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Theoretical Investigations of the Dynamical Neutron Diffraction by Magnetic Single Crystals

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Abstract

The fundamental equations for dynamical magnetic neutron diffraction including an external magnetic field are given. The reflecting power is calculated for some special cases for asymmetrical Laue geometry. Some *Pendellösung* effects are discussed. Analytical and numerical results for the integrated reflecting power are presented

I. Introduction

Dynamical neutron diffraction by magnetic crystals is treated by various authors (Ekstein, 1949; Stassis & Oberteuffer, 1974; Gusakov & Ruban, 1975; Belyakov & Bokun, 1975; Schmidt, Deimel & Daniel, 1975; Schmidt & Deimel, 1975; Sivardière, 1975; Schmidt & Deimel, 1976; Belyakov & Bokun, 1976; Mendiratta & Blume, 1976; Guigay & Schlenker, 1978). It is shown by Schmidt & Deimel (1975), Sivardière (1975), Schmidt & Deimel (1976), Belyakov & Bokun (1976), and Guigay & Schlenker (1978) that the magnetic diffraction depends on the angle φ between the average magnetic field $\mathbf{B}_A$ in the crystal and the Fourier transform $\mathbf{B}_H$ of the magnetic field in the crystal with respect to the scattering vector $\mathbf{H}$. The treatment of dynamical magnetic diffraction is very much simplified in the cases of $\mathbf{B}_A = 0$ or $\varphi = 0°$. The first explicit results for the wavevectors inside the crystal and the reflected and transmitted intensities for $\varphi \neq 0°$ and $\mathbf{B}_A \neq 0$ are given by Schmidt & Deimel (1976). Two special cases are treated in the work of Schmidt & Deimel (1976), the case of a vanishing nuclear contribution to the diffraction and the case of $\varphi = 90°$. This treatment is further restricted to the symmetrical Laue case and to crystals with a center of symmetry. Belyakov & Bokun (1976) show the fundamental equations for the general case and present results for the wavevectors inside the crystal in the symmetrical Laue case, with either $\varphi = 90°$, or a vanishing nuclear contribution to the diffraction and in the symmetrical Bragg case in general. Furthermore, for the symmetrical Bragg case and $|\mathbf{B}_A|/|\mathbf{B}_H| \gg 1$ results for the intensity and the polarization of the reflected beam are given. In the current work the treatment of Schmidt & Deimel (1976) for the case of vanishing nuclear diffraction will be generalized including a homogeneous magnetic field outside the crystal. Furthermore, for asymmetrical Laue geometry the reflected intensity will be given for some special cases. Finally, analytical and numerical results for the integrated reflecting power will be presented.

II. Fundamental equations

All the following results are obtained for the incidence of a plane neutron wave on an infinite parallel-sided crystal and for vanishing nuclear contribution to the diffraction. Furthermore it is assumed that the crystal has a center of symmetry and is not absorbing. The following parameters are used: The asymmetry parameter $b = \gamma_0/\gamma_H$ where $\gamma_0 = \cos \alpha_0$ and $\gamma_H = \cos \alpha_H$, α_0 being the angle between the normal to the crystal surface $\mathbf{n}$ and the direction of the incident beam and α_H being the angle between the direction of the reflected beam and $\mathbf{n}$; $B_0 = \mu_n|\mathbf{B}_A|/E$, $B_1 = \mu_n|\mathbf{B}_H|/E$ and $B_0^e = \mu_n|\mathbf{B}_e|/E$, where $\mathbf{B}_A$ is the average value of the magnetic field, $\mathbf{B}_H$ is the Fourier transform of the magnetic field with respect to the scattering vector $\mathbf{H}$, $\mathbf{B}_e$ is a homogeneous magnetic field outside the crystal parallel to $\mathbf{B}_A$, μ_n is the magnetic neutron moment, and E is the energy of the incident neutrons; φ, the angle between $\mathbf{B}_A$ and $\mathbf{B}_H$. From this, the fundamental equations for dynamical diffraction are:

$$(\varepsilon_0 + B_0)\psi_0^{(1)} + B_1 \cos \varphi \psi_H^{(1)} - B_1 \sin \varphi \psi_H^{(2)} = 0 \qquad (1a)$$

$$(\varepsilon_0 - B_0)\psi_0^{(2)} - B_1 \sin \varphi \psi_H^{(1)} - B_1 \cos \varphi \psi_H^{(2)} = 0 \qquad (1b)$$

$$\left(\frac{\varepsilon_0}{b} \mp B_0^e\left(1 - \frac{1}{b}\right) + B_0 + 2x\right)\psi_H^{(1)} + B_1 \cos \varphi \psi_0^{(1)} - B_1 \sin \varphi \psi_0^{(2)} = 0 \qquad (1c)$$

$$\left(\frac{\varepsilon_0}{b} \mp B_0^e\left(1 - \frac{1}{b}\right) - B_0 + 2x\right)\psi_H^{(2)} - B \cos \varphi \psi_0^{(2)} - B_1 \sin \varphi \psi_0^{(1)} = 0 \qquad (1d)$$

with

$$\varepsilon_0 = 1 - \frac{|\mathbf{k}'|^2}{|\mathbf{k}_0|^2}$$

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and

$$x = \frac{|\mathbf{H}|^2 + 2\mathbf{k}_o\mathbf{H}}{2|\mathbf{k}_0|^2} \simeq -\Delta\theta \sin 2\theta_B.$$

Here $\psi^{(1)}_{0,\mathrm{H}}$ and $\psi^{(2)}_{0,\mathrm{H}}$ are the components of the tensor

$$\begin{pmatrix} \psi^{(1)}_{0,\mathrm{H}} \\ \psi^{(2)}_{0,\mathrm{H}} \end{pmatrix}$$

corresponding to the forward-directed wave (index 0) and the reflected wave (index H) inside the crystal, $\mathbf{k}_0$ is the wavevector of the incident wave, $\mathbf{k}'$ is the wavevector of the forward-directed wave inside the crystal, $\mathbf{H}$ is the scattering vector, and $\Delta\theta$ is the deviation from the geometrical bragg angle θ_B. The upper signs in (1) refer to the incidence of neutrons polarized parallel to $\mathbf{B}_A$. Equation (1) is a homogeneous system and therefore ε_0 must satisfy the corresponding secular equation, which is to fourth degree in ε_0. For each solution of ε_0 a set of the quotients $\psi^{(1,2)}_{0,H}/\psi^e_0$, where ψ^e_0 is the amplitude of the incident wave can be obtained from (1). Then the quotients $\psi^{a(1,2)}_{0,H}/\psi^e_0$, where the $\psi^{a(1,2)}_{0,H}$ are the components of the tensors corresponding to the transmitted and reflected waves leaving the crystal, can be calculated using the boundary conditions at the entrance and the exit surfaces of the crystal. From this, the reflecting power P_H, which is the ratio of the intensity of the reflected beam to the intensity of the incident beam and P_0, the corresponding value for the transmitted beam as well as the polarization vectors $\mathbf{p}_{0,H}$ of the transmitted and the reflected beam can be obtained. The solutions $\psi^{a(1,2)}_{0,H}$ obviously depend on the polarization state of the incident wave. It is sufficient to know $\psi^{a(1,2)}_{0,H}$ for the incidence of neutrons with spin parallel to $\mathbf{B}_A$ and the incidence of neutrons with spin antiparallel to $\mathbf{B}_A$. For arbitrary polarization of the incident beam the values of $P_{0,H}$ and $\mathbf{p}_{0,H}$ can be obtained using the formulas given by Blume & Mendiratta (1976).

III. The reflected intensity for special cases

The formulas for the reflected intensity given in this section do not include *Pendellösung* oscillations. They are obtained from the intensity formulas including *Pendellösung* oscillations by integrating them with respect to the crystal thickness and are valid if the variation of the crystal thickness over the surface of the crystal is much greater than the *Pendellösung* length.

1. $\varphi = 0°$

In this case, the reflecting power P_H is given by

$$P_H = \frac{1}{2}\left[1 + \left(\frac{x - \Delta x}{B_1/\sqrt{b}}\right)^2\right]^{-1}$$

with

$$\Delta x = [(1-b)/2b](B_0 - B^e_0).$$

The reflected beam is completely polarized parallel to the incident beam. The reflection curve has a Lorentzian shape with a maximum deviated by an amount $[(1 - b)/2b](B_0 - B^e_0)$ from the geometrical Bragg angle and with a width given by $B_1/\sqrt{b}$. The deviation from the Bragg angle is due to refraction at the entrance surface of the crystal and is therefore proportional to the change of the magnetic field $B_0 - B^e_0$ at this surface. The result for P_H in the case of $\varphi = 0°$ is quite analogous to the corresponding result for X-ray diffraction (see, for example, Zachariasen, 1945).

2. $\varphi = 90°$

The reflecting power is given by

$$P_H = \frac{1}{2}\left[1 + \left(\frac{x - \Delta x}{B_1\sqrt{b}}\right)^2\right]^{-1}$$

with

$$\Delta x = [(1-b)/2b](B_0 - B^e_0) + B_0.$$

The reflected beam is completely polarized antiparallel to the incident beam in this case. Again there is a Lorentzian peak with a width $B_1/\sqrt{b}$. The deviation from the Bragg angle amounts to $B_0 + [(1 - b)/2b] \times (B_0 - B^3_0)$. It consists of two terms, the term due to refraction $[(1 - b)/2b](B_0 - B^e_0)$ and the term B_0 which is caused by a spin-flip within the crystal. The first term vanishes for $B_0 = B^e_0$, while the second does not depend on B^e_0.

3. $B_0 \gg B_1$

Applying this condition to the results for the symmetrical Laue case given by Schmidt & Deimel (1976), two peaks are obtained, separated by an amount B_0 for diffraction with and without spin-flip. Adoption of this result for the general asymmetrical Laue case means that the system of four equations (1) can be reduced to two equations in the following manner: because of the refraction at the surface the peak due to diffraction without spin-flip should appear at $[(1 - b)/2b](B_0 - B^e_0)$ and the other peak should appear at $B_0 + [(1 - b)/2b](B_0 - B^e_0)$. For diffraction without spin-flip $\psi^{(2)}_0$ and $\psi^{(2)}_H$ should be very much less than $\psi^{(1)}_0$ and $\psi^{(1)}_H$. Thus in (1a) and (1c) the terms containing $\psi^{(2)}_0$ and $\psi^{(2)}_H$ can be omitted. That (1b) and (1d) are consistent with the assumption that $\psi^{(2)}_{0,H} \ll \psi^{(1)}_{0,H}$ can be seen by solving the secular equation for the reduced system consisting of (1a) and (1c). It turns out that near the peak the coefficients of $\psi^{(2)}_0$ in (1b) and of $\psi^{(2)}_H$ in (1d) are nearly equal to $-2B_0$. Because B_0 is very much larger than B_1 it follows that $\psi^{(1)}_H \gg \psi^{(2)}_0$ and

$\psi_0^{(1)} \gg \psi_H^{(2)}$ in accordance with the assumption made above. Therefore the system (1) can be reduced to

$$(\varepsilon_0 + B_0)\psi_0^{(1)} + B_1 \cos\varphi\psi_H^{(1)} = 0$$

$$\left(\frac{\varepsilon_0}{b} - B_0^e\left(1 - \frac{1}{b}\right) + B_0 + 2x\right)\psi_H^{(1)} + B_1 \cos\varphi\psi_0^{(1)} = 0.$$

The result for P_H is

$$P_H = \frac{1}{2}\left[1 + \left(\frac{x - \Delta x}{B_1 \cos\varphi/\sqrt{b}}\right)^2\right]^{-1}$$

with

$$\Delta x = \frac{1-b}{2b}(B_0 - B_0^e).$$

In the same manner it can be shown that for the spin-flip peak (1b) and (1c) are consistent with $\psi_0^{(2)} \ll \psi_0^{(1)}$, $\psi_H^{(1)} \ll \psi_H^{(2)}$ and the system (1) is reduced to

$$(\varepsilon_0 + B_0)\psi_0^{(1)} - B_1 \sin\varphi\psi_H^{(2)} = 0$$

$$\left(\varepsilon_0 - B_0^e\left(1 - \frac{1}{b}\right) - B_0 + 2x\right)\psi_H^{(2)} - B_1 \sin\varphi\psi_0^{(1)} = 0.$$

The result for P_H is

$$P_H = \frac{1}{2}\left[1 + \left(\frac{x - \Delta x}{B_1 \sin\varphi/\sqrt{b}}\right)^2\right]^{-1}$$

with

$$\Delta x = B_0 + \frac{1-b}{2b}(B_0 - B_o^e).$$

IV. *Pendellösung* effects

In this section, for simplicity, the symmetrical Laue case is assumed. the *Pendellösung* causes a rather complicated oscillation of the reflecting power with $C = k_0 t/2\gamma_0$, where t is the thickness of the crystal. As an example, Fig. 1 shows the reflecting power as a function of C for $x = 0$, $\varphi = 45°$ and $B_0 = B_1$. For special cases the complicated structure becomes more simple. In particular, for $\varphi = 0$, $\varphi = 90°$ or $B_0 = 0$ there is only one period, which is given by $2\pi/B_1$ at the center of the peak. In the case of $B_0/B_1 \gg 1$ the *Pendellösung* period is given by $2\pi/(B_1 \cos\varphi)$ at $x = 0$ and $2\pi/(B_1 \sin\varphi)$ at $x = B_0$. These periods correspond to the peaks arising at $x = 0$ and $x = B_0$ (see § III.3).

In the opposite case, If $B_0/B_1 \ll 1$, one obtains at $x = 0$:

$$\left|\frac{\psi_H^{a(1)}}{\psi_0^e}\right|^2 = \sin^2 CB_1[1 - \sin^2\varphi\cos^2(CB_0\cos\varphi)]$$

$$\left|\frac{\psi_H^{a(2)}}{\psi_0^e}\right|^2 = \sin^2 CB_1 \sin^2\varphi\cos^2(CB_0\cos\varphi).$$

Therefore the reflecting power is given by

$$P_H = \sin^2 CB_1.$$

This is the familiar result with one period not affected by B_0. However, the polarization $(p_H)_z$ in the direction of $\mathbf{B}_A$ is given by

$$(p_H)_z = 1 - 2\sin^2\varphi\cos^2(CB_0\cos\varphi)$$

Thus, the *Pendellösung* variation of the polarization with C shows a period of $2\pi/(B_0 \cos\varphi)$.

Because $B_0 \ll B_1$ it is possible that even if the variation of the reflecting power is smeared out by integration with respect to the crystal thickness the variation of the polarization remains.

V. Integrated reflecting power

1. *Symmetrical Laue case*

The integrated reflecting power R^θ is the total reflecting power integrated with respect to the angle of incidence θ, which is related to x by $\theta = \theta_B - x/\sin 2\theta_B$. The results shown in this section were obtained by integrating the formulas without *Pendellösung* oscillations given by Schmidt & Deimel (1976). For $B_0 = 0$ and for $\varphi = 0°$ one obtains the familiar result:

$$R^\theta = \frac{\pi}{2}B_1.$$

In the case of $B_0/B_1 \gg 1$ the reflecting power is the sum of two Lorentz curves located at 0 and B_0 and with widths $B_1 \cos\varphi$ and $B_1 \sin\varphi$. Thus, in this case the integrated reflecting power becomes

$$R^\theta = \frac{\pi}{2}B_1(\cos\varphi + \sin\varphi).$$

It depends on the angle φ and is largest for $\varphi = 45°$. For intermediate values of B_0/B_1 numerical integrations were done for several values of φ. Fig. 2 shows the integrated reflecting power as a function of B_0/B_1

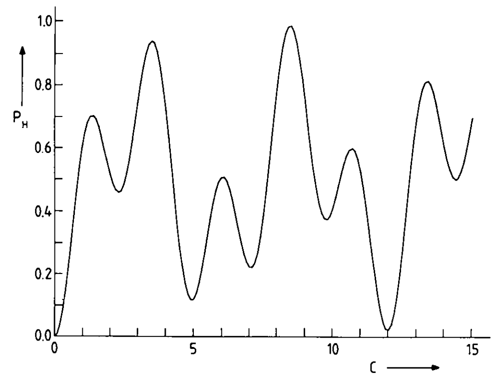

Fig. 1. *Pendellösung* oscillations of the reflecting power for $x = 0$, $B_0 = B_1$, and $\varphi = 45°$.

normalized to the integrated reflecting power for $B_0/B_1 = 0$ for some values of φ. For large values of B_0/B_1, R^θ tends to $(\pi/2)B_1$ (cos φ + sin φ) in each case, as was pointed out before.

2. *Asymmetrical Laue case*

For this case explicit results for the reflecting power are only known for $\varphi = 0$, $\varphi = 90°$ and $B_0/B_1 \gg 1$. In the first two cases one obtains the familiar result. In the third case, as for $b = 1$, the reflection curve splits into two Lorentzian peaks and the integration can be done easily. The result for R^θ is then the same as for $b = 1$ apart from a factor $\sqrt{b}$ due to the broadening of the peaks.

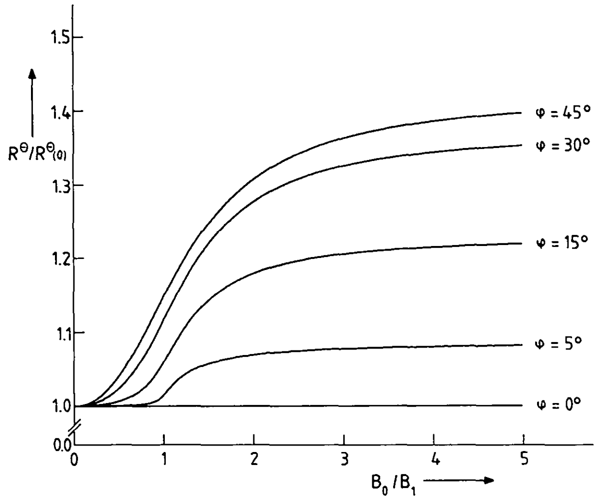

Fig. 2. Integrated reflecting power as a function of B_0/B_1 normalized to the value for $B_0/B_1 = 0$ for $\varphi = 0$, 5, 15, 30, and 45°.

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Experiments on Dynamical Magnetic Neutron Diffraction by $DyFeO_3$

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Abstract

The integrated reflecting powers of two nuclear and one magnetic reflexions of a $DyFeO_3$ crystal have been measured. The results indicate that dynamical magnetic diffraction occurs. In addition, the integrated reflecting power of the magnetic reflexion was measured as a function of the magnetic field applied to the crystal. The results are discussed.

1. Introduction

Dynamical diffraction of thermal neutrons has been demonstrated by various experiments (Knowles, 1956; Sippel, Kleinstück & Schulze, 1965; Shull, 1968). However, in all these experiments diffraction is only due to nuclear scattering. Dynamical magnetic diffraction was never observed. The theory of dynamical magnetic neutron diffraction was recently treated by Schmidt & Deimel (1976), Belyakov & Bokun (1976), Guigay & Schlenker (1978), and Schmidt (1983). The diffraction process depends on the angle φ between $\mathbf{B}_A$, the average magnetic field in the crystal, and $\mathbf{B}_H$, the Fourier transform of the magnetic field with respect to the scattering vector, and on the ratio B_A/B_H with $B_A = |\mathbf{B}_A|$ and $B_H = |\mathbf{B}_H|$. The aim of the experiments described here was twofold: firstly to establish dynamical magnetic neutron diffraction experimentally, and secondly to test the theoretical statements of Schmidt (1983).

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