

5. DYNAMICAL THEORY AND ITS APPLICATIONS

measured by Lambert & Malgrange (1982). The large angular amplification associated with the curvature of the dispersion surfaces near the exact Bragg setting was demonstrated by Kikuta *et al.* (1975) and by Zeilinger & Shull (1979).

In magnetic crystals, the investigations have been restricted to the simpler geometry where the scattering vector is perpendicular to the magnetization, and to few materials. *Pendellösung* behaviour was evidenced through the variation with wavelength of the flipping ratio for polarized neutrons by Baruchel *et al.* (1982) on an yttrium iron garnet sample, with the geometry selected so that the defects would not affect the Bragg reflection used. The inclination method was used successfully by Zelepukhin *et al.* (1989), Kvardakov & Somenkov (1990) and Kvardakov *et al.* (1990a) for the weak ferromagnet FeBO₃, and in the room-temperature weak-ferromagnetic phase of hematite, α -Fe₂O₃, by Kvardakov *et al.* (1990b) and Kvardakov & Somenkov (1992).

Experiments on the influence of defects in nearly perfect crystals have been performed by several groups. The effect on the rocking curve was investigated by Eichhorn *et al.* (1967), the intensities were measured by Lambert & Malgrange (1982) and by Albertini, Boeuf, Cesini *et al.* (1976), and the influence on the *Pendellösung* behaviour was discussed by Kvardakov & Somenkov (1992). Boeuf & Rustichelli (1974) and Albertini *et al.* (1977) investigated silicon crystals curved by a thin surface silicon nitride layer.

Many experiments have been performed on vibrating crystals; reviews are given by Michalec *et al.* (1988) and by Kulda *et al.* (1988). Because the velocity of neutrons is of the same order of magnitude as the velocity of acoustic phonons in crystals, the effect of ultrasonic excitation on dynamical diffraction takes on some original features compared to the X-ray case (Iolin & Entin, 1983); they could to some extent be evidenced experimentally (Iolin *et al.*, 1986; Chalupa *et al.*, 1986). References to experimental work on neutron scattering by imperfect crystals under ultrasonic excitation are included in Zolotoyabko & Sander (1995).

Some experiments with no equivalent in the X-ray case could be performed. The very strong incoherent scattering of neutrons by protons, very different physically but similar in its effect to absorption, was also shown to lead to anomalous transmission effects by Sippel & Eichhorn (1968). Because the velocity of thermal neutrons in a vacuum is five orders of magnitude smaller than the velocity of light, the flight time for neutrons undergoing Bragg scattering in Laue geometry in a perfect crystal could be measured directly (Shull *et al.*, 1980). The effect of externally applied fields was measured experimentally for magnetic fields by Zeilinger & Shull (1979) and Zeilinger *et al.* (1986). Slight rotation of the crystal, introducing a Coriolis force, was used by Raum *et al.* (1995), and gravity was tested recently, with the spectacular result that some states are accelerated upwards (Zeilinger, 1995).

5.3.7. Applications of the dynamical theory of neutron scattering

5.3.7.1. Neutron optics

Most experiments in neutron scattering require an intensity-effective use of the available beam at the cost of relatively high divergence and wavelength spread. The monochromators must then be imperfect ('mosaic') crystals. In some cases, however, it is important to have a small divergence and wavelength band. One example is the search for small variations in neutron energy in inelastic scattering without the use of the neutron spin-echo principle. Perfect crystals must then be used as monochromators or analysers, and dynamical diffraction is directly involved. As in the X-ray case, special designs can lead to strong decrease in the intensity of harmonics, *i.e.* of contributions of $\lambda/2$ or $\lambda/3$ (Hart & Rodrigues, 1978). The possibility of focusing neutron beams by the use of perfect crystals with the incident beam spatially modulated in amplitude through an absorber, or in phase through

an appropriate patterning of the surface, in analogy with the Bragg–Fresnel lenses developed for X-rays, was suggested by Indenbom (1979).

The use of two identical perfect crystals in nondispersive (+, −, ||) setting provides a way of measuring the very narrow intrinsic rocking curves expected from the dynamical theory. Any divergence added between the two crystals can be sensitively measured. Thus perfect crystals provide interesting possibilities for measuring very small angle neutron scattering. This was performed by Takahashi *et al.* (1981, 1983) and Tomimitsu *et al.* (1986) on amorphous materials, and by Kvardakov *et al.* (1987) for the investigation of ferromagnetic domains in bulk silicon–iron specimens under stress, both through the variations in transmission associated with refraction on the domain walls and through small-angle scattering. Imaging applications are described in Section 5.3.7.4. Badurek *et al.* (1979) used the different deflection of the two polarization states provided by a magnetic prism placed between two perfect silicon crystals to produce polarized beams.

Curved almost-perfect crystals or crystals with a gradient in the lattice spacing can provide focusing (Albertini, Boeuf, Lagomarsino *et al.*, 1976) and vibrating crystals can give the possibility of tailoring the reflectivity of crystals, as well as of modulating beams in time (Michalec *et al.*, 1988). A double-crystal arrangement with bent crystals was shown by Eichhorn (1988) to be a flexible small-angle-neutron-scattering device.

5.3.7.2. Measurement of scattering lengths by *Pendellösung* effects

As in X-ray diffraction, *Pendellösung* oscillations provide an accurate way of measuring structure factors, hence coherent neutron scattering lengths. The equal-thickness fringes expected from a wedge-shaped crystal were observed by Kikuta *et al.* (1971). Three kinds of measurements were made. Sippel *et al.* (1965) measured as a function of thickness the integrated reflectivity from a perfect crystal of silicon, the thickness of which they varied by polishing after each measurement, obtaining a curve similar to Fig. 5.1.6.7, corresponding to equation 5.1.6.8. Shull (1968) restricted the measurement to wavefields that propagated along the reflecting planes, hence at exact Bragg incidence, by setting fine slits on the entrance and exit faces of 3 to 10 mm-thick silicon crystals, and measured the oscillation in diffracted intensity as he varied the wavelength of the neutrons used by rotating the crystal. Shull & Oberteuffer (1972) showed that a better interpretation of the data, when the beam is restricted to a fine slit, corresponds to the spherical wave approach (actually cylindrical wave), and the boundary conditions were discussed more generally by Arthur & Horne (1985). Somenkov *et al.* (1978) developed the inclination method, in which the integrated reflectivity is measured as the effective crystal thickness is varied nondestructively, by rotating the crystal around the diffraction vector, and used it for germanium. Belova *et al.* (1983) discuss this method in detail. The results obtained by this group for magnetic crystals are mentioned in Section 5.3.6. Structure-factor values for magnetic reflections were obtained by Kvardakov *et al.* (1995) for the weak ferromagnet FeBO₃.

5.3.7.3. Neutron interferometry

Because diffraction by perfect crystals provides a well defined distribution of the intensity and phase of the beam, interferometry with X-rays or neutrons is possible using ingeniously designed and carefully manufactured monolithic devices carved out of single crystals of silicon. The technical and scientific features of this family of techniques are well summarized by Bonse (1979, 1988), as well as other papers in the same volumes, and by Shull (1986).

X-ray interferometry started with the Bonse–Hart interferometer (Bonse & Hart, 1965). A typical device is the LLL skew-symmetric interferometer, where the L's stand for Laue,

indicating transmission geometry in all crystal slabs. In these slabs, which can be called the splitter, the mirrors and the recombiner, the same pair of opposite reflections, in symmetrical Laue geometry, is used three times. In the first slab, the incident beam is coherently split into a transmitted and a diffracted beam. Each of these is then diffracted in the two mirrors, and the resulting beams interfere in the recombiner, again yielding a forward-diffracted and a diffracted beam, the intensities of both of which are measured. This version, the analogue of the Mach-Zehnder interferometer in optics, offers a sizeable space (several cm of path length) where two coherent parallel beams can be submitted to various external actions. Shifting the relative phase of these beams (*e.g.* by π , introducing an optical path-length difference of $\lambda/2$) results in the intensities of the outgoing beams changing from a maximum to a minimum.

Applications of neutron interferometry range from the very useful to the very exotic. The most useful one is probably the measurement of coherent neutron scattering lengths. Unlike the *Pendellösung* method described in Section 5.3.7.2, this method does not require the measured samples to be perfect single crystals, nor indeed crystals. Placing a slab of material across one of the beams and rotating it will induce an optical path-length difference of $(1 - n)t$ if t is the effective thickness along the beam, hence a phase shift of $2\pi(1 - n)t/\lambda$. With the expression of the refractive index n as given in Section 5.3.2.2, it is clear that for an isotopically pure material the scattering length b_{coh} can be deduced from the measurement of intensity *versus* the rotation angle of the phase shifter. This is a very versatile and much used method. The decrease in oscillation contrast can be used to obtain information of relevance to materials science, such as statistical properties of magnetic domain distributions (Korpiun, 1966) or precipitates (Rauch & Seidl, 1987); Rauch (1995) analyses the effect in terms of the neutron coherence function.

Many elegant experiments have been performed with neutron interferometers in efforts to set an upper limit to effects than can be considered as nonexistent, or to test expectations of basic quantum physics. Many papers are found in the same volumes as Bonse (1979) and Bonse (1988); excellent reviews have been given by Klein & Werner (1983), Klein (1988) and Werner (1995). Among the topics investigated are the effect of gravity (Colella *et al.*, 1975), the Sagnac effect, *i.e.* the influence of the Earth's rotation (Werner *et al.*, 1979), the Fizeau effect, *i.e.* the effect of the movement of the material through which the neutrons are transmitted (Arif *et al.*, 1988) and the Aharonov–Casher effect, *i.e.* the dual of the Aharonov–Bohm effect for neutral particles having a magnetic moment (Cimmino *et al.*, 1989).

5.3.7.4. Neutron diffraction topography and other imaging methods

These are the neutron form of the 'topographic' or diffraction imaging techniques, in which an image of a single crystal is obtained through the local variations in Bragg-diffracted intensity due to inhomogeneities in the sample. It is briefly described in Chapter 2.8 of *IT C*. It was pioneered by Doi *et al.* (1971) and by Ando & Hosoya (1972). Like its X-ray counterpart, neutron topography can reveal isolated defects, such as dislocations (Schlenker *et al.*, 1974; Malgrange *et al.*, 1976). Because of the small neutron fluxes available, it is not very convenient for this purpose, since the resolution is poor or the exposure times are very long. On the other hand, the very low absorption of neutrons in most materials makes it quite convenient for observing the gross defect structure in samples that would be too absorbing for X-rays (Tomimitsu & Doi, 1974; Baruchel *et al.*, 1978; Tomimitsu *et al.*, 1983; Kvardakov *et al.*, 1992), or the spatial modulation of distortion due to vibration, for example in quartz (Michalec *et al.*, 1975), and resonant magnetoelastic effects (Kvardakov & Somenkov, 1991). In particular, virtual slices of bulky as-grown samples can be investigated without cutting them using neutron

section topography or neutron tomography (Schlenker *et al.*, 1975; Davidson & Case, 1976).

Neutron topography also shows the salient dynamical interference effect, *viz Pendellösung*, visually, in the form of fringes (Kikuta *et al.*, 1971; Malgrange *et al.*, 1976; Tomimitsu & Zeyen, 1978). Its unique feature, however, is the possibility of observing and directly characterizing inhomogeneities in the magnetic structure, *i.e.* magnetic domains of all kinds [ferromagnetic domains (Schlenker & Shull, 1973) and antiferromagnetic domains of various sorts (Schlenker & Baruchel, 1978), including spin-density wave domains (Ando & Hosoya, 1972, 1978; Davidson *et al.*, 1974), 180° or time-reversed domains in some materials and helimagnetic or chirality domains (Baruchel *et al.*, 1990)], or coexisting phases at a first-order phase transition (Baruchel, 1989). In such cases, the contrast is primarily due to local variations in the structure factor, a situation very unusual in X-ray topography, and good crystal quality, leading to dynamical scattering behaviour, is essential in the observation process only in a few cases (Schlenker *et al.*, 1978). It is often crucial, however, for making the domain structure simple enough to be resolved, in particular in the case of antiferromagnetic domains.

Imaging can also be performed for samples that need be neither crystals nor perfect. Phase-contrast imaging of a specimen through which the neutrons are transmitted can be performed in a neutron interferometer. It has been shown to reveal thickness variations by Bauspiess *et al.* (1978) and ferromagnetic domains by Schlenker *et al.* (1980). The same papers showed that phase edges show up as contrast when one of the interferometer paths is blocked, *i.e.* when the sample is placed effectively between perfect, identical crystals set for diffraction in a nondispersive setting. Under the name of neutron radiography with refraction contrast, this technique, essentially a form of Schlieren imaging, was further developed by Podurets, Somenkov & Shil'shtein (1989), Podurets, Somenkov, Chistyakov & Shil'shtein (1989), and Podurets *et al.* (1991), who were able to image internal ferromagnetic domain walls in samples 10 mm thick.

References

- Albertini, G., Boeuf, A., Cesini, G., Mazkedian, S., Melone, S. & Rustichelli, F. (1976). *A simple model for dynamical neutron diffraction by deformed crystals*. *Acta Cryst.* **A32**, 863–868.
- Albertini, G., Boeuf, A., Klar, B., Lagomarsino, S., Mazkedian, S., Melone, S., Puliti, P. & Rustichelli, F. (1977). *Dynamical neutron diffraction by curved crystals in the Laue geometry*. *Phys. Status Solidi A*, **44**, 127–136.
- Albertini, G., Boeuf, A., Lagomarsino, S., Mazkedian, S., Melone, S. & Rustichelli, F. (1976). *Neutron properties of curved monochromators*. *Proceedings of the Conference on Neutron Scattering, Gallinburg, Tennessee*, USERDA CONF 760601-P2, 1151–1158. Oak Ridge, Tennessee: Oak Ridge National Laboratory.
- Al Haddad, M. & Becker, P. J. (1988). *On the statistical dynamical theory of diffraction: application to silicon*. *Acta Cryst.* **A44**, 262–270.
- Ando, M. & Hosoya, S. (1972). *Q-switch and polarization domains in antiferromagnetic chromium observed with neutron-diffraction topography*. *Phys. Rev. Lett.* **29**, 281–285.
- Ando, M. & Hosoya, S. (1978). *Size and behavior of antiferromagnetic domains in Cr directly observed with X-ray and neutron topography*. *J. Appl. Phys.* **49**, 6045–6051.
- Arif, M., Kaiser, H., Clothier, R., Werner, S. A., Berliner, R., Hamilton, W. A., Cimmino, A. & Klein, A. G. (1988). *Fizeau effect for neutrons passing through matter at a nuclear resonance*. *Physica B*, **151**, 63–67.
- Arthur, J. & Horne, M. A. (1985). *Boundary conditions in dynamical neutron diffraction*. *Phys. Rev. B*, **32**, 5747–5752.
- Bacon, G. E. & Lowde, R. D. (1948). *Secondary extinction and neutron crystallography*. *Acta Cryst.* **1**, 303–314.
- Badurek, G., Rauch, H., Wilfling, A., Bonse, U. & Graeff, W. (1979). *A perfect-crystal neutron polarizer as an application of magnetic prism refraction*. *J. Appl. Cryst.* **12**, 186–191.
- Baruchel, J. (1989). *The contribution of neutron and synchrotron radiation topography to the investigation of first-order magnetic phase transitions*. *Phase Transit.* **14**, 21–29.