International Tables for Crystallography (2006). Vol. B, Section 5.3.3.3, pp. 560–561.

These expressions are the sum of the spin-flip and non-spin-flip cross sections, which are equal to $(b \pm p\sin^2\beta)^2$ and $(p\sin\beta\cos\beta)^2$, respectively. In the case of non-polarized neutrons, the interference term $(\pm 2bp\sin^2\beta)$ between the nuclear and the magnetic scattering disappears; the cross section is then

$$(\mathrm{d}\sigma/\mathrm{d}\Omega) = b^2 + (p\sin\beta)^2. \qquad (5.3.3.8)$$

In the general case of a partially polarized beam we can use the density-matrix representation. Let $\rho_{\rm inc}$ be the density matrix of the incident beam; it can be shown that the density matrix of the diffracted beam is equal to the following product of matrices: $(\mathrm{q})\rho_{\rm inc}(\mathrm{q}^*)$. Using the relations between the density matrix and polarization vector presented in the preceding section, we can obtain a general description of the diffracted beam as a function of the polarization properties of the incident beam. Such a formalism is of interest for dealing with new experimental arrangements, in which a three-dimensional polarization analysis of the diffracted beam is possible, as shown by Tasset (1989).

5.3.3.3. *Dynamical theory in the case of perfect ferromagnetic or collinear ferrimagnetic crystals*

The most direct way to develop this dynamical theory in the two-beam case, which involves a single Bragg-diffracted beam of diffraction vector **h**, is to consider spinor wavefunctions of the following form:

$$\varphi(\mathbf{r}) = \exp(i\mathbf{K_0}\cdot\mathbf{r})\begin{pmatrix} D_0 \\ E_0 \end{pmatrix} + \exp[i(\mathbf{K_0}+\mathbf{h})\mathbf{r}]\begin{pmatrix} D_h \\ E_h \end{pmatrix} \qquad (5.3.3.9)$$

as approximate solutions of the wave equation inside the crystal,

$$\Delta\varphi(\mathbf{r}) + k^2\varphi(\mathbf{r}) = [u(\mathbf{r}) - \boldsymbol{\sigma}\cdot\mathbf{Q}(\mathbf{r})]\varphi(\mathbf{r}), \qquad (5.3.3.10)$$

where $u(\mathbf{r})$ and $-\boldsymbol{\sigma}\cdot\mathbf{Q}(\mathbf{r})$ are, respectively, equal to the nuclear and the magnetic potential energies multiplied by $2m/\hbar^2$. In the calculation of $\varphi(\mathbf{r})$ in the two-beam case, we need only three terms in the expansions of the functions $u(\mathbf{r})$ and $\mathbf{Q}(\mathbf{r})$ into Fourier series:

$$u(\mathbf{r}) = u_\mathbf{0} + u_\mathbf{h}\exp(i\mathbf{h}\cdot\mathbf{r}) + u_{-\mathbf{h}}\exp(-i\mathbf{h}\cdot\mathbf{r}) + \ldots,$$
$$\mathbf{Q}(\mathbf{r}) = \mathbf{Q_0} + \mathbf{Q_h}\exp(i\mathbf{h}\cdot\mathbf{r}) + \mathbf{Q_{-h}}\exp(-i\mathbf{h}\cdot\mathbf{r}) + \ldots.$$

We suppose that the crystal is magnetically saturated by an externally applied magnetic field $\mathbf{H}_a$. $\mathbf{Q_0}$ is then proportional to the macroscopic mean magnetic field $\mathbf{B} = \mu_0(\mathbf{M} + \mathbf{H}_a + \mathbf{H}_d)$, where **M** is the magnetization vector and $\mathbf{H}_d$ is the demagnetizing field. The results of Section 5.3.3.2 show that $\mathbf{Q_h}$ and $\mathbf{Q_{-h}}$ are proportional to the projection of **M** on the reflecting plane.

The four coefficients D_0, D_h, E_0 and E_h of (5.3.3.9) are found to satisfy a system of four homogeneous linear equations. The condition that the associated determinant has to be equal to 0 defines the dispersion surface, which is of order 4 and has four branches. An incident plane wave thus excites a system of four wavefields of the form of (5.3.3.9), generally polarized in various directions. A particular example of a dispersion surface, having an unusual shape, is shown in Fig. 5.3.3.1.

This is a much more complicated situation than in the case of non-magnetic crystals, in which one only needs to consider scalar wavefunctions which depend on two coefficients, such as D_0 and D_h, and which are related to hyperbolic dispersion surfaces of order 2, as fully described in Chapter 5.1 on X-ray diffraction.

In fact, all neutron experiments related to dynamical effects in diffraction by magnetic crystals have been performed under such conditions that the magnetization vector in the crystal is perpendicular to the diffraction vector **h**. In this case, the vectors $\mathbf{Q_h}$ and $\mathbf{Q_{-h}}$ are parallel or antiparallel to the vector $\mathbf{Q_0}$ which is chosen as the spin-quantization axis. The matrices $\boldsymbol{\sigma}\cdot\mathbf{Q_0}$, $\boldsymbol{\sigma}\cdot\mathbf{Q_h}$ and $\boldsymbol{\sigma}\cdot\mathbf{Q_{-h}}$ are then all diagonal matrices, and we obtain for the

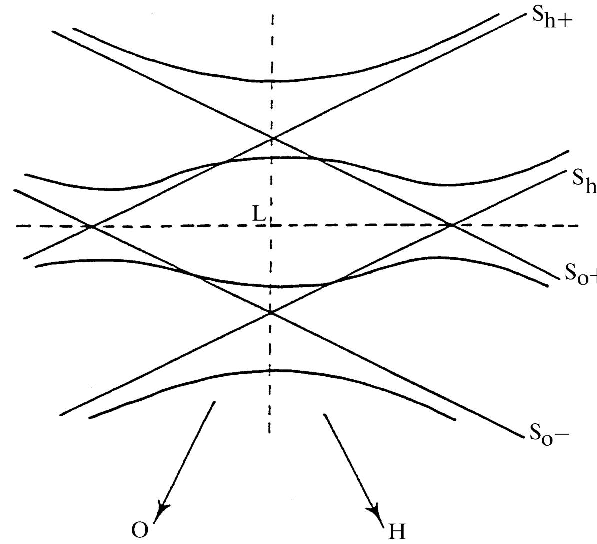

Fig. 5.3.3.1. Schematic plot of the two-beam dispersion surface in the case of a purely magnetic reflection such that $\mathbf{Q_h} = \mathbf{Q_{-h}} = \mathbf{Q_0}$ and that the angle between $\mathbf{Q_0}$ and $\mathbf{Q_h}$ is equal to $\pi/4$.

two spin states (±) separate dynamical equations which are similar to the dynamical equations for the scalar case, but with different structure factors, which are either the sum or the difference of the nuclear structure factor F_N and of the magnetic structure factor F_M:

$$F_+ = F_N + F_M \text{ and } F_- = F_N - F_M. \qquad (5.3.3.11)$$

F_N and F_M are related to the scattering lengths of the ions in the unit cell of volume V_c:

$$F_N = V_c u_\mathbf{h} = \sum_i b_i \exp(-i\mathbf{h}\cdot\mathbf{r}_i);$$
$$F_M = V_c|\mathbf{Q_h}| = -\frac{\mu_0 m}{2h^2}\mu_n\boldsymbol{\sigma}\cdot\sum_i \boldsymbol{\mu}_{i\perp}(\mathbf{h})f_i\left(\frac{\sin\theta}{\lambda}\right)\exp(-i\mathbf{h}\cdot\mathbf{r}_i).$$

The dispersion surface of order 4 degenerates into two hyperbolic dispersion surfaces, each of them corresponding to one of the polarization states (±). The asymptotes are different; this is related to different values of the refractive indices for neutron polarization parallel or antiparallel to $\mathbf{Q_0}$.

In some special cases the magnitudes of F_N and F_M happen to be equal. Only one polarization state is then reflected. Magnetic crystals with such a property (reflections 111 of the Heusler alloy Cu_2MnAl, or 200 of the alloy Co–8% Fe) are very useful as polarizing monochromators and as analysers of polarization.

If the scattering vector **h** is in the same direction as the magnetization, this reflection is a purely nuclear one (with no magnetic contribution), since F_M is then equal to 0. Purely magnetic reflections (without nuclear contribution) also exist if the magnetic structure involves several sublattices.

If **h** is neither perpendicular to the average magnetization nor in the same direction, the presence of non-diagonal matrices in the dynamical equations cannot be avoided. The dynamical theory of diffraction by perfect magnetic crystals then takes the complicated form already mentioned.

Theoretical discussions of this complicated case of dynamical diffraction have been given by Stassis & Oberteuffer (1974), Mendiratta & Blume (1976), Sivardière (1975), Belyakov & Bokun (1975, 1976), Schmidt *et al.* (1975), Bokun (1979), Guigay & Schlenker (1979*a*,*b*), and Schmidt (1983). However, to our knowledge, only limited experimental work has been carried out on this subject. Successful experiments could only be performed for the simpler cases mentioned above.

5.3.3.4. *The dynamical theory in the case of perfect collinear antiferromagnetic crystals*

In this case, there is no average magnetization ($\mathbf{Q_0} = 0$). It is then convenient to choose the quantization axis in the direction of $\mathbf{Q_h}$ and $\mathbf{Q_{-h}}$. The dispersion surface degenerates into two hyperbolic surfaces corresponding to each polarization state along this direction for any orientation of the diffraction vector relative to the direction of the magnetic moments of the sublattices. These two hyperbolic dispersion surfaces have the same asymptotes. Furthermore, in the case of a purely magnetic reflection, they are identical.

The possibility of observing a precession of the neutron polarization in the presence of diffraction, in spite of the fact that there is no average magnetization, has been pointed out by Baryshevskii (1976).

5.3.3.5. *The flipping ratio*

In polarized neutron diffraction by a magnetically saturated magnetic sample, it is usual to measure the ratio of the reflected intensities I_+ and I_- measured when the incident beam is polarized parallel or antiparallel to the magnetization in the sample. This ratio is called the flipping ratio,

$$R = I_+/I_-, \qquad (5.3.3.12)$$

because its measurement involves flipping the incident-beam polarization to the opposite direction. This is an experimentally well defined quantity, because it is independent of a number of parameters such as the intensity of the incident beam, the temperature factor or the coefficient of absorption. In the case of an ideally imperfect crystal, we obtain from the kinematical expressions of the integrated reflectivities

$$R_{\rm kin}(\mathbf{h}) = (I_+/I_-)_{\rm kin} = \left(\frac{|F_N + F_M|}{|F_N - F_M|}\right)^2. \qquad (5.3.3.13)$$

In the case of an ideally perfect thick crystal, we obtain from the dynamical expressions of the integrated reflectivities

$$R_{\rm dyn}(\mathbf{h}) = (I_+/I_-)_{\rm dyn} = \frac{|F_N + F_M|}{|F_N - F_M|}. \qquad (5.3.3.14)$$

In general, $R_{\rm dyn}$ depends on the wavelength and on the crystal thickness; these dependences disappear, as seen from (5.3.3.14), if the path length in the crystal is much larger than the extinction distances for the two polarization states. It is clear that the determination of $R_{\rm kin}$ or $R_{\rm dyn}$ allows the determination of the ratio F_M/F_N, hence of F_M if F_N is known. In fact, because real crystals are neither ideally imperfect nor ideally perfect, one usually introduces an extinction factor y (extinction is discussed below, in Section 5.3.4) in order to distinguish the real crystal reflectivity from the reflectivity of the ideally imperfect crystal. Different extinction coefficients y_+ and y_- are actually expected for the two polarization states. This obviously complicates the task of the determination of F_M/F_N.

In the kinematical approximation, the flipping ratio does not depend on the wavelength, in contrast to dynamical calculations for hypothetically perfect crystals (especially for the Laue case of diffraction). Therefore, an experimental investigation of the wavelength dependence of the flipping ratio is a convenient test for the presence of extinction. Measurements of the flipping ratio have been used by Bonnet *et al.* (1976) and by Kulda *et al.* (1991) in order to test extinction models. Baruchel *et al.* (1986) have compared nuclear and magnetic extinction in a crystal of MnP.

Instead of considering only the ratio of the integrated reflectivities, it is also possible to record the flipping ratio as a function of the angular position of the crystal as it is rotated across the Bragg position. Extinction is expected to be maximum at the peak and the ratio measured on the tails of the rocking curve may approach the kinematical value. It has been found experimentally that this expectation is not of general validity, as discussed by Chakravarthy & Madhav Rao (1980). It would be valid in the case of a perfect crystal, hence in the case of pure primary extinction. It would also be valid in the case of secondary extinction of type I, but not in the case of secondary extinction of type II [following Zachariasen (1967), type II corresponds to mosaic crystals such that the diffraction pattern from each block is wider than the mosaic statistical distribution].

5.3.4. Extinction in neutron diffraction (non-magnetic case)

The kinematical approximation, which corresponds to the first Born approximation in scattering theory, supposes that each incident neutron can be scattered only once and therefore neglects the possibility that the neutrons may be scattered several times. Because this is a simple approximation which overestimates the crystal reflectivity, the actual reduction of reflectivity, as compared to its kinematical value, is termed *extinction.* This is actually a typical dynamical effect, since it is a multiple-scattering effect.

Extinction effects can be safely neglected in the case of scattering by very small crystals; more precisely, this is possible when the path length of the neutron beam in the crystal is much smaller than $\Delta = V_c/\lambda F$, where λ is the neutron wavelength and F/V_c is the scattering length per unit volume for the reflection considered. Δ is sometimes called the ‘extinction distance’.

A very important fact is that extinction effects also vanish if the crystal is imperfect enough, because each plane-wave component of the incident beam can then be Bragg-reflected in only a small volume of the sample. This is the extinction-free case of ‘ideally imperfect crystals’. Conversely, extinction is maximum (smallest value of y) in the case of ideally perfect non-absorbing crystals.

Clearly, no significant extinction effects are expected if the crystal is thick but strongly absorbing, more precisely if the linear absorption coefficient μ is such that $\mu\Delta \gg 1$. Neutron diffraction usually corresponds to the opposite case ($\mu\Delta \ll 1$), in which extinction effects in nearly perfect crystals dominate absorption effects.

Extinction effects are usually described in the frame of the mosaic model, in which the crystal is considered as a juxtaposition of perfect blocks with different orientations. The relevance of this model to the case of neutron diffraction was first considered by Bacon & Lowde (1948). If the mosaic blocks are big enough there is extinction within each block; this is called *primary extinction.* Multiple scattering can also occur in different blocks if their misorientation is small enough. In this case, which is called *secondary extinction*, there is no phase coherence between the scattering events in the different blocks. The fact that empirical intensity-coupling equations are used in this case is based on this phase incoherence.

In the general case, primary and secondary extinction effects coexist. Pure secondary extinction occurs in the case of a mosaic crystal made of very small blocks. Pure primary extinction is observed in diffraction by perfect crystals.

The parameters of the mosaic model are the average size of the perfect blocks and the angular width of their misorientation distribution. The extinction theory of the mosaic model provides a relation between these parameters and the extinction coefficient,

references