# CRYSTAL DIFFRACTION OPTICS FOR X-RAYS AND NEUTRONS

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## INTRODUCTION

In the wavelength range near $\lambda = 1$ Å Bragg reflecting crystals are the only satisfactory dispersion elements for spectroscopy. Modern commercially available single crystals of silicon and germanium are inexpensive, almost uninfluenced by radiation damage and, from the diffraction viewpoint, ideally perfect.

Although perfect crystals Bragg reflect x-rays only over a very small range of incident angles, they do so with very high efficiency so that multiple Bragg reflections can be used without a serious loss of intensity. Multiple Bragg reflections allow great freedom in optical design and we will explore here the control and measurement of spectral profile, phase and polarization.

## DYNAMICAL DIFFRACTION THEORY

The theory of diffraction in perfect crystals is fully developed in a number of standard texts[1-9]. Two important cases are distinguished; in the Bragg-case (reflection) the diffracted wave $\underline{K}_h$ emerges from the same (entrance) surface as the incident wave $\underline{K}_o$ while in the Laue-case (transmission) the diffracted waves emerge from the lower surface. Only in the symmetric Laue-case when the Bragg planes are normal to the crystal surface does the Bragg law give the exact Bragg angle $\theta_L$, so that

$$2\ d \sin \theta_L = n\ \lambda$$

In all other cases the Bragg angle is shifted slightly by refraction by an amount which depends on the optical parameters and on the relative orientations of the crystal surface and the wavevectors. The principal features of multiple reflection systems can be understood in terms of the examples shown in Fig 1 which are drawn for the symmetric Bragg-case.

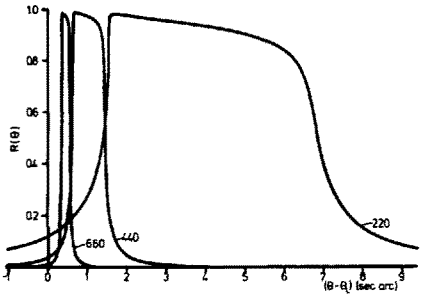


Fig 1(a) Graph of the reflectivity $R(\theta)$ for the fundamental (n=1) 220 Bragg reflection from silicon at $\lambda = 1.54\text{Å}$ and for the harmonics n=2 (440 at $\lambda = 0.77\text{Å}$) and n=3 (660 at $\lambda = 0.385\text{Å}$). The $\sigma$-state of polarization is assumed.

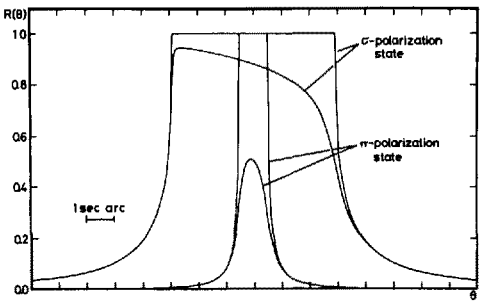


Fig 1(b) $R(\theta)$ for the two polarization states in the 440 Bragg reflection from germanium at $\lambda = 1.54\text{Å}$. The flat-topped curves show the result when absorption is ignored.

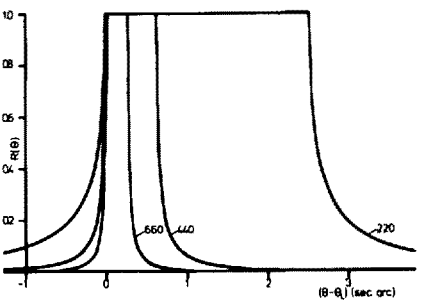


Fig 1(c) $R(\theta)$ for the fundamental (n=1) 220 Bragg reflection of neutrons from silicon at $\lambda = 3\text{Å}$ and for the harmonics n=2 (440 at $\lambda = 1.5\text{Å}$) and n=3 (660 at $\lambda = 1\text{Å}$).

The principal effect of refraction is to shift the Bragg peak and its harmonics away from $\theta_L$. The shift of the centre of the Bragg peak is given by

$$\theta_o - \theta_L = \frac{|\chi_{ro}|}{2\sin 2\theta}\left(1 + \frac{\sin(\theta-\phi)}{\sin(\theta+\phi)}\right)$$

where $\phi$ is the angle between the crystal surface and the Bragg planes and $\chi_{ro} = -8r_e\lambda^2(Z+f')/\pi a^3$ in diamond structure materials. For the harmonics the shift is proportional to $\lambda^2$ or $n^{-2}$. The peak widths $\Delta\theta_o$ are given by

$$\Delta\theta_o = \frac{2\,|C|\,|\chi_{rh}|}{\sin 2\theta}\sqrt{\frac{\sin(\theta-\phi)}{\sin(\theta+\phi)}}$$

where C is 1 for the $\sigma$-polarized state and $\cos 2\theta$ for the $\pi$-polarized

width $\Delta\theta_o$. Thus large sources can be used to gain intensity. In practice $\Delta\theta_o$ lies within the range $10^{-8} < \Delta\theta_o < 10^{-4}$ and the corresponding energy or momentum resolution is determined by the width of the convolution of the two crystal profiles. It is approximately

$$\Delta E/E \simeq \cot\theta . \Delta\theta_o$$

## MULTIPLE BRAGG REFLECTIONS

Since the peak reflectivity is almost 1 (Fig 1) multiple Bragg reflections can be made with little intensity loss. This design freedom allows one to control the shape of the Bragg reflection profile, to split and recombine beams and to control their paths through a system. For example, multiple reflections lead to the elimination of the tails of the Bragg reflection curve[12] and make possible interferometry with x-rays and neutrons[13-18]. In practice most of the multiple reflection diffraction optical systems are monolithic perfect crystals, but a new design freedom is added by the realisation that elastic adjustments are practicable if the crystal is suitably designed. In this way it has been possible to make ultra-stable scanning interferometers and fast interferometric choppers[19,20], harmonic-free monochromators[21] and tuneable polarizers [22].

## HARMONIC FREE SYSTEMS FOR SPECTROSCOPY

There is a fundamental requirement in spectroscopy for "monochromatic" beams with a controlled spectral passband. In practice, with single crystal monochromators, the excellent selectivity of the Bragg reflection (Fig 1) is spoiled by the presence of harmonics and by the background intensity in the wings or tails of the Bragg reflection curve. The former problem has been tackled by a variety of means, shown in Fig 3, while both problems can be solved with suitable multiple Bragg reflection optical systems.

Although specular reflection at a mirror (usually metal-coated quartz, bent to focus the beam) has been widely used to reduce crystal monochromator harmonics at synchrotron radiation sources, there appears to be little information on their actual performance. It is straightforward (e.g. from James (1948)[1]) to show that the harmonic reflectivity at twice the critical angle is 0.4%. In practice, if we include the influence of absorption, the harmonic content will be higher.

state. $\chi_{rh} = r_e \lambda^2 F_h/\pi a^3$ where $F_h$ is the structure amplitude for the active Bragg reflection. The peak widths are therefore proportional to $F_h n^{-2}$ for the harmonics. In the approximation of point stationary atoms - close to the neutron case at room temperature - Fig 1(c) represents a universal result in which changes of atom, wavelength or obliquity simply alter the scale of the abscissa. Comparisons between Fig 1(a) and Fig 1(c) show the influence of finite atoms on the peak widths and the (small) influence of thermal motion ensures that the left hand edges of the peaks in Fig 1(c) do not in fact quite coincide at $\theta = \theta_L$. In practice, x-ray absorption limits the peak reflectivity somewhat as Fig 1(a) and Fig 1(b) show and the diffuse nature of the electron density around atoms moves the near edge of the Bragg peak away from $\theta_L$.

## SPECTROSCOPIC DESIGN

There are only two generic designs for spectroscopic instruments[10,11].

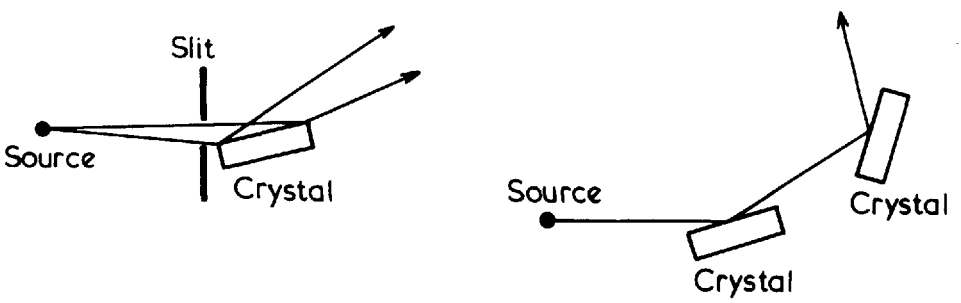


Fig 2(a) Bragg spectrometer (b) Compton or double crystal spectrometer

In the Bragg spectrometer the energy resolution is determined by the divergence of the incident beam $\Delta\theta_i$ which is fixed in practice by the geometry of the source and collimator slit. $\Delta\theta_i > 10^{-4}$ can be achieved in the laboratory with reasonable intensity and at synchrotron radiation sources it might be possible to work with $\Delta\theta_i < 10^{-5}$. The corresponding energy resolution follows from Bragg's law

$$\Delta E/E = \cot\theta . \Delta\theta_i$$

Higher resolution can be achieved in the double crystal arrangement because the angular pass band is determined by the Bragg

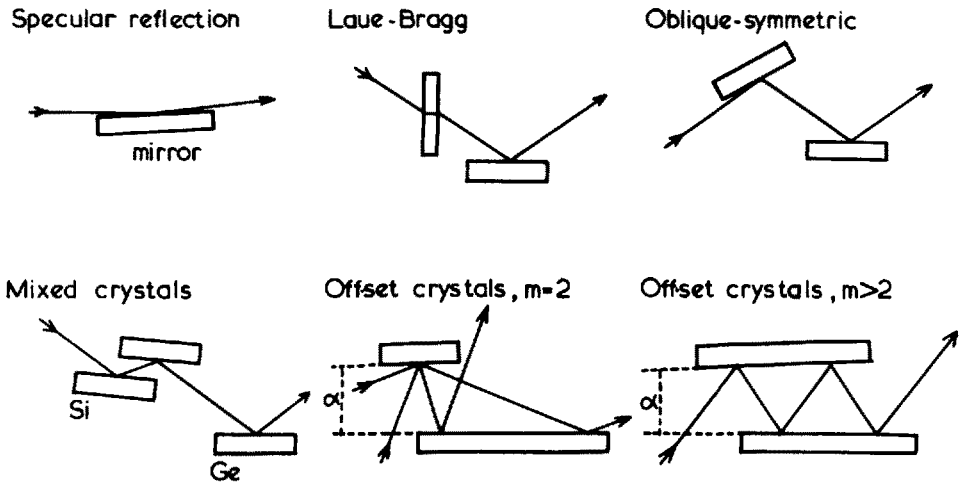


Fig.3 Systems used to control harmonic contamination in spectrometers.

Three of the harmonic reducing optical systems rely on shifting the relative positions of fundamental and harmonics by refraction (Fig 1). Bonse, Materlik and Schröder[23] found that the Laue-Bragg combination did not reduce the harmonic content below a few percent of the fundamental intensity. However, when the Laue-case device is an interferometer and the Bragg-case device is a double-reflection grooved crystal as it was in those experiments, harmonics can at least be conveniently reduced by this method. The refractive index shift can also be exploited by mixing oblique and symmetric Bragg reflections[24]. Unfortunately, the system cannot be scanned over a wide wavelength range unless several oblique crystal cuts are available. By mixing crystals with different refractive indices, for example, a double 220 reflection from silicon and a single 220 reflection from germanium, harmonic suppression to 0.3% can be achieved[23]. Such a system could with difficulty be scanned over wavelength but its passband is fixed.

The off-set grooved crystal systems suffer from none of these disadvantages. Their operation depends not on the refraction effect but on the fact that the Bragg reflection widths are different for the fundamental and the harmonics. The off-set angle $\alpha$ is larger than the

width of the harmonic peak and much less than the width of the fundamental (Fig 1). With m = 2 Bragg reflections there is no restriction on the wavelength range or passband compared with a single Bragg reflection, but the harmonic intensity is reduced to well below 0.5% [21]. As more Bragg reflections are added the harmonic ratio is reduced substantially and can be chosen in the range $10^{-2}$ to (say) $10^{-10}$ by appropriate design.

An incidental advantage of these elastic monolithic off-set crystal monochromators is that they have an excellent mechanical frequency reponse. They can be used for x-ray and neutron beam modulation and in stroboscopic diffraction topography[33].

## PHASE MEASUREMENT - DISPERSION SPECTROSCOPY

Since this topic is covered by Zeilinger[17] in the neutron case and by Graeff[18] for the x-ray case we will be concerned here only with crystal optical principles and with the results so far obtained.

Two different approaches have been adopted. Where film detection was used to determine phase shifts in nickel[25], harmonic control is necessary and was achieved as outlined in the previous section. On the DESY synchrotron source the complete nickel spectrum shown in Fig 4 was obtained in only three hours. By using energy selective detectors and a scanning interferometer[20,26,27] one may work simultaneously at two separate harmonic wavelengths which provide internal calibration of the sample thickness. With 0.8 kVA laboratory Bremsstrahlung sources each spectrum represents 600 hours of counting time but the resulting dispersion curves for zirconium, niobium and molybdenum are very encouraging as Fig 4 shows. With a 100 kW rotating anode x-ray generator these spectra could be obtained in only 5 hours.

In the corresponding neutron case the scattering is entirely characterised by a single parameter; the scattering length,which may be measured at any convenient wavelength. As complete spectra are not necessary, results have already been obtained[17,28] for many different elements.

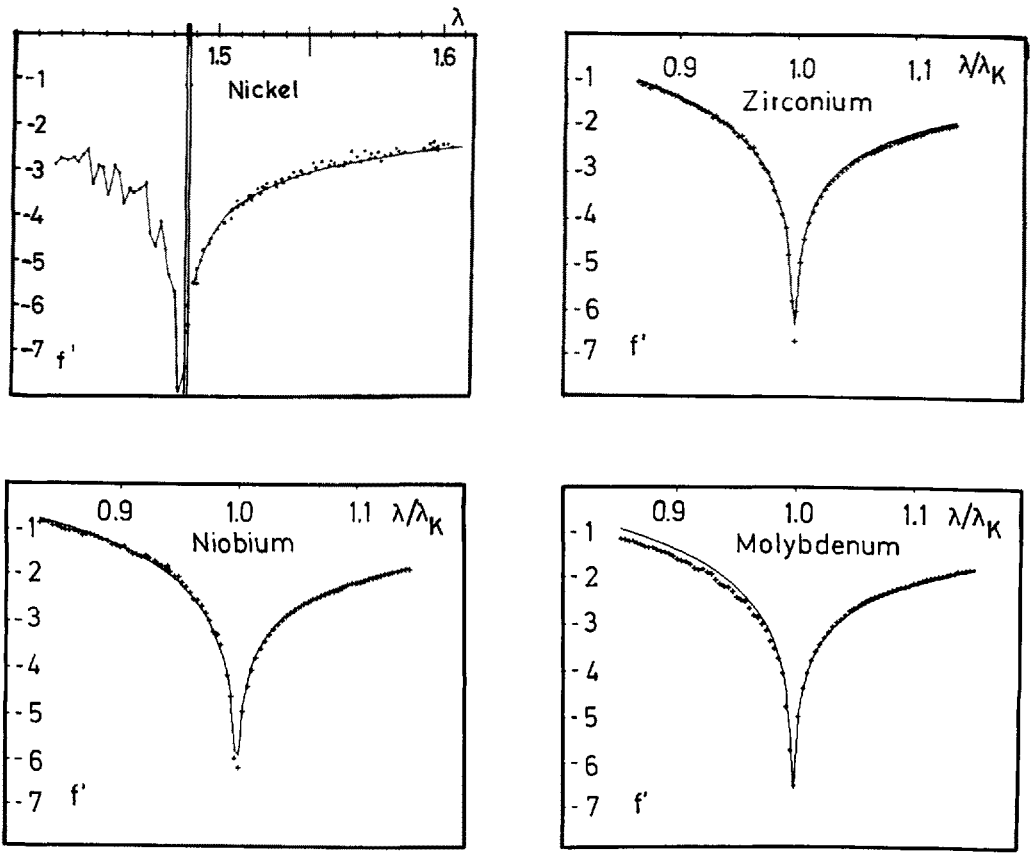


Fig. 4 K-electron dispersion curves obtained by x-ray interferometry.

## POLARIZING OPTICAL SYSTEMS

Prior to the 1960's very little work had been done with polarized x-rays[29]. Simple Rayleigh scattering and coherent Bragg scattering at $90^{o}$ were the only available methods for producing polarized beams. More recently partially polarized sources of synchrotron radiation have become available. Polarization phenomena are reviewed by Skalicky[30].

Recently a tuneable x-ray polarizer has been developed[22]. As Figure 1 shows, the peak reflectivity of a crystal is lower for the $\pi$-polarization state than for the $\sigma$-state. By making multiple Bragg reflections in a groove cut into a perfect crystal we can produce polarized beams. Figure 5 shows calculations of the polarizing ratio $I_{\pi}/I_{\sigma}$ for $1 < m < 10$ in the case of the 440 Bragg reflection from germanium for the wavelength range near the polarizing Bragg angle.

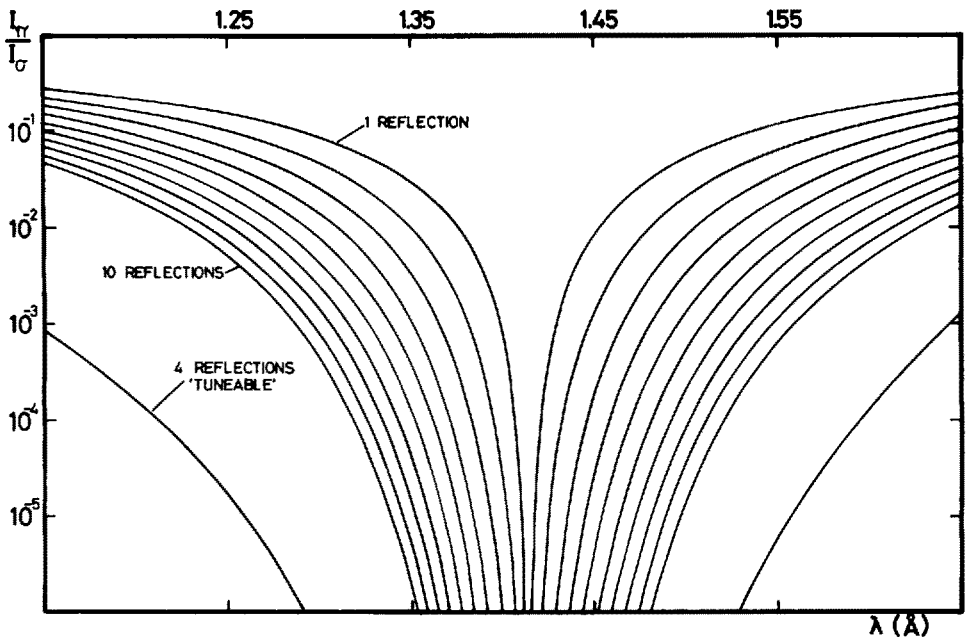


Fig. 5 Calculated polarization ratios for the 440 Bragg reflection from germanium with m-fold Bragg reflections.

As the π-state reflection curve is much narrower than the σ-state reflection curve (Fig 1), the polarization ratio can be further improved if one side of the groove is off-set by a small angle α with respect to the other as in the arrangement for harmonic elimination. The result, for a four-fold Bragg reflection system, is shown as the outermost curve in Fig 5. Detailed calculations in the Table below (for the silicon 422 Bragg reflection with m = 4 Bragg reflections at $\lambda = 1.38\text{Å}$) show how the same off-set grooved crystal acts both as a harmonic free monochromator and as a polarizer. As mentioned before, this general purpose device can also be used as a wavelength modulator and stroboscopic monochromator.

T A B L E

| α/sec.arc. | $I_\sigma(\alpha)/I_\sigma(o)$ | $I_\pi(\alpha)/I_\sigma(\alpha)$ | $I_{844}/I_{422}$ |
|---|---|---|---|
| 0.0 | 1.00 | $5.8\times10^{-2}$ | $9.6\times10^{-2}$ |
| 0.1 | 0.95 | $3.6\times10^{-2}$ | $1.0\times10^{-3}$ |
| 0.2 | 0.85 | $6.3\times10^{-3}$ | $5.0\times10^{-6}$ |
| 0.3 | 0.73 | $5.4\times10^{-4}$ | $1.5\times10^{-7}$ |
| 0.4 | 0.60 | $7.9\times10^{-5}$ | $2.5\times10^{-8}$ |
| 0.5 | 0.47 | $2.0\times10^{-5}$ | $7.5\times10^{-9}$ |

This same principle can also be used to polarize neutrons[22]. At $\lambda = 2\mathring{A}$ using the 110 Bragg reflection from saturated iron we find that the Bragg widths are approximately $\Delta\theta(+) \sim 7.4$ sec.arc and $\Delta\theta(-) \sim 2.1$ sec. arc. At an off-set $\alpha \sim 3$ sec.arc the polarization ratio is calculated to be $I_-(\alpha)/I_+(\alpha) = 5 \times 10^{-4}$.

BEAM STEERING AND CALIBRATION

Multiple Bragg reflections can also be used to simplify the experimental geometry. For example, the precision and range of goniometric control required in a classical double crystal spectrometer (Fig 2) is so daunting that few have been built. The arrangement shown in Fig 6a performs the same task but is far simpler to implement[31]. The double Bragg reflectors might each be off-set grooved crystals.

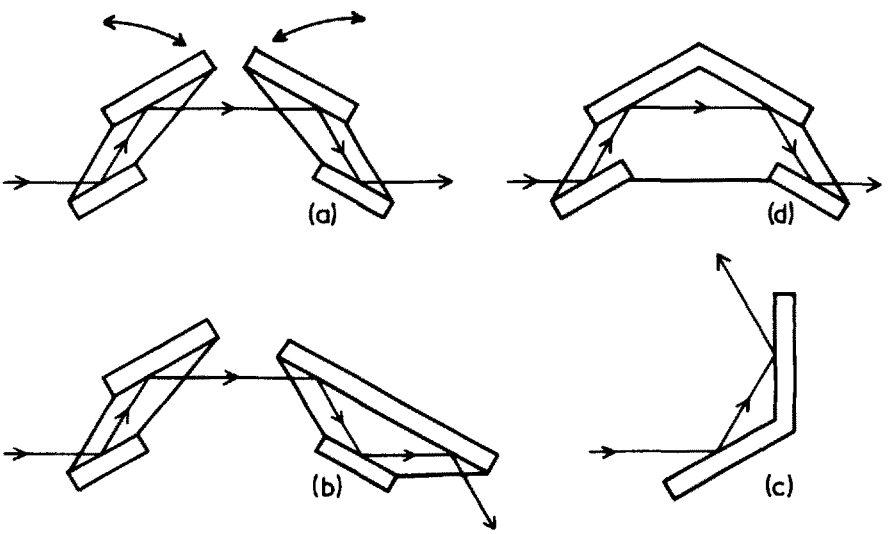


Fig.6 Beam steering and calibration by multiple Bragg reflections

If the final beam is required in the deviated direction then, with virtually no intensity penalty, another Bragg reflection can be added as in Fig 6b. In some situations only a fixed wave-length is required. Than can be prov ided by the monolithic double reflection monochromator shown in Fig 6c. A zero-deviation version, shown in Fig 6d has recently been constructed to calibrate synchrotron radiation spectrometers[32].

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