Exploiting the x-ray refraction contrast with an analyser: the state of the art

Alberto Bravin

European Synchrotron Radiation Facility, BP220, F-38043 Grenoble, France
E-mail: bravin@esrf.fr

Received 14 September 2002
Published 22 April 2003
Online at stacks.iop.org/JPhysD/36/A24

Abstract
Besides attenuation, x-ray refraction occurs in an x-ray beam passing through an object containing details of different refractive indices or of non-uniform thicknesses. The emerging x-rays present characteristic angular deflections, which are on the microradian scale. Different refraction directions can be resolved by using an angular analyser, for instance a perfect crystal, which can then be revealed by a detector placed behind it. By varying the alignment of the analyser with regard to the incoming x-ray beam, the unrefracted x-rays can either be recorded on a detector or rejected, thus, either contributing or not contributing to the image contrast.

The x-ray refraction contrast can greatly exceed the absorption contrast, for instance in imaging low Z materials, where the potentialities of the technique have been exploited from the beginning. An analyser-based imaging technique has been presented in various versions also assuming different names.

An important variation, based on an algorithm that differentiates the separation of the refraction contrast from the absorption contrast in an image, has been introduced with the name ‘diffraction enhanced imaging’ (DEI).

The interest in the potential medical application of the diffraction imaging technique has given origin to many projects utilizing analyser-based refraction imaging. Initiated with conventional x-ray generators, it has been rapidly developed in synchrotron radiation sites thanks to the highly intense collimated beams available that permit fast projection and tomographic imaging. Mammographic in vitro protocols and cartilage characterisation are the most exploited applications of DEI. The potential medical application has refocused the interest towards the utilization of presently available conventional high-power x-ray generators that can open the doors to large-scale utilization. The aim of this paper is to present a review of the technique in its theoretical and practical aspects; an abundant bibliography, highlights the most significant and recent results.

1. Introduction
X-ray imaging is utilized for the visualization of internal structures of objects in a large number of fields such as materials science, condensed matter physics, biology and medicine. In absorption imaging, the contrast results from variation in x-ray absorption arising from density differences and from variation in the thickness and composition of the specimen.

The sensitivity of the technique is drastically decreased when the sample consists of low Z elements, for which the differences in the absorption coefficients are very small for hard x-rays [1].

Weakly absorbing objects are often encountered in the life sciences, because organic matter is made mainly of carbon, hydrogen, oxygen and nitrogen.

Besides absorption, an x-ray beam traversing an object picks up information on its refraction properties that can also
be utilized as a source of contrast for displaying the internal properties of the sample.

The behaviour of x-rays as they travel through an object can be described in terms of a complex index of refraction. In the x-ray region, it can be written as \( n = 1 - \delta - i\beta \) where the real part \( \delta \) corresponds to the phase shift due to refraction and the imaginary part \( \beta \) to the absorption.

The real and imaginary parts have very different dependences on the photon energy; in the regime where the photoelectric effect dominates and far from absorption edges, \( \beta \propto E^{-4} \) while \( \delta \propto E^{-2} \). As a consequence, the values of \( \delta \) can be orders of magnitude larger than \( \beta \) terms; for example, the values for nylon (C\(_2\)H\(_4\)) at 25 keV are \( \delta = 3.50 \times 10^{-7} \) and \( \beta = 8.12 \times 10^{-11} \) [1].

X-rays passing through regions of different \( \delta \) values are subjected to phase shifts that correspond to being refracted. These changes, which can originate from the purely geometrical effect of the shape of the object or, for instance, from local homogeneity defects of the object, cannot often be visualized using absorption imaging techniques.

Different techniques have been developed for detecting the phase variations, which can be classified into three categories: deflectometry, interferometry and in-line holography [2]. This paper will focus on the first technique; for an extended discussion of the others we refer the reader to [3–5].

In this method, the x-rays transmitted through a sample are analysed by a perfect crystal; only the x-ray satisfying the Bragg law for the diffraction (2d sin \( \theta = \lambda \)) where d is the crystal d-spacing, \( \theta \) is the grazing angle of incidence to the crystal and \( \lambda \) is the radiation wavelength) can reach the detector and then contribute to the image formation [6].

The angular resolution is provided by the choice of reflection from the crystal, which acts as a spatial filter of the radiation refracted and scattered inside the object. As an example, the full-width at half-maximum of the rocking curve for the reflection 111 of the silicon spars the range 17.6–4.3 \( \mu \)rad for the energy interval 15–60 keV [7].

The implementation of this technique (called a ‘Schlieren’ method by the author) was reported by Förster et al [8] for measuring with micrometric precision the wall thickness of spherical plastic targets for laser nuclear fusion.

Different authors have presented slightly varying versions of the same technique that has been called ‘refraction contrast radiography’ [8], ‘phase dispersion introscopy’ [9] or ‘diffraction imaging’ (DI) [10]. The latter term, which refers only to the use of the analyser crystal, will be utilized in this work.

2. The image formation

DI has been applied to x-rays and to neutrons, in the latter case for obtaining images of the magnetic field gradient in the gap of a permanent magnet and the distribution of magnetic domains in a ferrosilicon crystal plate [6].

For a given sample, the contrast depends on the rocking curve width (the analyser), the crystal composition and quality, the x-ray energy, the x-ray divergence and the diffraction order. When all other parameters have been fixed, the contrast depends on the angular position of the analyser crystal.

The x-rays refracted from the sample deviate from their original direction by a small amount, which is proportional to \( \delta \). If they arrive at the analyser within its angular acceptance they are diffracted with an intensity modulated by the intensity of the reflectivity curve at that point. X-rays coming with angular deviations that are outside this small acceptance cannot be diffracted and do not contribute to the image formation. These missed scattered x-rays, which are always contributing to the blurring of the details in conventional images, appear in the diffraction images as absorbed x-rays, generating the so-called ‘extinction contrast’.

When the analyser is set at the angle corresponding to the peak of the rocking curve, it diffracts the unretracted x-rays with full efficiency. The images appear almost scatter-free, because only the small-angle scattering at angles within the rocking curve width can reach the detector.

When the analyser is set at one of the slopes of its rocking curve, the intensity of the x-ray beam diffracted by the analyser is changed by the refraction in the sample, giving rise to the ‘refraction contrast’.

By moving the analyser far off the Bragg angle, inverse contrast is observed: the analyser rejects the unretracted x-rays whereas the scattered ones are recorded and contribute to the image formation [11]. This case is very interesting with samples producing a very large amount of small-angle scattering (spoon-like tissues, for instance), where a visualization of the details can be achieved [12].

In general, the image formation can be described using either geometrical or wave optics. In geometrical optics, ray tracing of the radiation from the source to the detector can be performed by calculating, point by point, the refraction angle in the sample and by weighting the intensity of the refracted x-rays by the rocking curve of the analyser crystal calculated at the point \( \theta + \delta \theta \), where \( \theta \) is the analyser angular position and \( \delta \theta \) is the refraction angle.

The effect of the finite resolution of the detector and of the finite size of the source can be considered by convoluting the intensity with the point spread function of the detector and with the source distribution of standard deviation \( \Sigma = \sigma d_1 / d_2 \), respectively, where \( \sigma \) is the projection of the source size in the scattering plane, and \( d_1, d_2 \) are the source to sample and sample to detector distances, respectively [13].

For simple objects this approach gives results that are generally in good agreement with the experimental data [14, 15].

The wave optical approach provides more precise results because less stringent assumptions have to be considered [16].

The use of the Takagi–Taupin equations to compute the intensity of the electric field outgoing a crystal is necessary for perfect and flat or cylindrically bent crystals; in these cases the equations even have analytical solutions [17]. Gureyev et al [18] have identified the general limits of utilization of the two approaches by considering the area contrast (the contrast far from edges, where weak phase variation occurs) and the edge contrast. To date, no systematic comparative study of the two approaches, including different optics and sample compositions, with experimental data has been published even though it is considered to be a key issue in the full understanding of the parameters affecting image formation. Work on this subject is being carried out by the author [19].
3. The set-up

A highly stable set-up is required for DI and the optics have to be installed on optical tables or high precision diffractometers. In order to minimize the possible artefacts on the images, the vibrations of the optics have to be kept as low as possible. Angular oscillations of the analyser in the order of 1–5% of the rocking curve width are considered to be acceptable.

From the first experiment of Förster et al., whose set-up had an x-ray tube as source and a pre-monochromator crystal, a linear stage for the sample, an analyser crystal and an x-ray film as detector, the layout of the experiment has not changed dramatically. The use of synchrotron radiation (SR) beams and the possibility of fine-tuning the analyser crystal by utilizing high precision or piezoelectric motors, are probably the most important improvements.

With regard to the analyser crystal, the various research groups have exploited both Bragg and Laue geometries. Bragg set-ups are generally more stable, deliver more intense beams and are necessary for low-energy imaging. In the Laue case, an appropriate choice of the distances allows us to acquire both the absorption and the phase-contrast images at once (figure 1) [20]. Alternatively, the absorption image can be directly registered by detuning the analyser crystal and in this case the analyser acts as an x-ray absorber.

4. Sources

4.1. Conventional sources

The utilization of conventional x-ray sources implies severe practical restrictions to the experiments due to the limited x-ray intensity remaining after the ray passes through the monochromator. For instance, Beliaevskaya et al. [21] reported on a flux on the sample of $10^3$–$10^4$ ph s$^{-1}$ mm$^{-2}$; it involves exposure times of about 30 min even for thin samples and of the order of hours for thicker ones. Such long exposures are often subject to stability problems related to thermal drift of the optics and are not suitable for a routine application of the technique. Only the development of further high-power x-ray generators or the utilization of the recently presented monochromatic sources, based on the inverse Compton scattering principle, could permit the application of the technique outside synchrotrons, on a wide range of samples [22].

In the existing conventional set-ups an asymmetric crystal is often utilized to expand the x-ray beam upstream of the sample and to achieve a quasi-parallel small beam to match the acceptance of the analyser crystal [9]. Furthermore, a slit system is used to reduce the effective source size in the dispersion plane [15].

4.2. SR sources

The availability of SR sources has given a critical impulse to the development of DI.

Besides the highly intense beam furnished in a very large energy range, which permits image acquisition in seconds even in the case of thick samples, the fact that the energy can be easily varied and the high spatial coherence of the source have made possible the development of high-energy, low-dose projection imaging and tomography (see sections 9 and 10).

In SR beamlines, the x-ray beam is normally pre-monochromatized by a monochromator [12, 23, 24] (figure 2). The refraction imaging optics consists of either a single crystal (analyser) or a double crystal (monochromator-analyser). In the latter case, horizontal beam and fixed-exit optics can also be achieved [25], whereas in the first case the position of the detector has to be adjusted to the selected energy. A double analyser system (channel-cut) $(+n-n)$ has also been introduced by Menk et al. [26] by determining a horizontal beam on the detector that is utilized for a combined application of the ultra small-angle scattering (USAXS) technique and DI. Set-ups adapted for investigation in the range 10–60 keV and crystal reflections ranging from 111 to 777 have been developed. An easy way of utilizing high orders of reflection is to select high harmonics from the beam delivered by the monochromator; a practical method, introduced by Zhong et al. [27], consists in inserting a prism between the two crystals forming the pre-monochromator. As the angle by which an x-ray deviates from its original direction, when it passes through a refractive prism, is proportional to the square of the wavelength of the x-ray, a spatial separation of the harmonics occurs. The selection of a specific order of the harmonic order can be obtained by slightly tuning the second crystal of the monochromator.

5. Optics development

Recently, Protopopov et al. [28] proposed a special kind of multilayer mirror to analyse the refracted x-ray beam. For a given energy, the reflectivity curve of this multilayer has a

---

Figure 1. Experimental set-up with conventional source. The x-ray beam is expanded and monochromatized by an asymmetric cut crystal and is analysed by a Laue crystal.

Figure 2. Typical experimental set-up with SR source. The white beam is monochromatized by the beamline monochromator. The DI set-up consists of a monochromator, a linear/rotating stage for the sample, an analyser crystal and a detector.
resonant dip that is few microradians wide. If the multilayer is used as the analyser, it shows a minimum reflectivity for an unrefracted x-ray beam and, therefore, it acts as a perfect crystal with an ‘inverse’ rocking curve. The advantage of this set-up consists in the rejection of the direct beam, with only the pure refraction information being kept. The first multilayer prototype consists of 30 C W^{-1} bilayers on a silicon substrate, with a resonance at 0.8° at a wavelength of 1.5 Å. At this stage of development, the width of the resonance is strongly energy dependent and therefore mirrors have to be adapted for single energy utilization; preliminary results have been shown and new developments on the mirror quality have been announced [28].

6. Image processing

Chapman et al [29], proposed the diffraction enhanced imaging (DEI) method for extracting quantitative information from radiographs.

In DEI two images are acquired, one on each side of the rocking curve of the analyser, and an appropriate algorithm permits the calculation of two new images: the apparent absorption and the refraction maps of the object.

The apparent absorption image features the absorption and extinction contrast, which is due to the rejection of small-angle scattering by the analyser crystal. The refraction image is indeed a map of the component of the refractive angles of the photons passing through the sample in the diffraction plane, which is the only direction where the analyser is sensitive [30].

The apparent absorption image, $I_R$, and the refraction angle image, $\Delta \theta_2$, are calculated by applying on a pixel-by-pixel basis the equations:

$$I_R = \frac{I_R^+ + I_R^-}{2R}, \quad \Delta \theta_2 = \frac{R(\theta_0)}{dR(\theta)/d\theta)}|_{\theta=\theta_0} \left( \frac{I_R^+ - I_R^-}{I_R^+ + I_R^-} \right)$$

where $I_R^+$ and $I_R^-$ are the images taken at symmetric angles with respect to the centre of the curve (high (+) and low (−) angles, respectively), $R(\theta_0)$ is the intensity of the rocking curve at those angular positions and $dR(\theta)/d\theta$ is the absolute value of the derivative. These relations have been derived in geometrical optics by considering the first term in the Taylor’s series expansion for the formula for the contrast.

Recently, new algorithms have been studied for the extraction of quantitative information in cases where the small-angle scattering component is dominant (e.g. in the case of porous materials) or when simple geometrical optics does not truly describe the image formation. A discussion of these methods is included in studies that are in the process of being published [31, 32].

7. Applications

7.1. Mammography

In mammography, the discrimination between a tumour lesion and normal tissues is generally difficult because of the small difference in the linear attenuation coefficient in biological objects. Microcalcifications are also an important indirect sign of breast cancer; their number, size, morphology, location and distribution can refer to the presence of a lesion in their proximities. In optimal circumstances, with conventional x-ray methods, the minimum detectable diameter is 0.1–0.2 mm [33].

The high sensitivity of DI to small variations of the refractive index and the intrinsic capability of the analyser to produce almost scatter-free images are the main motivations for the increasing interest in DI mammography.

Several breast cancer specimens had been examined with the DEI technique at 18 keV using a 333 reflection [34]. Images compared with digital radiographs show, in most cases, a better visualization of spiculation or architectural distortion. Comparative studies of absorption versus diffraction images at comparable doses, using mammographic phantoms, formalin fixed and fresh breast tissues, have been carried out by Arfelli et al [23]; their results confirm superior image visualization of details in diffraction images.

With regard to the detection of microcalcifications, unpublished studies report a 50% increase in the number of visualized microcalcifications in breast samples with the DI technique compared to conventional mammography [35].

In figure 3 the absorption image at 20 keV of a mammographic phantom (a) is compared with the diffraction image (b) acquired at the same energy and utilizing a Si 111 analyser crystal tuned at the half width of the rocking curve. The improvement in the visibility of details is evident.

In another study [14] it has also been shown that at 25 keV, in images of thin nylon fibres (diameter 0.1–0.85 mm), an increase in contrast by an average factor of about 1.5 has been observed in the image recorded at the maximum of the 111 rocking curve with respect to the absorption image. A further improvement by an average factor of 5 has been shown in

![Figure 3. Details of a mammographic phantom containing fibrous structures (upper image) and simulated microcalcifications (lower image) at 20 keV: (a) absorption image, (b) diffraction image recorded at half the maximum of the rocking curve with a Si (111) analyser crystal, courtesy of the SYRMEP group, Elettra, Trieste, Italy.](image-url)
333 images with respect to the 111. Even higher gains have been achieved with the 444 reflection; the contrast gain is smaller for thicker samples.

At 18 keV and using a Si 333 reflection, quantitative evaluation of the apparent absorption image of a mammographic phantom (American College of Radiology, RMI 156, Rad Meas Inc Middleton, WI) has shown a contrast as high as 40%; this contrast level is about 20–30 times higher than that measured in a SR radiograph recorded at the same energy in the absorption regime [29].

7.2. Cartilage studies

Osteo-arthritis is a poorly understood disease that can affect the cartilage and other tissues in the joints of ageing people. Conventional radiography is sensitive only in cases of advanced disease in which there has been a loss of cartilage; structural abnormalities in the early stages of the degenerative process are generally not visualized in radiographs.

Measurements have been performed on human articular cartilage in disarticulated, as well as in intact joints [36]. Gross cartilage defects, even at early stages of development, have been studied at 18 and 30 keV and compared with the absorption technique and show a clear, early visualization of the damage.

8. Computed tomography

X-ray tomography is highly developed for measurements of the absorption distribution in two- and three-dimensional objects. Tomographic imaging permits the resolution of structures of a complex sample that are overlaid in projection imaging.

CT DI has been introduced by Dilmanian et al [37]; they demonstrated the possible simultaneous reconstruction of the attenuation coefficients and refractive indices by applying the DEI algorithms to the sinograms before backprojecting the data for image reconstruction. This method leads to uncertainties of about 20% in the reconstructed values of refractive indices.

A different algorithm, which includes second-order terms of the geometrical optical description of the image formation, has been developed and utilized in Monte Carlo simulation by Pavlov et al [38]; it now needs experimental confirmation.

9. High-energy studies

SR sources permit the delivery of highly intense monochromatic beams in an energy range that may extend up to hundreds of kilo-electronvolts [39].

The utilization of high energies \((E > 30 \text{ keV})\) is necessary in imaging thick samples or high Z materials. At high energies, the absorption contrast is reduced compared to that at low energies, while the refraction contrast is only partially depressed. In fact, as already discussed in earlier paragraph, the \(\delta\) term has an energy dependence proportional to \(1/E\). Conversely, to a first approximation, the rocking curve width for a given crystal reflection decreases linearly with increasing energy [40], and then the sensitivity of the analyser to the variation of the refractive index is only proportional to \(1/E\).

Thus, for a given object, the ratio \(\text{SNR} / \sqrt{D}\), where SNR is the signal to noise ratio and \(D\) is the dose delivered to the sample, should give higher values in diffraction compared to absorption imaging; this means that for a given SNR lower dose imaging is possible, and conversely, for a given dose, higher SNR can be obtained.

High-contrast mammographic images have been obtained at energies above 30 keV without a significant loss of contrast. In particular, images at 50 keV have been recorded with a significantly reduced dose compared with conventional techniques [25].

10. Combination of different techniques

X-ray scattering and imaging are complementary techniques, which can access the inner structures of a sample. In particular, USAXS can provide quantitative information on microstructures in the range from a few nanometres to some dozens of microns by also giving significant volume-averaged information of these structures. As in the case of DI, USAXS images are formed from angle-filtered rays that are scattered by the electron density variations in the sample. Information on the shapes and sizes of the objects are achievable by comparing images produced at different scattering vectors.

A combination of USAXS and DI has been initiated by Menk et al [26] by utilizing a double crystal analyser that permits the application of both techniques without moving the sample. By tuning the angle of the analyser crystal, it is possible to choose a scattering angle that corresponds to a desired object size, and to use for imaging only those x-rays that were deviated under this angle.

11. Conclusion and outlook

Several groups, worldwide, are now using the DI technique; most of the studies are conducted at SR beamlines where highly intense monochromatic x-ray beams are available. The applications have been restricted until now to biology and medicine, whereas the exploitation in other fields, such as materials science, for instance, has not yet been documented. The high sensitivity of the technique to small variations of the refraction index, even in thick materials, may be used, for example, in the study of composite materials, in the identification of strain distribution or for distinguishing between objects of the same composition, but whose scattering properties are different, based on structural order.

The interest in the technique is also proved by the existence of several patents concerning different aspects of the techniques [41–44] demonstrating the existence of interest that is other than scientific in the utilization of the technique.

A critical input for the utilization, on a large scale, of the refraction contrast imaging technique could be obtained from the availability of intense, monochromatic, non-synchrotron, radiation sources [22, 45].

Acknowledgments

I thank Dr P Coan for her support in the preparation of the manuscript. This work is supported by the EU grant contract HPRI-CT-1999-50008.
References

[22] Carroll F E 2002 Am. J. Roent. 179 583–90
[31] Pagot E et al unpublished
[34] Pisano E et al 2000 Radiol. 214 895–901