REVIEW LECTURE

Ten years of X-ray interferometry

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[Plates 1–4]

X-ray interferometers were the first optical instruments which utilized many Bragg reflecting components in monolithic blocks of perfect crystal. They have made important contributions to our knowledge of fundamental constants, of Bragg reflection X-ray optics, of strains and defects in crystals and of the optical constants of materials in the X-ray region of the electromagnetic spectrum. Based on an oversimplified optical analogue, their mode of operation is described in detail. Current applications of crystal interferometers and possible future work is briefly reviewed.

INTRODUCTION

Ten years is a reasonable time for a scientific curiosity to be transformed into a useful instrument, and that is how long it has taken for X-ray interferometers. In this review I shall concentrate attention, not on the detailed description of the theoretical background, but on the practical application that has followed the invention of single-crystal Bragg reflexion interferometers (Bonse & Hart 1965a). They find applications whenever the wavelength (or de Broglie wavelength) of radiation lies inside the limiting sphere of whatever perfect crystals happen to be available. All of the optical instruments which have so far been constructed use single crystals of silicon as the starting material, usually for no reason other than cost and ease of availability, suitable material is sold on the open market at about £1 cm⁻³, and this price, like that of the integrated solid-state circuits for which the silicon is grown, has not increased during the time-scale of this review. X-ray interferometers may also be used to investigate defects in crystals so that the interferometer itself becomes the object of investigation. For this purpose X-ray interferometers have been constructed from silicon, germanium and quartz. Doubtless many other materials could also be studied in this way.

It is not possible to appreciate the value of an instrument without at least a rudimentary impression of the principles governing its operation. Fortunately an analogue which can be very useful in describing X-ray interferometers does exist and it will be developed in the first section of this review.

The most obvious application has been to the establishment of the absolute
length scale for submultiples of the wavelength of light without recourse to X-ray spectral line standard. In spite of a considerable effort by several laboratories this project is still tantalizingly close to fruition with only small systematic errors awaiting eradication. The result will have immediate applications in determining the Avogadro number and other fundamental constants, in establishing the absolute scale of \( \gamma \)-ray wavelengths and, perhaps in the realization of an atomic definition of mass which will replace the existing alloy artefact standard.

In studies of defects in crystals some particularly simple X-ray interferometers have been used to investigate irradiation damage and its subsequent annealing and to determine the configurations of crystal dislocations. As a result of recent improvements in the experimental methods, the X-ray optical constants of materials at wavelengths near their atomic absorption edges have been measured with an adequate precision to be theoretically interesting. This work has been done with conventional X-ray sources and with synchrotron X-ray sources.

Finally we shall be concerned with X-ray interferometric spectroscopy where dispersive interferometers might be used. Little progress has so far been made because high intensity tunable X-ray sources have not been available. This situation will change as more synchrotrons and electron storage rings are adapted to become useful and potent sources of X-radiation.

Two review articles have been written, one by Bonse (1969) and the other by Hart (1971).

**AN OPTICAL ANALOGUE**

Figure 1 is a representation of a single perfect crystal X-ray interferometer showing the generation of many waves from the one which is incident on the first crystal \( S \) at the Bragg angle. At each Bragg reflexion the incident wave generates a wave travelling at an angle \( 2\theta \) to the forward direction. Bragg's law must be satisfied so that

\[
K_0 + h = K_h, \quad |K_0| = |K_h| = \lambda^{-1}, \quad |h| = d^{-1}
\]  

or

\[
2d \sin \theta = \lambda.
\]

\( K_0 \) and \( K_h \) are the wavevectors of the two waves, \( h \) is the reciprocal lattice vector for the Bragg planes whose spacing is \( d \) and \( \lambda \) is the X-ray wavelength. The relative amplitudes and phases of the two waves in a particular case are determined by the requirements of Maxwell's wave equation and by the application of Snell's law of refraction at the crystal surfaces. As we shall not be concerned with a detailed discussion of the Bragg intensities, it will suffice for us to note that two kinds of solution exist. The electric displacement ratio \( D_h/D_0 \) may be either positive or negative. These two solutions correspond (in simple crystal structures) to the two cases in which either the maxima or the minima of the standing wave pattern lie on the planes of atoms. In the general case, linear superpositions of the two solutions are obtained so that the position of the standing wave pattern with respect to the planes of atoms will depend on the wave amplitudes. Let us confine our attention exclusively to the antiphase
FIGURE 1. Schematic plane-wave diagram for a monolithic crystal interferometer. The short vertical lines represent planes of atoms spaced $d$ apart and all three wafers $S$, $M$ and $A$ are in perfect alignment. Moiré patterns are formed in the exit beams by the overlap of the standing wave pattern $P$ with the analyser crystal $A$. 
solution when the minima of electric displacement (black in figure 1) lie on the planes of atoms. This is the only relevant solution in the case of crystals which show a strong anomalous transmission effect. The optical analogue which is developed below gives reliable predictions for that case. When only weakly absorbing crystals are used the X-ray interferometers still form moiré fringes but the mechanism of destructive interference involves amplitude switching from one wavefield type to the other rather than absorption. Bragg reflecting crystals act as phase gratings rather than as amplitude gradings.

If the gaps between S and M and between M and A are precisely equal, then, as figure 1 shows, a standing wave pattern P is projected onto the final wafer A. With a suitable X-ray sensitive detector one would see unlocalized X-ray fringes with a spacing d in this position. Since d is approximately 1 Å\(^+\) we look instead for a way to magnify the fringe spacing and this is the function of the analyser crystal A. Just as the scattering power for a perfect crystal is periodic in position so too is the absorption cross-section. Both periodicities are described by the same reciprocal vector \( h \). Large-scale X-ray moiré patterns are therefore observed when the detector views the standing wave pattern through the analyser crystal. Let the grid P in figure 2 represent the X-ray standing wave pattern P and let the black bars of grids A1, A2 represent the absorbing regions of crystal A. If the whole crystal (that is S, M and A) is ideally perfect, figure 1 shows that the intensity maxima of P (light) are not aligned with the planes of absorption and that a high intensity is obtained below the analyser crystal. This corresponds to the superposition of P and A1 in figure 2; the observed moiré field is bright. Suppose now that we contrive to move A through a distance \( \frac{1}{4}d \) parallel to \( h \). As figure 2 shows, the moiré field is now dark in the region where P and A2 are superimposed. There are no output beams from the X-ray interferometer.

Other deformations of the analyser crystal A are possible. Let us consider some of them in optical analogues.

With out significantly changing the spacing of atoms in the crystal we might rotate the analyser A3 through an angle \( \phi \) about an axis normal to its surface. Horizontal fringes are observed in the moiré field as figure 2 shows. Their spacing \( A_R \) is given by

\[
A_R = d\phi^{-1}.
\]  

If the analyser A4 has Bragg planes which are precisely parallel to but different in spacing from the standing wave pattern P then vertical fringes appear as in figure 2. The moiré fringe spacing \( A_D \) is inversely proportional to the difference in spacing \( \Delta d \) between the standing wave pattern at P and the Bragg planes of the analyser crystal A4, \( \Delta d = d_A - d_P \) and

\[
A_D = d(\Delta d/d)^{-1}.
\]

When both deformations are present sloping moiré fringes are observed. From the sign of the slope of the moiré fringes the relative signs of \( \Delta d \) and \( \phi \) may be deduced.

\[\dagger\] 1 Å = 10\(^{-10}\) m = 0.1 nm.
Figure 2. Moiré patterns formed between the standing wave pattern P and various analyser crystals. A1, A2, A3 have the same spacing as P. A4, A5, A6 have a larger spacing so that $\Delta d = d_A - d_P > 0$. 
as figure 2 shows in the overlap of A5 and A6 with P. When Δd is positive a rotation of A results in a moiré pattern rotation of the same sign.

These simple fringe patterns were quantitatively investigated by Bonse & Hart (1966). They elastically strained the analyser mount of a single crystal interferometer so as to produce rotational moiré patterns. By heating the analyser crystal they were able to alter its lattice parameter so as to produce dilational moiré patterns.

Although we have been concerned with changes in the analyser crystal, the moiré patterns are equally sensitive to changes in the standing wavefield P. Such changes may be caused by variations in position, orientation or spacing in any of the other elements of the interferometer. More important, the standing wave pattern can be altered by phase shifting objects placed in one of the interfering beams. A half-wave plate will advance the phase of a beam by π and will therefore cause the standing wave pattern to move sideways through a distance Δd. A wedge placed with its edge horizontal will cause the standing wave pattern to rotate so that a set of horizontal moiré fringes will be observed. A vertical set of moiré fringes is obtained when the edge of the wedge is vertical. In these experiments a detailed analysis is complicated because the wedge alters both the phase and the direction of the X-ray beams. But in each case, to a good approximation, the moiré fringes delinate loci of constant optical path. Thus, X-ray interferometers can be used to map both crystal deformations and the distribution of electrons in samples.

STUDIES OF DEFECTS IN CRYSTALS

Homogeneous elastic strains such as pure rotation and dilations were studied by Bonse & Hart (1966). Those experiments served to verify quantitatively the reliability of the optical analogue. At the same time crystal dislocations were found in one silicon crystal from which an interferometer had been cut. The systematic observation of the images in moiré patterns obtained with many different Bragg reflexions (Hart 1972) enables one to determine the Burgers vector of the crystal dislocation in both magnitude and direction.

Since the elastic theory for dislocations in isotropic media is well known, we can use that theory to prepare optical analogues which can be compared directly with X-ray moiré patterns. Figure 3 shows some predicted patterns in which we assume that the standing wave pattern P is perfectly periodic and that the analyser A contains a single edge dislocation. In each case the number of extra half fringes in the moiré pattern is given by

\[ N = |b \cdot h|, \]

where \( b \) is the dislocation Burgers vector. The position of the extra moiré fringe is determined by the signs of Δd and φ. In each case the extra half plane of atoms lies above the crystal dislocation which is marked with the conventional symbol \( \perp \). When Δd < 0 (figure 3a) the extra moiré fringe appears on the same side of the dislocation as the extra half plane in the crystal. Reversing the sign of Δd inverts the
Figure 3. Moiré patterns formed with a perfect standing wavefield and an imperfect analyser. The symbol ⊥ shows the position of the extra half-plane of atoms $(b, n = 1$ so that only one extra half-fringe appears). (a) $\Delta d = d_A - d_2 < 0$; (b) $\Delta d > 0$; (c) $\phi > 0$; (d) $\phi < 0$; (e) $\phi < 0$, $\Delta d < 0$; (f) $\phi < 0$, $\Delta d > 0$; (g) $\phi > 0$, $\Delta d > 0$; (h) $\phi > 0$, $\Delta d < 0$. The extra half line in the analyser grid can be seen most easily by viewing at a small angle to the plane of the figure.
moiré dislocation (figure 3b). Because $|b| = d$ there is only one extra moiré fringe (equation (5)). Dilational X-ray moirés with $N = 2$ were obtained in the early experiments by Bonse & Hart (1966).

In practice, rotational moiré patterns are more conveniently produced than are dilational moiré patterns. For $\phi > 0$, corresponding to a counterclockwise rotation of the standing wave pattern $\mathbf{P}$ when looking towards the X-ray source, Figure 3c shows the extra moiré fringe appears on the right-hand side of the crystal dislocation. When the rotation is reversed the extra moiré fringes appear to the left of the crystal dislocation (figure 3d). When both rotations and dilations occur simultaneously the extra moiré fringes can appear in any one of the four quadrants (figures 3e–f). Figure 4, plate 1, shows a series of moiré images of one crystal dislocation (Hart 1972). The pictures were all obtained with $\phi > 0$. The Burgers vector $\mathbf{b} = (u, v, w)$ can be determined from the X-ray moiré patterns by direct substitution in equation (5) of the appropriate values of $N$ and $\mathbf{h}$

$$2u - 2w = \pm 1,$$
$$-2u + 2v = \mp 2,$$
$$-4u + 2v + 2w = \mp 3.$$ 

Thus $\mathbf{b} = (\pm \frac{1}{2}, \mp \frac{1}{2}, 0)$. The sign ambiguity refers to the fact that we have not yet established a sign convention. Comparisons between the X-ray moiré patterns and the optical analogues show that the extra half-plane of atoms lies below the crystal dislocation. If we define the direction of the dislocation line as $L = [101]$, directed towards the viewer, then, using the FS/RH perfect crystal convention (Hirsch et al. (1955)), we find that $\mathbf{b} = \frac{1}{3}[1\bar{1}0]$.

It is not necessary that the defects should be grown into the crystal. When two dissimilar materials are joined together it is well known that mutual strains occur when the temperature changes. Such strains can be imaged in an X-ray moiré pattern (figure 5, plate 2). Since the method is non-destructive, it may be used to follow the deformation resulting from, for example, various heat treatments.

In practice it is often either inconvenient or impossible to obtain large crystals from which X-ray interferometers may be cut. Since, in these experiments, the function of the mirror $M$ is merely to make optical contact between the beam splitter $S$ and the analyser $A$, it is convenient to bring these two members into physical contact so that the mirror is unnecessary. This approach has been developed by Lang and Muncey (1965), Bradler & Lang (1968) and by Lang (1968) in studies of dislocations and fault boundaries in quartz and silicon crystals. By irradiation with charged particles a crystal may be transformed into two coherent parts separated by a damaged layer. Such bicrystals also produce Bragg reflexion moiré patterns which have been used to study the properties of the damaged layer by Authier & Montenay-Garestier (1965), Bonse, Hart & Schwuttke (1969) and by Schwuttke & Brack (1973). The first moiré patterns of this type were produced by the coherent overgrowth of impure cadmium sulphide on a purer substrate (Chikawa 1965).
Figure 4. X-ray moiré patterns of a dislocation in silicon obtained with Mo Kα radiation. Field approximately 1.4 mm x 1.4 mm: (a) 202 Bragg reflection; (b) 220 Bragg reflection; (c) 422 Bragg reflection; (d) 440 Bragg reflection.
Figure 5. X-ray moiré pattern of an aluminium disk (5 mm diameter) evaporated onto the analyser wafer of a silicon interferometer, (a) as deposited, (b) after a short period of annealing at 500 °C (courtesy J. Hellier).
Figure 8. X-ray phase-contrast micrograph (a) of a thin slice of granite. For comparison (b) is an optical micrograph. Area approximately 2 mm x 2 mm. From Ando & Hosoya (1972).
Figure 11. Experimental dispersion curves for nickel near its absorption edge obtained with synchrotron radiation (Braman & Matherly 1974). The region A is covered with nickel foil, the region B is not.
The scale of X-ray moiré patterns is not altered when the X-ray wavelength is changed. For this reason it was first suggested by Bonse & Hart (1965a) that Bragg spacings could be measured without the need to know the X-ray wavelength. The influence of the X-ray linewidth, which introduces the dominant error in the conventional method, can therefore be completely eliminated when Bragg spacings are measured with X-ray interferometers.

Suppose that the analyser crystal A in figure 2 is moved parallel to the reciprocal vector \( h \). The field becomes light, as in A1, and then dark, as in A2, periodically for each increment \( \frac{1}{2} d \) in the distance moved. A value of the Bragg spacing \( d \) is obtained by counting the number of X-ray fringes \( (p + \varepsilon) \) per unit distance. It is convenient to represent the X-ray fringe order as the sum of an integer part \( p \) and a fraction \( \varepsilon \). Distance is measured with an optical interferometer using light from the standard lamp whose radiation defines the International Standard of Length \( \lambda_0 \). Then, if one optical fringe represents a translation of \( \frac{1}{2} \lambda_0 \),

\[
d = \frac{1}{2} \lambda_0 (p + \varepsilon).
\]  

Since the optical standard is a defined quantity and since crystals up to 1 m or so in length can be obtained (containing approximately \( 10^{10} \) Bragg planes if \( d = 1 \) Å) very precise measurements of \( d \) should be possible. In this experiment it is crucially important that both the X-ray interferometer and the optical interferometer should measure the same baseline. As sufficient care has not always been taken in this aspect of the experimental design it is useful for us to examine briefly the necessary criteria and to see how they might be met.

Let us choose right-handed Cartesian axes with \( x \) parallel to \( h \), \( y \) parallel to \( K_0 \wedge K_h \) and the origin \( O \) at the centre of the standing wave pattern on the entrance surface of the analyser crystal. In this context it is not necessary for us to provide a more precise definition of the centre of the standing wave pattern. The X-ray interferometer measures distance with respect to the origin. On the other hand, the measure of distance made by the optical interferometer is referred to the centre of the optical mirror at \((x_0, y_0, z_0)\). It is impossible for these two reference points to coincide. But what separation is tolerable in practice?

Without difficulty it is possible to arrange that both the optical beams and the X-ray beams lie within (say) 10 μm of the horizontal \( xz \) plane. If, during the translation \( X \), a parasitic rotation \( \phi \) occurs about the \( z \) axis, then a systematic error is introduced. The X-ray interferometer records \( X/d \) fringes while the optical interferometer records \( \varepsilon(X + y_0 \phi)/\lambda_0 \) fringes. The fractional error is \( y_0 \phi/X_0 \). If the fringes are recorded by an electronic photon counter such as a scintillation counter then the appearance of rotational moiré fringes in the X-ray beam results in a loss of fringe contrast. The contrast change is negligible if the rotational moiré fringe spacing is very much greater than the height of the X-ray beam. Rotations of \( 10^{-8} \) rad are easily detected in practice (\( \Delta R = 2 \) cm) so that the error on a 100 μm traverse length is only one part in \( 10^9 \).
It is not easy to make \( x_0 \) less than 1 cm. If the optical mirror is ground onto the silicon crystal then \( x_0 \) changes by \( 2.56 \times 10^{-6} \) cm K\(^{-1} \) when the temperature changes and by less than \( 10^{-7} \) cm when the air pressure changes during a deep depression. If, for example, glass optical mirrors are glued onto the silicon X-ray interferometer then the strains induced by environmental changes will be even larger.

By far the most important design compromise is the one necessary when the optical interferometer is used in transmission, for \( z_0 \) cannot then be zero. Suppose that there is a parasitic rotation \( \delta \theta \) about the \( y \) axis when the translation stage moves through its range \( X \): The fractional error is 15 parts in \( 10^6 \) for a traverse length of 100 \( \mu \)m if \( z_0 = 3 \) mm and \( \delta \theta = 0.1^\circ \). This particular systematic error can be entirely avoided when the optical interferometer is used in the back-reflexion mode because \( z_0 \) may be adjusted to zero.

**Experimental details**

A first step towards absolute determinations of Bragg spacing was taken by Bonse & te Kaat (1968) who made an X-ray interferometer in two parts which were reassembled on a translation device. This was followed by a monolithic X-ray interferometer which incorporated an elastic spring strip translation device as part of the same silicon crystal (Hart 1968). Optical interferometers have been attached to both of these devices (Bonse, te Kaat & Spicker 1971; Curtis, Morgan, Hart & Milne 1971). A third system has been developed at the National Bureau of Standards (Deslattes 1969; Deslattes & Henins 1973).

Figure 6 shows, in outline, the details of the experimental arrangements used in the National Bureau of Standards experiment and in the joint Bristol/National Physical Laboratory experiment. The experiments started by Bonse and his coworkers are continuing in a collaboration with the Physikalisch Technisches Bundesanstalt. In all of these experiments the translation stage depends on the elastic deformation of spring elements. Deslattes’s (1969) design (figure 6a) was based on a scanning stage constructed by Blayney (unpublished). It is in essence a pinjointed structure in which the joints are the flexure regions left between pairs of closely spaced parallel holes. The scanning stage consists of a pair of platforms constrained, by means of two struts \( S \) and four flexure regions \( F \), to have only the parallel relative motion which results from shearing of the parallelogram that is formed by the two struts and the two platforms. Hart’s (1968) scanning stage is a folded version of the well-known spring strip elastic traverse (Jones 1951) but it is constructed from a monolithic block of silicon so as to avoid many of the problems which arise in polylithic elastic systems. In both systems the relative orientations of the two parts of the X-ray interferometer can be adjusted by elastic deformations of the supports.

In the National Bureau of Standards experiment (figures 6a, b and Deslattes & Henins 1973) the optical interferometer is a plano-concave Fabry–Parot cavity working with a mean order number of 1000 and a fineness of more than 1000 so that the optical fringes are very sharp. The full width at half intensity of the cavity
Figure 6. X-ray and optical interferometers used to measure absolute lattice spacings in silicon crystals. Appropriate detectors are marked D. In (a) a two-component X-ray interferometer is mounted on a brass scanning system, described in the text. The offset z₀ between the X-ray and optical beams is shown in (b). The whole system is a monolithic silicon crystal in (c). Beam steering with polarizing optical components is shown in the plan (d) of the interferometers. Scale marks approximately 1 cm long.
response function is about 3 Å. Because the system works in transmission, there is an offset between the optical and X-ray interferometers of \( z_0 \approx 3 \text{ mm} \). As we found in the previous section, this imposes severe demands on the quality of the translation stage. In contrast to this, the optical interferometer in the Bristol/National Physical Laboratory experiment works in back reflection from a silicon mirror (figures 6c, d) and \( z_0 \) is adjusted to zero with a precision of better than 10 µm. Because these are elastic systems, body rotations may occur when forces are applied to the translation stage. For this reason the optical interferometer employs four reflections from the silicon X-ray interferometer to ensure that its output is unchanged when body rotations of the silicon occur. Beam steering is accomplished by polarization sensitive reflectors as indicated in the ray diagram. By employing electro-optic phase modulation and synchronous detection this interferometer is capable of detecting relative motions of a few hundredths of an ångström.

Figure 7 shows a typical record in which the ordinate is proportional to the instantaneous X-ray counting rate and the abscissa is the output of the optical interferometer. The time constant is 1 s in each channel and the noise level is a few hundredths of an ångström. From two such records obtained near successive zeros of the optical signal the ratio \( d/\lambda_0 \) is determined to approximately 10 p.p.m. (parts per million). This precision is comparable with the best results obtained by the
traditional method. Over ranges of several hundred optical fringes the precision of measurement improves by the number of optical fringes traversed.

Experimental results

Deslattes & Henins (1973) claimed to be able to measure the lattice parameter of silicon with a standard error of 0.05 p.p.m. though that error was increased to 0.25 p.p.m. (Deslattes et al. 1974) when the influence of drift was included. The Bristol/National Physical Laboratory experiment is still in progress but one advantage of monolithic construction is evident in that the measured relative drift between the X-ray and optical interferometer is less than 0.1 Å/d.

Let us attempt to estimate the systematic errors which will result when the translation stage is imperfect. When δθ is not zero the mean intensity of the X-ray fringe pattern is altered because the analyser crystal is no longer set at the Bragg angle. For the 220 Bragg reflexion of molybdenum Kc 1 radiation with 0.5 mm thick crystals, both experiment and theory show that an intensity change of 1% corresponds to a rotation δθ of 8 × 10⁻⁷ rad. An intensity change of 0.1% results from a rotation of 2.5 × 10⁻⁷ rad. Under typical conditions it is feasible, with a 10 s time constant, to detect a 1% change in the X-ray intensity. There is therefore a systematic error of up to 24 p.p.m. in the National Bureau of Standards experiment for a 100 μm traverse length. R. D. Deslattes (1974, private communication) expects from design computations that δθ < 5 × 10⁻⁷ rad for his translation stage, though he has not measured its performance. Deslattes & Henins (1973) state that their system is ‘capable of traversing more than 100 optical fringes without any noticeable loss of X-ray fringe contrast’. If we assume, on the basis of this statement, that X = 36.7 μm and that δθ = 8 × 10⁻⁷ rad, then their systematic error becomes 76 p.p.m. Even with the optimistic estimate that δθ = 5 × 10⁻⁸ rad, the potential systematic error is 4.6 p.p.m. and it would be extremely difficult to check the traverse performance to that precision. To make this error less than the standard error which would be achieved under drift-free conditions one would have to demonstrate that δθ < 10⁻¹⁰ rad! Or, of course, one could make the offset 20 equal to zero. In the monolithic system with z₀ < 10 μm and δθ < 8 × 10⁻⁷ rad, the systematic error is only 0.08 p.p.m. for a 100 μm traverse length.

As we saw in the previous section, parasitic rotations about the z-axis need not lead to systematic errors of more than 0.001 p.p.m. in the apparatus depicted in figure 6.

The measurement of the lattice spacing of silicon made by Deslattes & Henins (1973) has been incorporated into a new determination of Avogadro’s number (Deslattes et al. 1974). The previously unnoticed systematic error propagates into one three times larger in the Avogadro number.

It is quite clear that, although the absolute lattice parameters of silicon has not yet been measured with errors which are demonstrably lower than those claimed for conventional methods, the sensitivity and scale of the existing measuring machines is such that a measurement of δ to 0.1 p.p.m. is in sight. The calibration
of the lattice parameter of a crystal is the key link in several measurement chains. A calibrated crystal would be essential for improved measurements of \( \hbar / \epsilon \) from the high energy limit of the Bremsstrahlung radiation, of \( \hbar / \epsilon c \) and the fine structure constant from electron–positron annihilation experiments and of \( \hbar c / M \) for thermal neutrons. The calibration of the nuclear \( \gamma \)-ray wavelength scale is also dependent on the precision measurement of a crystal lattice spacing.

On rather a longer time-scale, it is worth noting that when the precision with which Avogadro’s number is measured approaches 0.01 p.p.m., it is possible to use an atomic standard of mass in place of the artefact standard kilogram.

**X-ray Phase Contrast Microscopy**

Objects placed in the interfering beams of an X-ray interferometer can be imagined on film with spatial resolution almost as high as that obtained in the visible part of the electromagnetic spectrum. The refractive index \( n \) is calculable from (James 1948)

\[
\frac{n = 1 - x - i \beta}{1 - \delta} = 1 - \frac{e^2 \lambda^2}{2 \pi \hbar c^2} \sum N_i (Z_i + f'_{i} + i f''_{i}),
\]

(7)

where \( N_i \) is the number of atoms per unit volume with atomic number \( Z_i, f'_{i} \) and \( f''_{i} \) are the real and imaginary dispersion corrections at the wavelength \( \lambda \) and \( e, \mu \) and

**Table 1. Real Parts of the Refractive Index Decrements, \( 10^6 \alpha \) for Various Crystalline Materials**

<table>
<thead>
<tr>
<th>Material</th>
<th>( \alpha_0 / \text{Å} )</th>
<th>W K( \alpha_1 )</th>
<th>Ag K( \alpha_1 )</th>
<th>Mo K( \alpha_1 )</th>
<th>Cu K( \alpha_1 )</th>
<th>Cr K( \alpha_1 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>C</td>
<td>3.5688</td>
<td>0.2073</td>
<td>1.485</td>
<td>2.389</td>
<td>11.29</td>
<td>25.02</td>
</tr>
<tr>
<td>Si</td>
<td>5.4307</td>
<td>0.1369</td>
<td>0.965</td>
<td>1.586</td>
<td>7.562</td>
<td>16.81</td>
</tr>
<tr>
<td>Ge</td>
<td>5.6774</td>
<td>0.2783</td>
<td>2.000</td>
<td>3.144</td>
<td>14.44</td>
<td>31.89</td>
</tr>
<tr>
<td>LiF</td>
<td>4.0236</td>
<td>0.1444</td>
<td>1.025</td>
<td>1.666</td>
<td>7.890</td>
<td>17.51</td>
</tr>
<tr>
<td>NaCl</td>
<td>5.6388</td>
<td>0.1224</td>
<td>0.881</td>
<td>1.418</td>
<td>6.728</td>
<td>14.94</td>
</tr>
</tbody>
</table>

**Table 2. Values of \( t_x / \mu \text{m} \) for the Crystalline Materials Listed in Table 1**

<table>
<thead>
<tr>
<th>Material</th>
<th>W K( \alpha_1 )</th>
<th>Ag K( \alpha_1 )</th>
<th>Mo K( \alpha_1 )</th>
<th>Cu K( \alpha_1 )</th>
<th>Cr K( \alpha_1 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>C</td>
<td>100.8</td>
<td>37.67</td>
<td>29.69</td>
<td>13.64</td>
<td>9.152</td>
</tr>
<tr>
<td>Si</td>
<td>152.7</td>
<td>56.56</td>
<td>44.72</td>
<td>20.37</td>
<td>13.62</td>
</tr>
<tr>
<td>Ge</td>
<td>75.10</td>
<td>27.97</td>
<td>22.21</td>
<td>10.67</td>
<td>7.179</td>
</tr>
<tr>
<td>LiF</td>
<td>144.7</td>
<td>54.05</td>
<td>42.38</td>
<td>19.53</td>
<td>13.08</td>
</tr>
<tr>
<td>NaCl</td>
<td>170.8</td>
<td>63.53</td>
<td>50.02</td>
<td>22.90</td>
<td>15.33</td>
</tr>
</tbody>
</table>

c are fundamental constants. The real parts \( \alpha \) of the refractive index decrements \( \delta \) are listed for common crystals in Table 1. Since the refractive indices are so close to unity phase plates are quite thick. The material thicknesses \( t_\alpha \) which cause a phase advance of \( 2\pi \) are calculated in Table 2. There is a simple reason why \( t_\alpha \) appears to be almost a constant for all of the materials chosen. It is that \( N \) \( Z \) is proportional to the density because \( Z / \Lambda \) is approximately a constant for all atoms. The different
crystals in the tables cover only a small range of density. To quite a high degree of approximation the X-ray optical thickness of a material is proportional to its density. On the other hand, apart from the influence of absorption edges, the mass absorption coefficient \( \mu \) varies with \( Z^3 \lambda^3 \) so that X-ray absorption microscopy is a sensitive way of mapping atomic number variations in a sample. Absorption contrast is too small to be useful for light elements at short X-ray wavelengths and it is in this range that X-ray phase contrast microscopy might be an important experimental technique.

Specimens will of course be imaged with both absorption contrast and with X-ray phase contrast in the same photograph. However, the sign of the phase contrast can be inverted by moving the analyser crystal parallel to \( h \) through a distance \( \frac{1}{2}d \). If the contrast is to be linear with phase shift, then two photographs should be taken, one with the analyser crystal displaced by \( \frac{1}{4}d \) and the other with a displacement of \( -\frac{1}{4}d \) from the positions of the extrema depicted in the optical analogues (figure 2, positions A.1 and A.2). Provided that the phase variations are small, or provided that the X-ray interferometer is precisely set at the \( \pm \frac{1}{4}d \) positions, the mean intensity map shows only the X-ray absorption contrast. Pure phase contrast is obtained in the intensity difference map.

For example, Ando \& Hosoya (1972) showed that different patterns were obtained by X-ray phase contrast and by optical microscopy of specimens of biological and mineralogical interest. Figure 8a, plate 3, shows one of their X-ray phase-contrast photographs of a 180 \( \mu \)m thick slice of granite obtained with molybdenum K\( \alpha \) radiation. The spatial resolution obtained is not very much worse than the resolution of the optical micrograph (figure 8b) which was taken for comparison with the X-ray phase contrast picture. In the optical micrograph the black areas are biotite and the other grey parts contain feldspar and quartz. Suppose we may assume, for approximate calculations, that both the phase shift and X-ray linear absorption coefficient are proportional to the crystal density. Using molybdenum K\( \alpha \) radiation we find that the linear absorption coefficient \( \mu \approx 5.5 \rho \) cm\(^{-1} \) and that \( t \approx 0.0016 \rho \) cm. For 180 \( \mu \)m of quartz the attenuation factor \( \exp (-\mu t) \) is 0.77 and the phase advance is \( 2\pi(p+\varepsilon) = 2\pi \times 4.25 \). The absorption contrast is given by

\[
\frac{\delta I_A}{I} = -\mu t \frac{\delta \rho}{\rho} = -0.77 \frac{\delta \rho}{\rho},
\]

where \( \delta I_A/I \) is the fractional change of intensity caused by a fractional density change \( \delta \rho/\rho \). On the other hand, moiré fringes of the form \( I = I_0 [\cos 2\pi(p+\varepsilon) + 1] \) allow a phase contrast \( \delta I_p/I \) of

\[
\frac{\delta I_p}{I} = \pm (p+\varepsilon) \frac{\delta \rho}{\rho} = \pm \frac{t \delta \rho}{t \rho} = \pm 4.25 \frac{\delta \rho}{\rho}.
\]

Under optimum conditions the phase contrast is therefore five times higher than the absorption contrast in this case. Unfortunately, the increase in density of biotite over feldspar and quartz arises at least in part from iron and magnesium. The mass
absorption varies rapidly with atomic number and is approximately ten times higher for iron than for silicon at the wavelength of molybdenum Kα. As the biotite is imaged with less intensity than the matrix it is not clear in only one X-ray moiré photograph how much of the contrast is attributable to phase shifting and how much is due to absorption. However, M. Ando & S. Hosoya (1975, private communication) also took X-ray absorption photographs of the granite sample. They detected no contrast in the biotite rich region and were therefore sure that the image obtained is an X-ray phase contrast image. At short wavelengths and for low atomic number materials the phase contrast becomes of more import than the absorption contrast.

ATOMIC SCATTERING FACTORS

Especially in the spectral regions close to the characteristic absorption edges of a substance, the variations of its atomic scattering factors are theoretically interesting. Measurements of the refractive index are particularly challenging because in the interesting spectra region the absorption is inevitably high. For \( \mu t \approx 1 \) reasonable moiré fringe contrast is still obtained but for most solids near the K absorption edges of their constituent atoms the phase shifts are only about 10\( \pi \), (\( p + c \approx 5 \)). Even so, it is essential that \( \alpha \) should be measured to better than 0.05 % so that \( f' \) (equation (7)) can be determined to within a small per centage.

At characteristic wavelengths a number of results have been obtained using fairly obvious modifications of traditional optical techniques. The X-ray moiré fringes were recorded on film (Bonse & Hart 1965a,b; Kato & Tanemura 1967; Bonse & Hellkötter 1969; Creagh & Hart 1970; Bonse & Materlik 1972). Since there are few suitable characteristic wavelengths near the absorption edge of any particular material and since the exposure times were several hours, these approaches will probably be of little use in future measurements. A free choice of the operating wavelength is essential in these experiments.

With neutrons, where the absorption is rather low even near absorption edges, Rauch, Treimer & Bonse (1974) Baupsiess, Bonse, Rauch & Treimer (1974) were able to use a simple arrangement (figure 9a). With the phase shifting sample S is rotated through an angle \( \gamma \) the optical path difference is

\[
\Delta t [\arccos (\theta + \gamma) - \arccos (\theta - \gamma)].
\]

Because \( t \) can be large in the sense that the sample produces a large phase shift, many fringes can be recorded with quite small rotations (a few degrees). The fringes which are formed in this way are detected with a neutron sensitive counter so that low beam powers can be used. Although the mean intensities range between 0.5 neutron s\(^{-1}\) and 2 neutrons s\(^{-1}\), fringes are clearly visible in experiments lasting a few hours. From these fringe patterns, neutron scattering lengths were measured for aluminium and bismuth at approximately 2 Å neutron wavelength.

Experiments on neutron interferometry are planned by this group at high flux...
facilities where novel methods for the examination of magnetic structures and for the measurement of neutron spin sensitive interference effects will be feasible.

Although this rather simple interferometer provides for extremely stable operating conditions, it is not suitable for the corresponding X-ray measurements because the interference orders involved are then so small. Furthermore, the approaches which were previously mentioned for characteristic radiation are impracticable with tunable laboratory X-ray sources. Two methods have been designed to circumvent these difficulties, one using a synchrotron source to provide high enough intensities and the other using photon counters with scanning X-ray interferometers.

Figure 9. X-ray and neutron interferometers used to measure the optical constants of the samples S.

Cusatis & Hart (1974) were able to measure phase shifts with a scanning interferometer similar to the one used to make absolute measurements of the lattice spacing of silicon (figure 6c). No optical mirrors are necessary in this application and a very small scanning range will suffice. As the translational stiffness of the X-ray interferometer was approximately 1 dyn Å⁻¹ (1 dyn = 10⁻⁵ N) the necessary forces were generated electrostatically between two metal plates C1 and C2 (figure 9b). They were approximately 1 cm² in area and 10 μm apart so that 10 V sufficed to drive the interferometer 1 Å. The force generated is proportional to the square of the field between the plates; in practice, when fringe profiles are fitted by a computer this is not an inconvenience. Figure 10 shows several traces obtained with the white radiation from a laboratory Bremsstrahlung source. The wavelength selected by the 220 Bragg reflexion is 0.6956 Å, quite close to the zirconium K absorption edge. The wavelength resolution of the interferometer is 0.001 Å. The
first harmonic, diffracted by the 440 Bragg planes at 0.348 Å, is also present and, as the record shows, the Bragg spacing is half that of the fundamental. Since the harmonic is not near the absorption edge it is used to measure the thickness of the zirconium foil. Theoretical estimates of the refractive index at 0.348 Å should be fairly reliable. It is straightforward at these wavelengths to separate the harmonics by amplitude analysis of the output pulses obtained from the scintillation counter. Second and higher order harmonics were not present in the primary beam. A substantial improvement will be obtained when the scintillation counter is replaced by a semiconductor detector.

Bonse & Materlik (1974) used the high intensity X-ray beam from the Deutsches

*Figure 10. Phase-shift measurements made with an electrostatically driven scanning X-ray interferometer. Upper curves at \( \lambda = 0.6956 \) Å, lower curves obtained simultaneously at \( \lambda = 0.348 \) Å. Left hand part is obtained with a zirconium foil in one beam. The right hand part of the record is from the empty interferometer.*
Elektronen Synchrotron (D.E.S.Y.) at Hamburg to measure the dispersion corrections for nickel near its K absorption edge. Their experimental arrangement is sketched in figure 9c. Since the spectrum of synchrotron radiation is essentially white, the X-ray interferometer has pass bands near the wavelength $\lambda$ for which it is intentionally set and also near wavelengths $\frac{1}{2}\lambda$, $\frac{1}{4}\lambda$ etc. If all these were recorded on the film, then rather complicated multiple order moiré fringes would be obtained. Bonee & Materlik (1974) found that several peaks occurred in the rocking curve which was obtained by rotating the double Bragg reflector while keeping the X-ray interferometer stationary. By energy dispersive analysis with a semiconductor detector they showed that each peak consisted of a single harmonic and that each harmonic could be separately recorded on film. Figure 11, plate 4, shows such a recording made near the nickel absorption edge.

In this experiment $\phi$ is fixed to select the fundamental wavelength $\lambda$ then, with a nickel foil $S$ in one of interfering beams, the whole spectrometer is rotated ($\theta$) while the film is moved in synchronism with the rotation axis to record the dispersed spectrum of rotational moiré fringes. The distance of a fringe from the lower edge of the fringe is a measure of the refractive index. A distinctive dip can be seen in the fringe pattern near the K absorption edge of nickel. The scale mark $\Delta \lambda$ indicates the energy resolution of the system. A more direct way to observe the variation of refractive index with wavelength is to compare the intrinsic rotational moiré fringe spacing in the region $B$ with the fringe spacing in the region $A$ which includes phase shifts due to the nickel. In the spectral range covered by figure 11, plate 4, the refractive index decrement varies by about 8%. The photograph is an exact X-ray analogue of the famous demonstration (due to Wood 1911) of the anomalous dispersion of sodium vapour near the D lines.

**DISPERSE X-RAY INTERFEROMETRY**

In all of the applications so far mentioned the X-ray interferometers were achromatic. For all of the Bragg reflexions involved in an interferometer the same Bragg planes were used. Some degree of chromatic resolution can be achieved if a mixture of Laue-case (transmission) and Bragg-case (reflection) optical components are used. Bonee & Hart (1968) demonstrated that such dispersive interferometers do allow measurements of phase shifts, but no developments followed. When more than one reciprocal lattice point lies on the Ewald sphere many new designs, which are all highly dispersive, become possible. Figure 12 indicates two possible Michelson interferometers. If the ray direction is reversed by the sequential operation of the $hkl$ and $pqf$ Bragg reflexions then, neglecting the influence of refraction,

$$\arcsin \{ \lambda (h^2 + k^2 + l^2)^{1/2} / 2 \alpha \} + \arcsin \{ \lambda (p^2 + q^2 + r^2)^{1/2} / 2 \alpha \} = \frac{1}{2} \pi,$$

so that $\lambda$ is thereby fixed. The interferometer in figure 12a (Deslattes 1968) operates at the wavelength of Co Kα, with the 202 and 404 Bragg reflexions from germanium near room temperature. Direct path length variations could be introduced by
translations of one retroreflector. There are severe limitations on the dimensions of such an interferometer so that no systematic experimental work has yet been done. An interesting design is proposed in figure 12(b). By combining the appropriate pair of Bragg reflections, for example $\overline{2}20$ and $400$, Bragg angles of $\frac{1}{2}\pi$ and $\frac{3}{4}\pi$ may be sequentially obtained so that a simple interferometer results. $220$ Bragg reflexions are also available for beam steering, should that be necessary. In this interferometer the wavelength pass-band is near $\lambda = \frac{1}{2}a$, or, if higher orders are used, near harmonies of this wavelength. As in the other instrument displacements of the retroreflectors can be used to introduce directly changes in the optical path length. There are many possible designs of these kinds. In each case the spectral pass band, which

![Figure 12. Possible designs of X-ray Michelson interferometers.](image)

is fixed by crystal geometry, is unlikely to be within a characteristic X-ray linewidth. However, intensity problems are not severe because both synchrotron sources of X-rays and high flux neutron reactors are becoming available to experimenters. Within the spectral ranges of these interferometers it should be possible to do Fourier transform spectroscopy so as to achieve higher spectral resolution than permitted in conventional systems. Finally it is notable that such high resolution spectroscopy is already possible with the simplest X-ray and neutron interferometers. Order scanning over the requisite range can be achieved with weakly absorbing wedges of beryllium and aluminium. Since the thicknesses $t_n$ (table 2) for both X-rays and neutrons are a few tens of micrometres, and since monolithic interferometers 100 mm long have been made, visibility curves are measurable over several thousand orders. The spectral pass-band of a non-dispersive interferometer is fixed by the collimation system, and it may be chosen at will by changing the mean Bragg-angle for the incident beam. Provided that sufficiently intense sources are available both X-ray and neutron Fourier transform spectroscopy should be viable high-resolution methods.
Summary

Development of the simplest monolithic crystal interferometers has reached the point where they have become X-ray optical tools. In the near future they should make contributions to our knowledge of the fundamental constants, of X-ray dispersion parameters and of neutron scattering lengths. Phase-contrast X-ray and neutron microscopy await development.

The detailed theory of coherent diffraction systems has not been worked out. The exploration of other designs, not mentioned in this review, is still in its infancy. Multiple Bragg reflexion systems which use the same monolithic crystal design principles have already been applied in X-ray optical systems. But these are topics for other reviews.

References


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