McCann, D. R. & Cartz, L. (1972). *J. Appl. Phys.* **43**, 4473–4477.
Morosin, B. & Schirber, J. E. (1969). *Phys. Lett. A*, **30**, 512–513.
Morosin, B. & Schirber, J. E. (1972). *Solid State Commun.* **10**, 249–251.
Schirfel, D. (1977). *Rev. Sci. Instrum.* **48**, 24–30.
Shimomura, O., Takemura, K., Fujii, Y., Minomura, S., Mori, M., Noda, Y. & Yamada, Y. (1978). *Phys. Rev. B*, **18**, 715–719.
Takemura, K., Fujii, Y., Minomura, S. & Shimomura, O. (1979). *Solid State Commun.* **30**, 137–139.

Acta Cryst. (1984). A**40**, 502–507

Adjustable Resolution Bragg Reflection Systems

By M. Hart, A. R. D. Rodrigues* and D. P. Siddons†

Wheatstone Laboratory, King's College, Strand, London WC2R 2LS, England

(*Received* 21 *February* 1984; *accepted* 8 *March* 1984)

Abstract

In this paper is described the realization of an idea due to J. W. M. Dumond who, almost 50 years ago, suggested that Bragg diffraction peaks might be made narrower by multiple reflection [Dumond (1937). *Phys. Rev.* **52**, 872–883]. The optical elements which result belong to a family of harmonic free monochromators, X-ray and neutron polarizers whose properties make feasible many new Bragg diffraction optical systems.

Introduction

Nearly fifty years ago Kirkpatrick first suggested that the intrinsic width of the Bragg reflection from perfect single crystals might be reduced by making several successive reflections from different crystals. Dumond (1937) took up the idea and investigated the possibilities theoretically. Although Dumond calculated that a double Bragg reflection, successively from two calcite cleavage planes, could be used to reduce the width of the composite Bragg peak from $3\frac{1}{2}''$ to only $1\frac{1}{4}''$ at 0·71 Å wavelength he stated that Bollmann, Bailey & Dumond (1938) found experimentally that the attainable narrowing was insignificant. Dumond (1937) approximated the shape of the Bragg reflection peak with a Lorentzian $R_L(y)=1/(1+y^2)$, where y represents the angle of incidence, and went on to show that no useful narrowing of the Bragg peak could be expected.

In this paper we show that Kirkpatrick's ideas can be successfully implemented with appropriate design and we demonstrate narrowing of the quadruply diffracted 422 rocking curve in silicon from 3·0″ to only 0·8″ at 1·54 Å wavelength. Two important developments have made this work possible; ideally perfect crystals are now commercially available and the concept of monolithic construction with elastic adjustment Okkerse (1963) has been developed to achieve adequate stability and control in multiple Bragg reflection systems.

* Present address: Departamento de Fisica, Universidade Federal do Parana, CX Postal 1862-8000, Curitiba PR, Brasil.

† Present address: Director of Research & Development, Precision Electronic Components Ltd, Toronto, Canada.

Theoretical and experimental background

Zero absorption approximation

The original Kirkpatrick–Dumond idea is indicated in Fig. 1. Taking two identical crystals whose reflectivities are given by

$$R_B(y)=R_1(y)=R_2(y+\Delta\theta)=\big||y|-(y^2-1)^{1/2}\big|^2, \quad (1)$$

$$R(\Delta\theta)=R_1(y)R_2(y+\Delta\theta), \quad (2)$$

the double reflectivity profile [$R(\Delta\theta)$ in Fig. 1] may have a much lower angular spread than the Bragg peaks of the individual single crystals. This is the 'offset narrowing' concept which Kirkpatrick and Dumond proposed.

Dumond (1937) noted, as Fig. 1 shows well, that as the composite Bragg peak is narrowed (so that $\Delta\theta$ approaches 2 on the y scale) a progressively larger fraction of the integrated intensity resides in the tails of the peak. The corresponding decrease in signal-to-noise ratio when the narrowed peak is used as a probe is most unhelpful. Dumond also reported that 'Bollman and Bailey have found experimentally with three calcite crystals reflecting Mo K radiation on their cleavage planes and have subsequently verified by graphical integration with the theoretical diffraction patterns of Prins that the simple application of the principle of displaced superposition of diffraction patterns is not enough to give a "tool" adequate satisfactorily to reveal the Prins diffraction pattern of a third crystal' (Bollman, Bailey & Dumond, 1938). At long wavelengths, spectroscopists in the 1930's

0108-7673/84/050502-06$01.50

commonly used the approximation $R_L(y) = 1/(1+y^2)$ to describe the shape of the Bragg peaks from their monochromators. As Dumond (1937) realized, the composite peak $R_L(\Delta\theta) = R_L(y)R_L(y+\Delta\theta)$ is always broader at half of its maximum height than the original Lorentzian. Based on the same Lorentzian approximation to the Bragg peak, Dumond also concluded that although the width of the peak became smaller, no *useful* narrowing of the peak could be obtained by undisplaced superposition ($\Delta\theta = 0$) because the integrated reflection power is decreased more rapidly than the width.

These gloomy conclusions remained unchallenged until now. It turns out that the Bragg peaks are sufficiently different from Lorentzians to invalidate Dumond's conclusions and that commercially available crystals are now sufficiently perfect for us to demonstrate narrowing of the Bragg peaks. Such crystals were not available until after 1960 and detailed peak-shape analysis was impracticable before computers became widely available.

On the other hand, Dumond concluded, on the basis of the Lorentzian approximation, that spectrometer resolution *could* be improved by undisplaced superposition ($\Delta\theta = 0$). Whereas the peak of the Lorentzian is 1 and its full width at half unit maximum is 2, when subjected to n undisplaced multiple reflections the resulting curve $R_L^n(y)$ has unit maximum and a width of $2(2^{1/n}-1)^{1/2}$; 0·87 for $n = 4$. In practical cases where $R(y)$ is more nearly given by (1) the width of $R^n(y)$ is almost independent of n and has a limiting value of 2, thus showing that Dumond's conclusion is spurious, being based on an inappropriate approximation.

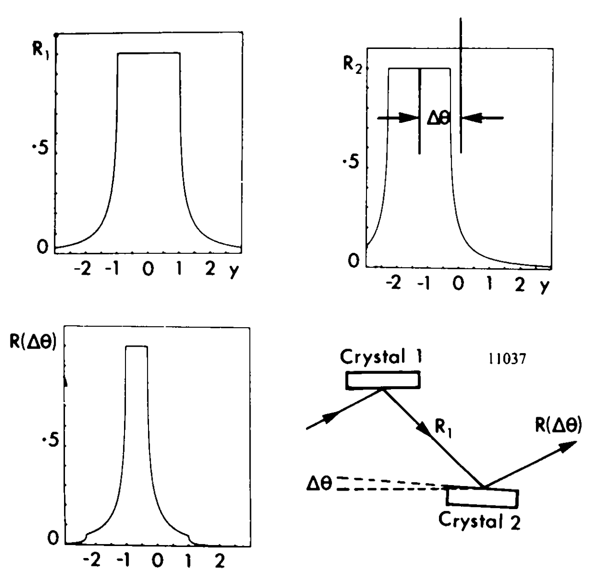

Fig. 1. The 'offset narrowing' concept of Dumond and Kirkpatrick for two successive Bragg reflections in the approximation of zero absorption. In typical cases with X-rays or neutrons at 1 Å wavelength, $y = 1$ corresponds to an angle of between 0·1 and 10″.

Normal absorption

Fig. 2 compares the calculated reflectivity of silicon with the Lorentzian. Curves are also shown for cases of multiple parallel superposition when $\Delta\theta = 0$. A number of features are apparent: because the peak reflectivity is close to one, multiple Bragg reflections do not have a strong influence on the peak reflectivity. The intensity in the tails is, however, drastically reduced by multiple Bragg reflections. Whereas the Lorentzian narrows significantly, the Bragg peak width at half height is hardly changed. For our present application it is important to notice that multiple Bragg reflections allow control of the peak shape and suppression of the tail intensity.

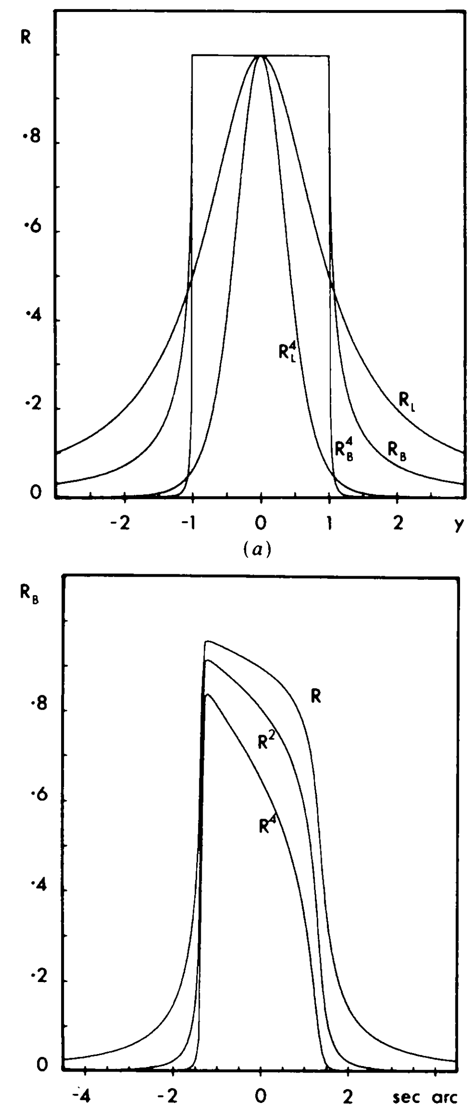

Fig. 2. (*a*) Comparison between the Darwin and Lorentzian peak shapes for single and quadruple reflections. (*b*) Intrinsic R, double R^2 and quadruple R^4 Bragg reflectivities for the 422 reflection of Cu $K\alpha$ radiation from silicon.

Multiple reflection offset narrowing

Each crystal in Fig. 1 could be replaced with a multiply Bragg reflecting crystal. Two different schemes are possible in principle as Fig. 3 shows, though when $n=m=1$ they are identical. The integrated reflection powers are different in the two cases;

$$R_{\rm I}(\Delta\theta)=\int_{-\infty}^{\infty}\prod_{p=0}^{m+n-1}R(y+p\Delta\theta)\,{\rm d}y \qquad (3a)$$

$$R_{\rm II}(\Delta\theta)=\int_{-\infty}^{\infty}R^{n}(y)R^{m}(y+\Delta\theta)\,{\rm d}y. \qquad (3b)$$

In the zero absorption approximation the type II system is simplest to understand. When m and n are large enough the composite profile $R_{\rm II}(\Delta\theta)$ is rectangular; the peak reflectivity is unity and the width is $(2-\Delta\theta)$ on the y scale. There are no tails. The individual reflection profiles $R(y)$ have unit height and width 2 on the y scale.

By contrast the tails of the type I system are higher.

Historical perspective

To the casual observer these new Bragg reflection optical systems may appear to be similar to other devices which we have described. Indeed they are identical! Let us take the type II device in Fig. 3 as an example.

Reflection-curve tail suppression

Bonse & Hart (1965) first demonstrated that multiple Bragg reflections in grooved crystals ($\Delta\theta=0$) *do*

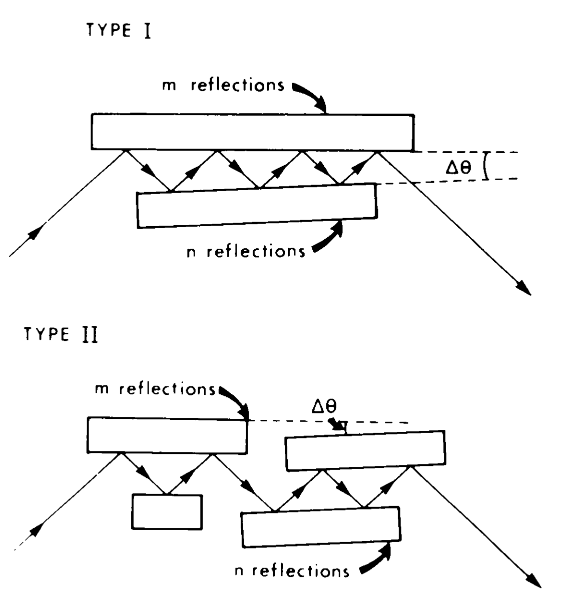

Fig. 3. Multiple reflection systems showing two different offset schemes. Note that the offset $\Delta\theta$ need not be angular but might also be achieved through a change in lattice spacing Δd; for example, in synchrotron radiation experiments an offset can be achieved accidentally through beam heating.

not significantly narrow the reflection profile, contradicting Dumond's result based on the consideration of Lorentzian shape functions, but *do* result in the elimination of the tails of the Bragg peak. The principle has been widely used in X-ray small-angle scattering and in X-ray and neutron spectrometers.

Harmonic-free monochromators

The harmonic Bragg reflections, which also occur roughly at the same Bragg angle but with wavelengths λ/n, $n=2,3,\ldots$ *etc.*, are narrower than the fundamental. The harmonic reflections have widths $2F_n/n^2F_0$ on the y scale. Thus, no harmonic peak is wider than $\frac{1}{4}$ of the fundamental width in simple crystal structures and an offset $\Delta\theta\geq 0{\cdot}25$ on the y angle scale is sufficient to suppress the harmonics. Such harmonic-free monochromators were first described by Hart & Rodrigues (1978) and are now in routine use at synchrotron radiation sources.

Tuneable polarizers

The reflection profile for the π state of polarization is $2|\cos 2\theta|$ y units wide. If the two grooves are offset by this angle then they act as a perfect polarizer with an angular passband for σ-polarized radiation of $2(1-|\cos 2\theta|)$. The device works as a neutron polarizer too (Hart & Rodrigues, 1979).

Adjustable resolution devices

Finally, if the offset is increased to $2(1-\Delta)$ the angular pass-band becomes 2Δ, which may be very small if high angular resolution is required; the offset groove forms the basis of a Bragg reflection spectrometer with *adjustable* resolution. We present here some measurements and calculations of the performance of a silicon Bragg reflection spectrometer with adjustable resolution. By making two similar devices ('narrowers') and rocking one against the other in the non-dispersive setting we demonstrate controlled and adjustable Bragg peak narrowing as envisaged by Dumond almost fifty years ago.

Experimental results

Although the results in Fig. 2(a) are directly applicable in neutron and γ-ray diffractometry when the absorption is very low indeed, in the majority of experiments with X-rays the absorption is important. Nevertheless, a large number of computations lead us to believe that the type II systems always result in sharper composite profiles than can be obtained in type I systems with the same total number of Bragg reflections.

After exhaustive calculations for absorbing crystals we concluded that the simplest demonstration of a variable resolution diffractometer using readily available crystals would employ the 422 Bragg reflection of silicon with copper $K\alpha_1$ radiation, with $m = n = 2$ in a type II system.

Crystal unit design and calibration

We emphasize that although the design principles are clear in the zero absorption case, in real X-ray cases with strong absorption detailed calculations are required to determine the optimum number of Bragg reflections for a particular application. To achieve the necessary long-term stability the offset multiple Bragg reflector is cut as an elastically adjustable monolithic crystal UNIT (Fig. 4). The offset angles required are only up to 10″ and that is achieved by bending of the leaf-spring region of the crystal under the force generated between a fixed permanent magnet and an air-cored electromagnet. By constructing two nominally identical complete systems we were able to use them on a double-crystal diffractometer to explore their intrinsic and variable reflection profiles.

Before attempting to obtain double-crystal rocking curves the single-UNIT magnetically scanned rocking curves were measured. So that known offset angles could be conveniently calibrated and fixed in later experiments, the whole series of measurements was automatically controlled and the data stored in a microcomputer system (Rodrigues & Siddons, 1979) which was connected for display and computational purposes to a Hewlett-Packard 9485A calculator and plotter. The monolithic crystal systems (UNIT 1 and UNIT 2) were designed for use with horizontal goniometer axes at synchrotron radiation sources. In these experiments we used vertical axes at a conventional laboratory X-ray source and consequently the elastic-spring regions twist under gravity. This tilt error was compensated by applying appropriate couples with weights attached to the crystal. Fig. 5 shows the intrinsic magnetically scanned rocking curves for the two UNITS and the corresponding theoretical curve which is simply $R^2 * R^2$ where $*$ denotes convolution. Since R^2 is almost rectangular (Fig. 2) we expect, and observe, that the convolution is almost an isosceles triangle. Notice too that the two Bragg reflections in each part of the offset monolith are already sufficient to suppress noticeably the tails of the rocking curve. The absolute peak intensities from the two UNITs differ by about 5%, pointing to residual strain or inhomogeneities in the two crystals

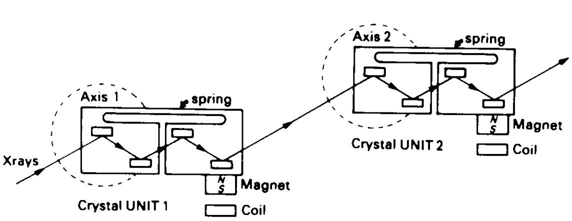

Fig. 4. Experimental arrangement showing two crystal UNITs set up on a two-axis diffractometer. Each unit contains four Bragg reflections which are internally adjustable (with the coil, magnet and spring system) as two pairs of fixed double Bragg mirrors. Including the tilt adjustment between UNITs, the arrangement is equivalent to an eight-axis goniometer.

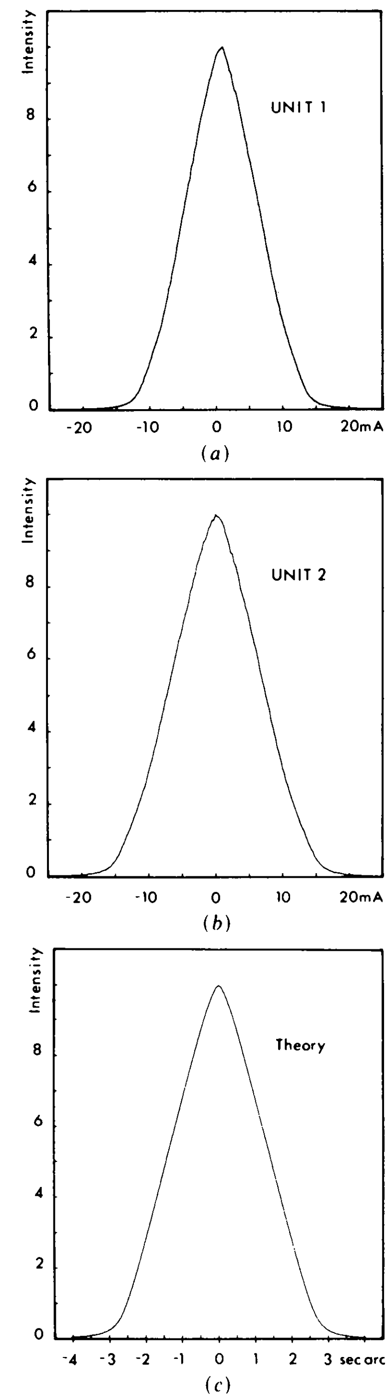

Fig. 5. (*a*), (*b*) UNIT rocking curves; intensity as a function of coil current for each of the four reflection UNITs using Cu $K\alpha_1$ radiation and 422 Bragg reflections. (*c*) Theoretical curve $R^2 * R^2$ which is used to determine the relationship between magnet current and angle for each UNIT.

used and the peak widths differ by about 20% in magnet current, reflecting slight constructional differences between the two UNITs (different spring dimensions, magnet strengths, coil/magnet geometry *etc.*). Comparison with theoretical rocking curves (Fig. 5*c*) allows direct calibration of the magnetic drive characteristic in mA per s of arc.

Since the Bragg angle for the 422 reflection of Cu $K\alpha_1$ is 44·01°, the diffracted beams are about 97% linearly polarized after a single Bragg reflection and are completely polarized in multiple-reflection offset-crystal systems (Hart & Rodrigues, 1979). At other Bragg angles the situation is much more complicated but with polarized sources, such as storage rings, only one state of polarization would be important.

Double-crystal measurements

Having calibrated the magnetic offset scan systems for the two crystals UNITs they were mounted, in the non-dispersive setting, on a double-axis diffractometer. By minimizing the width of the observed rocking curves the relative tilt between UNIT 1 and UNIT 2 was adjusted to zero.

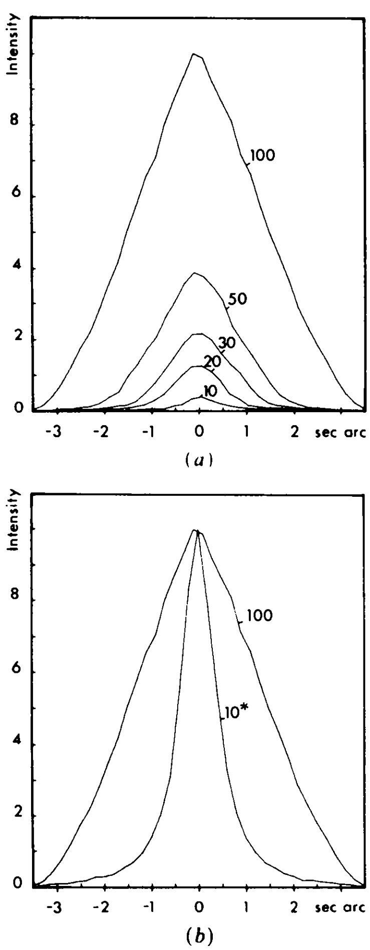

Fig. 6. (*a*) Measured rocking curves with both UNITs offset to the same angle; see text. (*b*) 100 and 10% offset intensity rocking curves normalized to the same peak intensity.

Fig. 6 shows the series of symmetric rocking curves obtained by offsetting both crystals to the same intensity point of their magnetic rocking curves; by offsetting UNIT 1 to n% of its peak intensity, UNIT 2 to n% of its peak intensity and then rocking one crystal UNIT against the other. The resultant curve is symmetric because it is a self-convolution. The width at half-maximum intensity decreases systematically as the UNIT offsets increase from 100 to 50, 30, 20 and 10% of peak (single-UNIT) intensity. Within the achievable precision of the experimental adjustments the peak intensities in the rocking curves are in good agreement with calculations. Whereas the width at half intensity of R^4 (Fig. 2*b*) is 2·2″, the width of the convolution $R^4 * R^4$ is 3·0″ (100% intensity, or $\Delta\theta = 0$ rocking curve in Fig. 6). For triangle functions, to which R^4 closely approximates, James (1948) gives the ratio of function width to self convolution width as 1·35 which is very close to the value 1·36 found here.

The curve marked 10* in Fig. 6(*b*) is the rocking curve obtained with both UNITs offset to 10% of peak intensity, normalized to the same height as the zero-offset rocking curve. The achieved narrowing from 3·0″ full width to only 0·8″ can be clearly seen. Detailed analysis shows the narrowed peak to be symmetric to the limit set by counting statistics; between 10 and 100% of peak intensity the mid-chord is constant to 0·01″, or about 1% of the peak width, and $\frac{3}{4}$ of the integrated intensity lies in that range.

Applications

In optical spectroscopy with prisms or diffraction gratings one commonly controls resolution by adjusting the width of a convenient slit. Such control now becomes feasible in systems which use Bragg reflecting crystals as dispersion elements and in such systems remote electrical control as provided by our magnetically adjusted elastic systems is very convenient. There are other ways in which Bragg-peak narrowing can be achieved but are not of general utility in spectroscopy and diffractometry.

Apart from the method described here the Borrmann effect, oblique Bragg reflections and *Pendellösung* fringes have been used to create angularly narrow diffraction probes. Authier (1961) first used the Borrmann effect as a means of controlling ray divergence in propagation experiments. Partial polarization is also achieved (Cole, Chambers & Wood, 1961) but the efficiency is so low that the method is not of general use and, in any case, can only be used with spatially narrow beams. *Pendellösung* fringes have also been used to increase the local gradients in rocking curves and thereby to increase angular sensitivity (Cusatis, Hart & Siddons,

1982) but again this method is not practicable for general spectroscopic use since the signal-to-noise ratio would be very poor. The third method which seeks to control the Bragg width by oblique Bragg reflections has been widely used (Renninger, 1961, 1967; Kikuta & Kohra, 1970; Kikuta, 1971; Matsushita, Kikuta & Kohra, 1971; Hashizumi & Kohra, 1971; Bonse & Graeff, 1973; Kohra, 1972). Kohra & Kikuta (1968) summarize fairly completely the relevant literature so that not all references need be mentioned here. In essence, cutting the surface of a crystal at angle α to the Bragg planes reduces the range of Bragg reflection by a factor $b^{1/2}$, where $b=\sin(\theta-\alpha)/\sin(\theta-\alpha)$. Oblique incidence at $\frac{1}{2}°$ to the surface yields a narrowing factor of ten at a Bragg angle $\theta=60°$ but such small glancing angles are difficult to achieve, result in specular external reflection and, by definition, do not permit the tuneability necessary for spectroscopy. However, it would appear to be possible to achieve large narrowing factors by combining this method and the present one. For example, Kikuta (1971) achieved $b=573$ and we demonstrate herein a further factor of four so that the total narrowing in a combined device could be $4b^{1/2}=100$ with the 422 reflection from silicon. Since the energy resolution of a single-crystal Bragg spectrometer based on the 422 Bragg reflection is $1{\cdot}4\times10^{-6}$ (Beaumont & Hart, 1974), the energy resolution of an oblique-offset multiple-reflection Bragg spectrometer would then be $1{\cdot}4\times10^{-8}$; quite adequate for many experiments which have been proposed using synchrotron radiation sources, for example, the measurement of inelastic X-ray scattering due to phonons. The construction has the added advantage that setting up could be done in the low-resolution mode with high intensity and the resolution would be varied to suit the problem in hand.

References

AUTHIER, A. (1961). *Bull. Soc. Fr. Minéral. Cristallogr.* **84**, 51–89.
BEAUMONT, J. H. & HART, M. (1974). *J. Phys. E*, **7**, 823–829.
BOLLMAN, V. L., BAILEY, H. H. & DUMOND, J. W. M. (1938). *Phys. Rev.* **54**, 792–801.
BONSE, U. & GRAEFF, W. (1973). *Z. Naturforsch. Teil A*, **28**, 558–564.
BONSE, U. & HART, M. (1965). *Appl. Phys. Lett.* **7**, 238–240.
COLE, H., CHAMBERS, F. W. & WOOD, C. G. (1961). *J. Appl. Phys.* **32**, 1942–1945.
CUSATIS, C., HART, M. & SIDDONS, D. P. (1982). *Acta Cryst.* A**39**, 199–202.
DUMOND, J. W. M. (1937). *Phys. Rev.* **52**, 872–883.
HART, M. & RODRIGUES, A. R. D. (1978). *J. Appl. Cryst.* **11**, 248–253.
HART, M. & RODRIGUES, A. R. D. (1979). *Philos. Mag.* B**40**, 149–157.
HASHIZUMI, H. & KOHRA, K. (1971). *J. Phys. Soc. Jpn*, **31**, 204–216.
JAMES, R. W. (1948). *The Optical Principles of the Diffraction of X-rays.* London: Bell.
KIKUTA, S. (1971). *J. Phys. Soc. Jpn*, **30**, 222–227.
KIKUTA, S. & KOHRA, K. (1970). *J. Phys. Soc. Jpn*, **29**, 1322–1328.
KOHRA, K. (1972). *Proc. VIth International Conference on X-ray Optics and Microanalysis*, pp. 35–43. Univ. of Tokyo Press.
KOHRA, K. & KIKUTA, S. (1968). *Acta Cryst.* A**24**, 200–205.
MATSUSHITA, T., KIKUTA, S. & KOHRA, K. (1971). *J. Phys. Soc. Jpn*, **30**, 1136–1144.
OKKERSE, B. (1963). *Philips Res Rep.* **18**, 413–431.
RENNINGER, M. (1961). *Z Naturforsch. Teil A*, **16**, 1110–1111.
RENNINGER, M. (1967). *Adv. X-ray Anal.* **10**, 32–39.
RODRIGUES, A. R. D. & SIDDONS, D. P. (1979). *J. Phys. E*, **12**, 403–408.

Acta Cryst. (1984). A**40**, 507–514

Propagation of X-ray Beams in Distorted Crystals (Bragg Case). I. The Case of Weak Deformations

BY J. GRONKOWSKI* AND C. MALGRANGE

Laboratoire de Minéralogie–Cristallographie, Universités Pierre et Marie Curie (Paris VI) et Paris VII, Tour 16, 4 place Jussieu, 75230 Paris CEDEX 05, France

(*Received 1 August 1983; accepted 12 March 1984*)

Abstract

It has been commonly admitted that the theories of X-ray propagation in distorted crystals based on the principles of geometrical optics [Penning & Polder (1961). *Philips Res. Rep.* **16**, 419–440; Kato (1963). *J. Phys. Soc. Jpn*, **18**, 1785–1791; Kato (1964). *J. Phys. Soc. Jpn*, **19**, 67–71, 971–985] were applicable only in the transmission (Laue) case. It is demonstrated in this paper that they can be applied more generally in all cases where beams can be defined, *i.e.* also in the Bragg case outside the total reflection range. Simple formulae for the case of a constant strain gradient in symmetric Bragg geometry are derived from a general formulation of the basic equation of geometrical theory using a new universal parameter *a*. They are verified by solving Takagi's

* Present and permanent address: Institute of Experimental Physics, Warsaw University, 00-681 Warsaw, Hoża Street 69, Poland.

0108-7673/84/050507-08$01.50