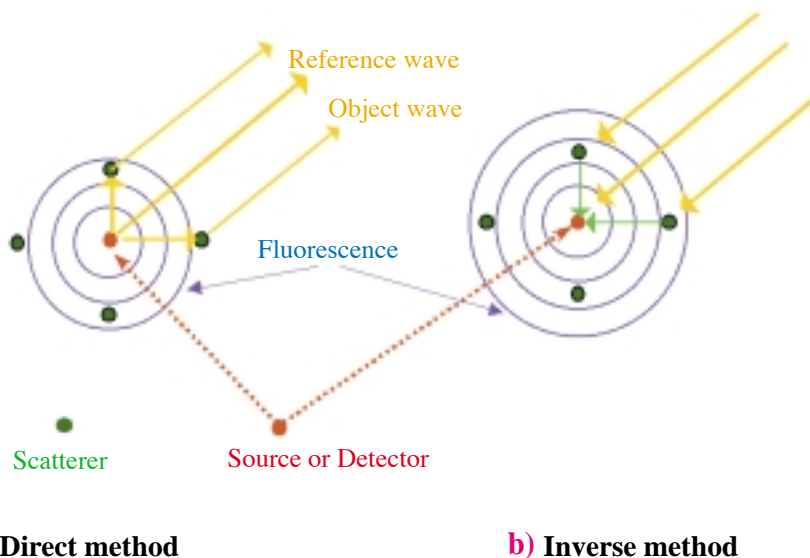
The knowledge of atomic and molecular structures is fundamental to physics, chemistry and biology. From the beginning of this century, much effort has been put into the development of reliable methods for structure determination. Diffraction techniques are the most common and the most highly developed for structural studies. However, not all of the problems can be solved by diffraction: well-known difficulties are that single crystals cannot be grown for all substances, and that a full structure determination is difficult from powder data alone. Sometimes the phase problem causes diffraction to fail – even with single crystals. As for non-periodic systems, like amorphous materials, quasi crystals etc., detailed structural information cannot be derived easily. Further difficulties arise with new artificial materials, which have structures that cannot always be solved by traditional crystallographic methods. Examples are the study of the local environment of low-concentration impurities in a sample, the atomic order in buried interfaces, and the atomic environment of surface adsorbates.

Therefore, it is not surprising that scientists are trying to find new techniques for structural investigations. Recently, atomic-resolution X-ray holography has emerged. It is based on the same principles as traditional holography with light: i.e. a coherent wave (called the reference wave) illuminates the object and the detector surface. The intensity modulation caused by the interference between the reference wave and the wave scattered by the object (called the object wave) is recorded. This interference pattern contains both the phase and the magnitude information of the object

wave. Therefore, the original wavefront can be reconstructed, giving the 3-D spatial arrangement of the objects. Although holography is used in many areas of science and everyday life, atoms in solids could not be imaged until recently. This is because the resolution is limited by the pixel size of the detector and the size of source, in addition to the wavelength. While it is relatively easy to decrease the wavelength to the Angstrom level, giving the possibility of atomic resolution, it is difficult to reduce the source size or to increase the detector's resolution. Abraham Szöke pointed out that individual atoms in a solid could be used as sources of radiation [1]. Based on his idea, experiments were performed, first using electrons [2] and later using photons [3] as hologram forming waves. Moreover, Gog and co-workers showed that the atoms present in a sample could be used as detectors instead of sources of radiation [4]. In spite of these pioneering efforts, many

problems relating to the experiment and evaluation of results remained to be solved. The most significant ones were the long measuring time, the experimental setup for synchrotron studies, background correction, and the method of reconstruction.

These problems prompted us to start a series of experiments at the ESRF. Our aim was to develop a prototype experimental setup for synchrotron holographic studies and at the same time to find reliable methods for background correction and reconstruction. In this article we would like to describe the most important results from these experiments. For clarity, first the principle of the "inside source and detector" holography is given. In our experiments the electronic system of the atoms is used as sources or detectors of X-ray radiation. Figure 1a shows the formation of a hologram in the case where the atoms act as point sources. A central atom is excited by an external



a) Direct method

b) Inverse method

Fig. 1: The XFH principle for (a) inside "source" and (b) "detector" holography.

source. In the de-excitation process, a fluorescent photon is emitted in the form of a spherical wave. This wave can reach the detector surface directly or after scattering by the neighbouring atoms. The two waves interfere producing an intensity modulation which is measured on a spherical surface surrounding the sample. The intensity modulation contains the hologram. In the other case i.e. when atoms are used as "point detectors" the external wave is incident to the sample in the form of a plane wave (Figure 1b). This can reach the central atom directly or by scattering on the neighbouring atoms. The two waves interfere at the detector atom (central atom) and the resulting field excites the electronic system. The probability of excitation is proportional to the strength of the field. Changing the direction of the incident radiation changes the phase relation between the direct and scattered waves resulting in oscillations of the fluorescent intensity. These oscillations contain the holographic information [5].

EXPERIMENTAL DEVELOPMENTS

It is clear from the above description, that the two types of measurements require a similar experimental setup. The main difference is that in the inside source case the fluorescent radiation has to be measured as a function of the detector position in relation to the sample. While using the atoms as "point

detectors" the direction of the incident beam has to be varied and the external detector has to collect the fluorescent photons from the full solid angle about the sample. The first step in our work was to develop the proper experimental setup for synchrotron measurements. It was constructed in a way that both inside "source and detector" holographic experiments could be done without remounting the sample. One of the critical components of the setup is the detection system. This should satisfy three conditions: first it has to discriminate the fluorescent photons from a relatively high background, second its angular acceptance has to be variable from the 1×1 degree² to the full solid angle (or as large a solid angle as possible), third it has to handle high count rates without appreciable dead time. The mechanics is another crucial part of the experimental setup. Although angular positioning and reproducibility do not have to be extremely precise, it does have to be fast - capable of 10 turns/second for one of the axes. For the two other axes rotation speed can be slower, about 2 degrees/second, but they both have to carry relatively heavy loads (3 kg). The third part of the experimental setup which needs attention is the data acquisition system. During the fast motion one has to read several counters in quick succession (every 200 microseconds).

We developed a setup which satisfies the above conditions, improving the system after every experimental run.

Here we will not describe all the steps but instead present the latest version of the setup which uses the full power of synchrotron undulator radiation. The experiments were done at beamlines **ID32**, **ID18** and **ID22**.

The experimental setup is shown in Figure 2, and a description of its characteristics follows. The optics are the least complicated part. A given harmonic of an undulator is used without monochromatisation since a small bandwidth is unnecessary. However, a mirror and an absorber are needed in order to single out a given harmonic – the so-called "pink" beam – which then passes through a thin foil, which scatters part of the incident beam. The scattered radiation is used for incident beam monitoring. The mechanical system comprises two coaxial vertical rotation stages (θ , θ') coupled in such a way that the lower goniometer (θ) carries the upper one (θ'). The upper goniometer in turn holds another smaller one with a horizontal axis (ϕ). The sample is fixed to this horizontal rotation stage so that its flat surface is perpendicular to this axis when illuminated by the X-ray beam. The detector is mounted on the lower goniometer. The third part of the experimental setup is the detector assembly. Since the count rate is very high, it could not be handled by single photon counters. Therefore, we used a Si diode in the current mode. However, this does not have energy resolution, which is necessary to suppress the unwanted radiation, so the energy

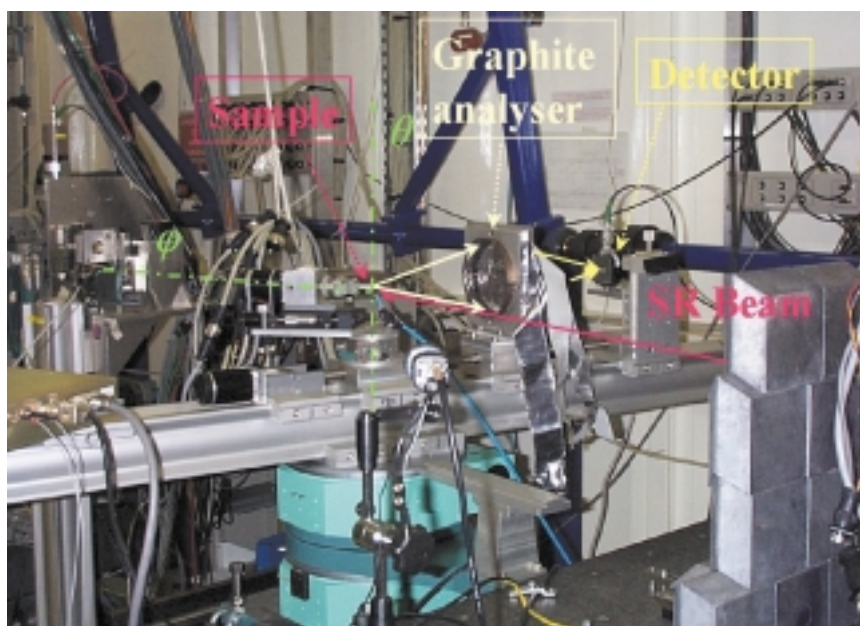
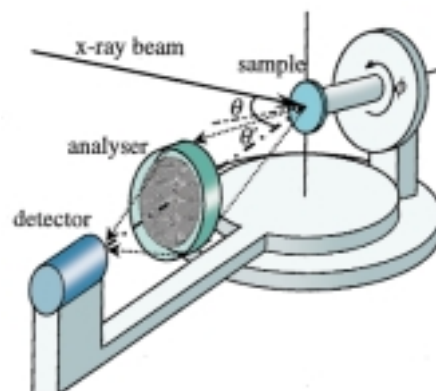


Fig. 2: Sketch of the experimental setup installed at the ESRF for the study of hard X-ray holographic measurements.





analysis was done before the detector by a doubly focusing pyrolytic graphite monochromator (Figure 2 [6]). This arrangement resulted in a large loss of detection solid angle compared to the laboratory measurements. Fortunately, this was more than compensated for by the high flux of synchrotron radiation. Typical estimated count rates were in the range of 10^{10} counts per second. Taking into account the statistical noise only, this would result in measuring times of a few seconds. However, the mechanical motion of the sample-detector system is not fast enough to scan a hemisphere in this short time. Therefore, the collection of a holographic data set at a single energy takes a few minutes.

HOLOGRAPHIC IMAGES

In the following part we would like to present a few examples of holographic imaging. Useful information can be derived from the holograms without any back-transformation. Since we use the atoms as point sources or detectors in the hologram forming process, the hologram shows the local symmetry of their environments. This is illustrated in Figure 3. The pictures show the recorded fluorescence intensities, projected on the sample surface of the upper hemisphere, after absorption correction. What is really seen is the standing wave patterns (or Kossel lines) rather than the holograms themselves, since the holographic oscillations are of much smaller amplitude. The pictures a, b and c were taken from samples having a three, four and five-fold symmetry axis perpendicular to their surface, respectively from an NiO[111] single crystal, an epitaxial FePt $L1_0$ film and an AlPdMn quasi-crystal. One can find other off-perpendicular symmetry points on these holograms from which the full local symmetry of the site can be deduced [7].

Of course, what we are finally interested in is the 3-D arrangement of atoms in real space. The next two figures show an example of this. The holograms of a CoO[111] sample are shown at four different energies (Figure 4). The reconstructed image of the Co atoms can be seen in Figure 5 [7]. In the evaluation process all four holograms were used to build a single high-resolution atomic structure. We would like to point out that

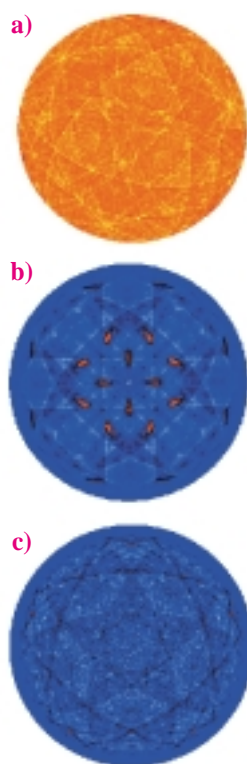


Fig. 3: Holograms and standing wave line patterns taken from samples having their surface parallel to crystallographic plane with 3 (NiO), 4 (FePt) and 5 (AlPdMn) fold axis. The FePt epitaxial film was prepared with A. Marty and B. Gilles, the quasi-crystal was supplied by F. Schmithusen and J. Chevrier.

Fig. 4: Holograms of CoO taken at energies 6925, 13861, 17444, 18915 eV respectively.

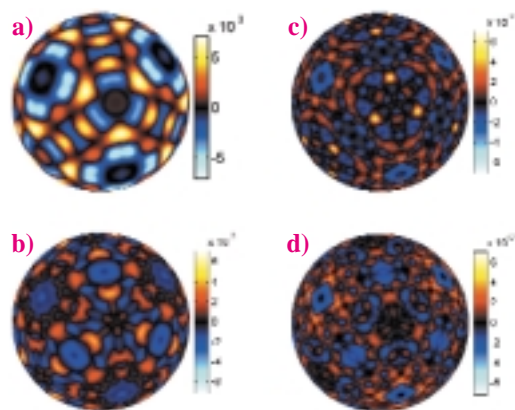
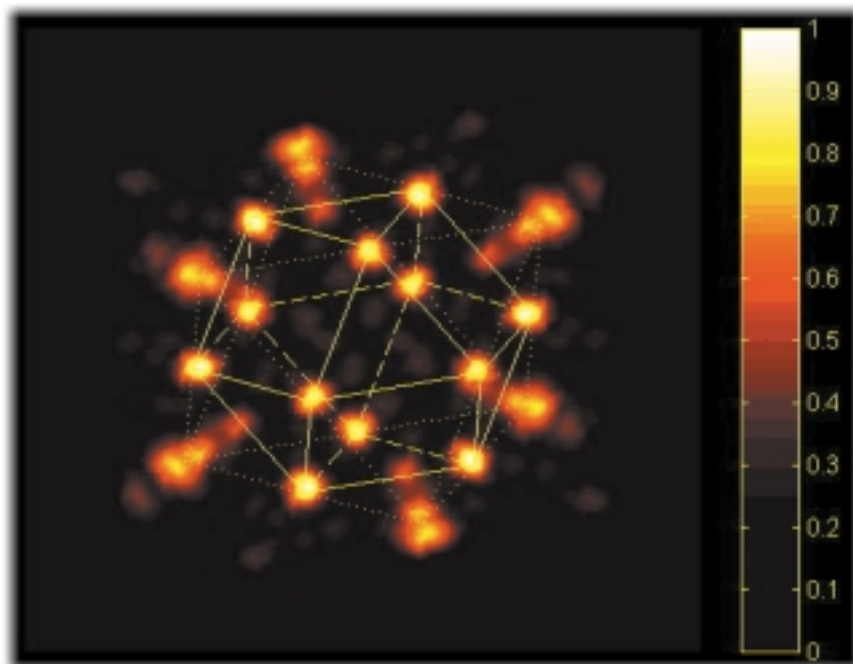


Fig. 5: The 3-D arrangement of Co atoms reconstructed from the holograms of Figure 4.



in this case the resolution is isotropic in contrast to all previous measurements, and its value (0.5 Å) is near to the diffraction limit.

FUTURE TRENDS

In spite of the substantial progress in the experimental and evaluation methods, there are many problems which need to be solved before the widespread application of this technique. Let us start with the easiest one to solve: the collection time of a hologram, which at a single energy is between 5 and 30 minutes. This does not seem too long, especially when compared with the duration of the first demonstration experiment (two months [3]). However, it would be advantageous to decrease the measuring time to 1 minute or below. The reason for this is that many artifacts inherent to holographic reconstruction can be eliminated by measuring the same sample at several (in the order of 10) energies [4,5,7]. In practical terms this would result in measuring a full data set for holographic reconstruction in 10 minutes instead of a few hours. The second problem is that we are unable to image light atoms. In all experiments so far, only relatively heavy atoms (from

iron up) could be imaged. However, in many applications especially in the case of biological samples, it would be crucial to see O, C, N etc. The third problem that hinders wider application of holography is the form of the sample. Presently single crystal samples with a large (5*5 mm²) flat surface can be measured. It would be very useful to extend the capabilities of holographic imaging to small (~10⁻³ mm³) arbitrarily shaped crystallites.

Of course there are many other areas where holography could be improved, but we stop here after mentioning the three most important ones. We have continued to work on the above mentioned problems, and have promising results, especially on the measuring time, on the reconstructed cluster size and the imaging of light atoms [8] and on the first application to thin films. ■

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APPOINTMENT OF A DIRECTOR OF ADMINISTRATION AND A DIRECTOR OF RESEARCH

The ESRF Council has opened the selection procedure for the appointment of

- a **Director of Administration** (from 1 February 2002) and
- **one of the ESRF's two Directors of Research** (from 1 June 2002).

Both appointments are for periods of five years.

For the time being, the eight delegations to the ESRF Council are invited to nominate candidates. People interested in contributing to this phase, for example by suggesting

candidates, are invited to contact one of the Heads of Delegation before mid November 2000. (The Council aims at having completed the recruitment for these positions one year before the start of the appointments.)

Further information on the ESRF, on the posts and on the procedure (notably the e-mail addresses of the Heads of Delegation) can be found on the ESRF web site at <http://www.esrf.fr>.