particular for the examination of a correlation between lattice parameter and the dopant or impurity concentration (Baker, Tucker, Moyer & Buschert, 1968). Such an arrangement can also be a very suitable tool in deformation studies, since it allows the separation of the effect of deformation on the Bragg angle from that due to lattice-parameter change (Skupov & Uspeckaya, 1975).

The basis of the accurate lattice-parameter comparison proposed by Bowen & Tanner (1995) is the use of a high-purity silicon standard (*cf.* §5.3.3.9 below) with a well known lattice parameter. To compensate an error that may result from a slight misalignment of crystal planes in relation to the axes of the instrument, the authors recommend a twofold measurement of the diffraction-peak position of the reference crystal (for a given diffraction position and after rotating the specimen holder through 180° about the axis normal to its surface) and a similar twofold measurement of the diffraction-peak position of the sample – after replacing the reference crystal by the sample. The mean positions of the reference crystal and of the sample are used in calculations of the Bragg-angle difference and then of the unknown interplanar spacing. The method uses a standard double-crystal diffractometer fitted with a monochromator (therefore, a third crystal), which provides a well defined wavelength, and with a specimen rotation stage. The measurement is accompanied by a detailed error analysis. The accuracy of absolute lattice-parameter determination as high as a few tens of parts in 10^6, and a much greater relative sensitivity are reported.

By combining a triple-axis spectrometer with the Bond (1960) method, the device can be used for absolute measurements (Pick, Bickmann, Pofahl, Zwoll & Wenzl, 1977). The device described in the latter paper is an automatic triple-crystal diffractometer that permits intensity measurement to be made in any direction in reciprocal space in the diffraction plane with step sizes down to $0.01''$ and therefore can be used for very precise measurements [see also §5.3.3.4.3.2, paragraph (5)].

5.3.3.7.3. *Multiple-beam methods*

The other possibility of recovering the crystal-angle scale in differential measurements with a double-crystal spectrometer (*cf.* §§5.3.3.7.1, 5.3.3.7.2) is to obtain reflections from two crystal planes [for example, from (hkl) and $(\bar{h}\bar{k}l)$ planes] by means of a double-beam arrangement and to measure them simultaneously.

The second X-ray beam may come from an additional X-ray source (Hart, 1969) or may be formed from a single X-ray source by using a beam-splitting crystal (Hart, 1969, second method; Larson, 1974; Cembali, Fabri, Servidori, Zani, Basile, Cavagnero, Bergamin & Zosi, 1992). In particular, two beams with different wavelengths ($K\alpha_1, K\beta_1$) separated with a slit system can be used for this purpose (Kishino, 1973, second technique). The principle of the double-beam method is shown in Fig. 5.3.3.12. The beams are directed at the first crystal (the reference crystal) so that the Bragg condition is simultaneously fulfilled for both beams, and they then diffract from the second crystal (the specimen). As the second crystal is rotated, a double-crystal diffraction profile is recorded first in one detector and then in the other. The angle $\Delta\theta$ of crystal rotation between the two rocking curves is given by (Baker & Hart, 1975):

$$\Delta\theta = (\theta_1 - \theta_2) = \tan\theta\,\Delta d/d. \qquad (5.3.3.43)$$

This formula leads to the lattice-parameter changes Δd.

A double-beam diffractometer can be used for the examination of variations in lattice parameters of about 10 parts in 10^6 within a sample in a given direction. An example was reported by Baker, Hart, Halliwell & Heckingbottom (1976), who used Larson's (1974) arrangement for this task.

The highest reported sensitivity (1 part in 10^9) can be achieved in the double-source double-crystal X-ray spectrometer proposed by Buschert, Meyer, Stuckey Kauffman & Gotwals (1983). The device can be used for the investigation of small concentrations of dopants and defects.

The method can also be applied for the absolute determination of a lattice parameter, if that of the reference crystal is accurately known and the difference between the two parameters is sufficiently small. Baker & Hart (1975), using multiple-beam X-ray diffractometry (Hart, 1969, first technique), determined the d spacing of the 800 reflection in germanium by comparing it with the d spacing of the 355 reflection in silicon. The latter had been previously determined by optical and X-ray interferometry (Deslattes & Henins, 1973; the method is presented in Subsection 5.3.3.8).

In the case of two different wavelengths and diffraction from two different diffraction planes $(h_1k_1l_1)$ and $(h_2k_2l_2)$, the lattice parameter a_0 of a cubic crystal can be determined using the formula (Kishino, 1973)

$$a_0 = \tfrac{1}{2}\{(L\lambda_1)^2 + [(M\lambda_2 - L\lambda_1\cos\theta_{1-2})/\sin\theta_{1-2}]^2\}^{1/2}, \qquad (5.3.3.44)$$

where $L = (h_1^2 + k_1^2 + l_1^2)^{1/2}$, $M = (h_2^2 + k_2^2 + l_2^2)^{1/2}$, and θ_{1-2} is the difference between the two Bragg angles for the specimen crystal, estimated from the measurement of $\Delta\theta = |\theta_{1-2} - \theta'_{1-2}|$ if the difference θ'_{1-2} for the first (reference crystal) is known beforehand. The idea of Kishino was modified by Fukumori, Futagami & Matsunaga (1982) and Fukumori & Futagami (1988), who used the Cu $K\alpha$ doublet instead of $K\alpha_1$ and $K\beta_1$ radiation. Owing to the change, they could use only one detector (Kishino's original method needs two detectors), but a special approach is sometimes needed to resolve two peaks that relate to the components of the doublet. A similar problem of separation of two peaks (recorded by two detectors) is reported by Cembali *et al.* (1992). By introducing a computer simulation of the reflecting curves (using a convolution model), the authors managed to determine the separation with an error of $0.01''$ and to achieve a precision of some parts in 10^7. The same precision is reported by Fukumori, Imai, Hasegawa & Akashi (1997), who introduced a precise positioning device and a position-sensitive proportional counter to their instrument.

As in the other multiple-crystal methods, the most important experimental problem is accurate crystal setting. Larson (1974), as a result of detailed analysis, gave the dependence between the angular separation of two peaks and angles characterizing misalignment of the first and second crystals.

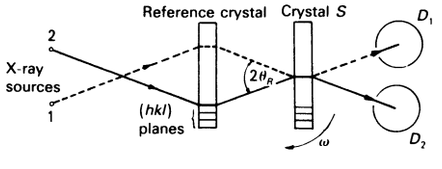

Fig. 5.3.3.12. Schematic representation of the double-beam comparator of Hart (1969).

5.3.3.7.4. *Combined methods*

The idea of multiple-beam measurement (§5.3.3.7.3) can be applied to other arrangements that combine the features of the double-beam comparator with those of the triple-crystal spectrometer; there are additional advantages in such a system.

The application of the double-beam technique makes it possible to realize a triple-reflection scheme for comparing lattice parameters on the basis of a double-axis spectrometer. The arrangement proposed by Ando, Bailey & Hart (1978), shown in Fig. 5.3.3.13, consists of a sample and a reference crystal, which are made from the same material but differ in purity (or strain, stoichiometry, vacancy concentration, *etc*). The angular difference $\Delta\theta$ in the Bragg angles of the sample and the reference crystal, θ_S and θ_R, respectively,

$$\Delta\theta = \theta_S - \theta_R, \tag{5.3.3.45a}$$

is measured as the sample angle $\Delta\omega$ between the double-reflected peak D and the triply diffracted peak T:

$$\Delta\theta = \Delta\omega, \tag{5.3.3.45b}$$

$$\Delta\omega = \omega_D - \omega_T. \tag{5.3.3.45c}$$

Assuming that $\Delta\theta$ is entirely due to changes Δd in atomic spacings, the authors use the following relation for determination of the latter:

$$\Delta d/d = -\cot\theta\Delta\theta. \tag{5.3.3.46}$$

The experimental requirements are simple and inexpensive, owing to simple shapes of both the reference crystal and the sample crystal, so that the measurement can be made quickly. By combining the two reference crystals into a single monolithic reference crystal, excellent stability, difficult to achieve with triple-axis arrangements (*cf.* §5.3.3.7.2), is obtained at the same time. The disadvantage of the method is that it covers a smaller range of lattice parameters than the other double-beam methods (Hart, 1969; Larson, 1974) described in §5.3.3.7.3. A new version of the double-crystal triple-reflection scheme (Häusermann & Hart, 1990) allows one to achieve a precision of 1 part in 10^8 in 2 min of measurement time, which includes the data analysis; 30 min are needed to change the sample. Errors due to the crystal tilt and thermal drifts are considered.

Another example of the triple-reflection scheme realized by means of the double-beam technique has been presented by Kovalchuk, Kovev & Pinsker (1975), who realized the triple-crystal arrangement on the basis of a double-crystal spectrometer by parallel mounting of the two crystals to be compared (the sample and the reference crystal) on one common axis. The advantage of this system is that Bragg angles as high as 80° are available. The device can be applied in studies of the real structure of a single crystal.

High-sensitivity ($\Delta d/d$ up to $\pm 3 \times 10^{-8}$) lattice-parameter-comparison measurement over a wide range of temperatures can be performed by means of the triple-crystal (more accurately, triple-axis) X-ray spectrometer realized by Buschert, Pace, Inzaghi & Merlini (1980). The arrangement (Fig. 5.3.3.14) consists of four crystals. The first is used for obtaining a very wide but extremely parallel exit beam, which is incident on both the standard crystal S and an unknown crystal X, placed side by side on a common axis in the cryostat. The reflected beams from S and X are recorded by partially transmitting detectors DA_2 and DB_2, so that the beams reflect from the third crystal and are detected by the counters DA_3 and DB_3. There is a small, sensitive, angle adjustment to rotate the crystal X with respect to the standard S and it is used to bring the peaks of S and X into approximate coincidence. The angular difference in the peak positions on the third axis is used for determination of lattice-parameter changes from (5.3.3.46), so that

$$\Delta\theta = \Delta\theta_3/2 - \Delta\theta_2, \tag{5.3.3.47}$$

where $\Delta\theta_2$ and $\Delta\theta_3$ are the differences in peak positions at axes (2) and (3), respectively. The device was used, for example, to study the effect of isotope concentration on the lattice parameter of germanium perfect crystals (Buschert, Merlini, Pace, Rodriguez & Grimsditch, 1988). The measured differences in the lattice parameter, of the order of 1 part in 10^5, were compared with those evaluated theoretically, and a very good agreement was obtained.

Another variant of a multiple-beam arrangement, based on a triple-crystal spectrometer, was proposed by Kubena & Holý (1988). The authors compared the distances of lattice planes in a direction perpendicular to the surface of the sample while studying the growth striations. One well collimated and monochromated beam coming from the first crystal was directed into the sample, and then two beams – one transmitted and one diffracted in the sample – diffracted in the reference crystal. Intensities of the diffracted beams were measured by two detectors. The difference of lattice spacings of the sample and the reference crystal was determined from the difference in positions of respective peaks. The accuracy of the lattice-spacing comparison of 2 parts in 10^7 and the precision of 1 part in 10^7 were obtained.

A four-crystal six-reflection diffractometer (Fewster, 1989) was built to study crystals distorted by epitaxy and defects in nearly perfect crystals. Fig. 5.3.3.15 is a schematic diagram of this device. The two-crystal four-reflection Bartels monochromator (Bartels, 1983) defines a narrow reflectivity profile. The analyser selects the angular range diffracted from the sample. The device may be used for recording both near-perfect rocking curves from distorted crystals (when rotations of the sample and the analyser are coupled) and a diffraction-space map for studying the diffuse scattering (when the two rotations are

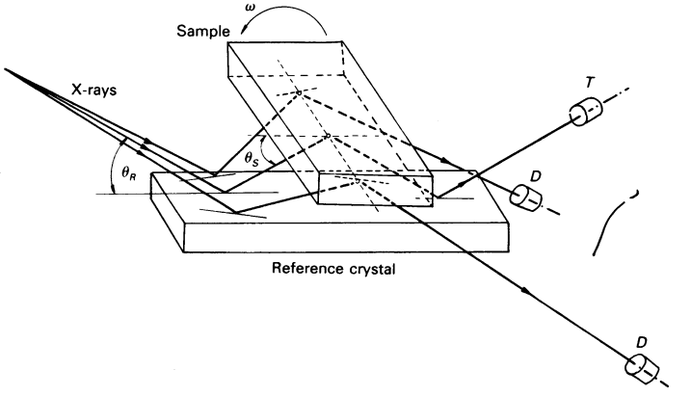

Fig. 5.3.3.13. The double-axis lattice-spacing comparator of Ando, Bailey & Hart (1978); a triple-diffracted beam is used.

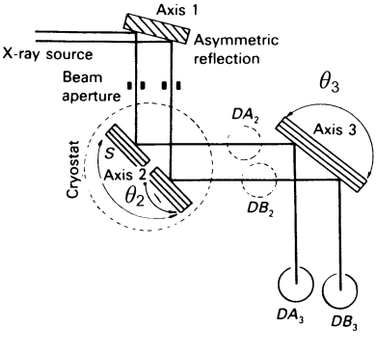

Fig. 5.3.3.14. Schematic representation of the double-beam triple-crystal spectrometer of Buschert *et al.* (1980).

uncoupled). Various applications of such high-sensitivity multiple-crystal X-ray spectrometers for reciprocal-space mapping and imaging (topography), which are outside the scope of the present paper, are reviewed by Fewster (1993, and references therein).

As was shown a few years later by Fewster & Andrew (1995), the device can also be used for absolute lattice-parameter measurements of single-crystal and polycrystalline materials with a relative accuracy of a few parts in 10^6. The authors checked the angular resolution and the sample centring of their instrument, and discussed systematic errors due to refraction, the Lorenz and polarization factor, the diffracting-plane tilt and the peak-position determination.

5.3.3.8. *Optical and X-ray interferometry – a non-dispersive technique*

The accuracy of an absolute measurement can be improved, in relation to that obtained in traditional methods (*cf.* Subsection 5.3.3.5), either if the wavelength of the radiation used in an experiment is known with better accuracy [*cf.* equation (5.3.1.3)] or if a high-quality standard single crystal is given, whose lattice spacing has been very accurately determined (Baker & Hart, 1975; mentioned in §5.3.3.7.3). The two tasks, *i.e.* very accurate determination of both lattice spacings and wavelengths in metric units, can be realized by use of combined optical and X-ray interferometry. This original concept of absolute-lattice-spacing determination directly in units of a standard light wavelength has been proposed and realized by Deslattes (1969) and Deslattes & Henins (1973).

The principle of the method is presented in Fig. 5.3.3.16. The silicon-crystal X-ray interferometer is a symmetric Laue-case type (Bonse & te Kaat, 1968). The parallel translation device consists of the stationary assembly (*a*) formed by two specially prepared crystals, and a moveable one (*b*), to which belongs the third crystal. One of the two mirrors of a high-resolution Fabry–Perot interferometer is attached to the stationary assembly and the second to the moving assembly. A stabilized He–Ne laser is used as a source of radiation, the wavelength of which has been established relative to visible standards. The first two crystals produce a standing wavefield, which is intercepted by the third crystal, so that displacement of the third crystal parallel to the diffraction vector (as suggested by the large arrow) produces alternate maxima and minima in the diffracted beams, detected by X-ray detector (*c*). Resonant transmission maxima of the optical interferometer are detected simultaneously by the photomultiplier indicated at (*d*). Analysis of the fringes (shown in Fig. 5.3.3.17) is the basis for the calculation of the lattice-spacing-to-optical-wavelength ratio (d/λ), which is given by

$$\frac{2d}{\lambda} = \frac{n}{m}\,\frac{\cos\alpha}{\cos\beta}, \qquad (5.3.3.48)$$

where n and m are the numbers of optical and X-ray diffraction fringes, respectively, and α and β are the measured angular deviations of the optical and X-ray diffraction vectors from the direction of motion. The measurements are carried out in two steps. First, the lattice parameter of silicon along the [110] crystallographic direction was measured in the metric system, independently of the X-ray wavelength used in the experiment. As the next step, a specimen of known lattice spacing, treated as a reference crystal, was used for the accurate wavelength determination of Cu $K\alpha_1$ and Mo $K\alpha_1$. Accuracy better than 1 part in 10^6 was reported (see Section 4.2.2).

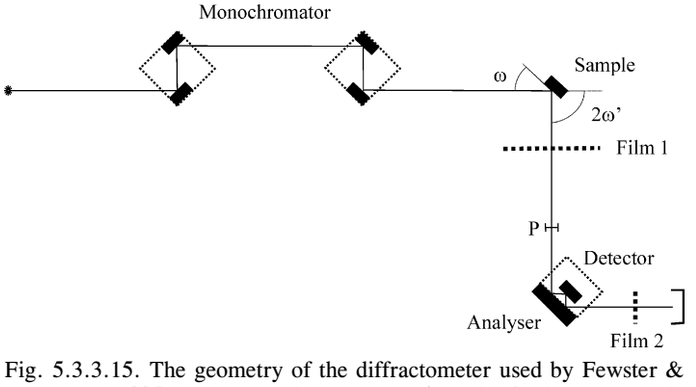

Fig. 5.3.3.15. The geometry of the diffractometer used by Fewster & Andrew (1995). The scattering angle, $2\omega'$, is the fundamental angle for determination of the interplanar spacing and P is the analyser-groove entrance.

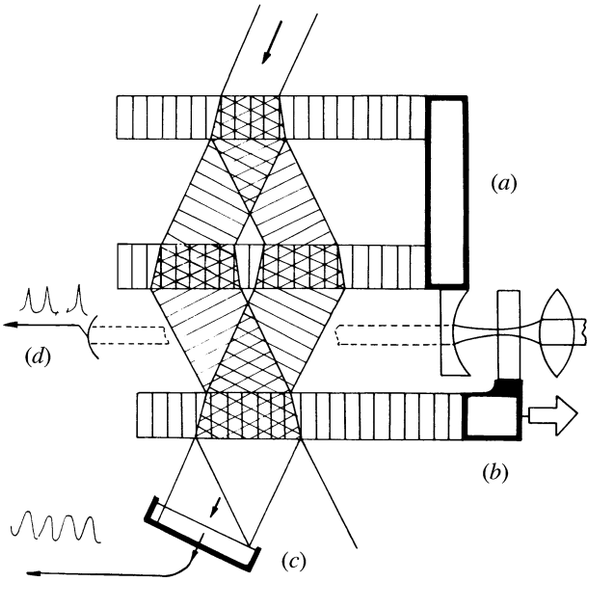

Fig. 5.3.3.16. Optical and X-ray interferometry. Schematic representation of the experimental set-up (after Deslattes & Henins, 1973; Becker *et al.*, 1981).

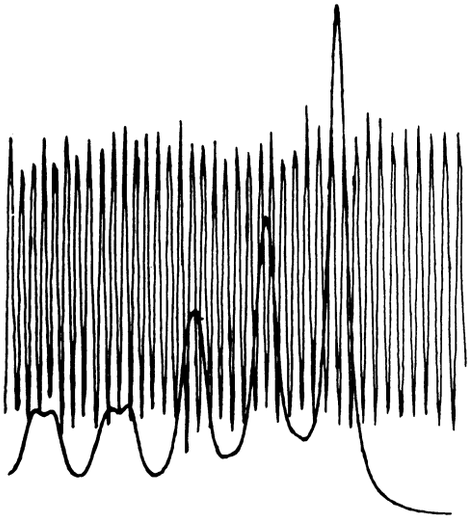

Fig. 5.3.3.17. Portion of a dual-channel recording of X-ray and optical fringes (Deslattes, 1969).

references