PERFECT CRYSTAL NEUTRON OPTICS

A. Zeilinger
Atominstitut der Österreichischen Universitäten
A-1020 Wien, Austria
and
Massachusetts Institute of Technology Cambridge, MA 02139, USA

1. Introduction and Fundamentals

In recent years neutron optics started to gain increasing attraction despite the handicap this field experiences because the index-of-refraction (compare /1-3/) of all materials and of magnetic fields for thermal neutrons

$$n = 1 - \frac{1}{2E}(V_N(0) \pm \mu B) \qquad (1)$$

is very close to 1. This is because the kinetic energy E of thermal neutrons ($\sim 10^{-2}$ eV) is very much larger than both the mean nuclear interaction potential $V_N(0)$ ($\sim 10^{-8}$ to 10^{-7} eV) and the interaction potential of the neutron magnetic moment μ with a magnetic field B ($6.03 \cdot 10^{-8}$ eV for B = 1 T).
This limitation was surpassed in two different ways. On the one hand the use of very cold and ultra cold neutrons (see /1/) makes it possible to work in a neutron energy range where the index-of-refraction is high.
On the other hand the introduction of perfect crystals allows the construction of optical components which provide large deflection angles even for thermal neutrons. This feature has finally lead to the successful development of neutron interferometers /4/.
The results of the dynamical theory of neutron diffraction /5/ as obtained from the Schroedinger equation turns out to be closely analogous to the X-ray case, even the size of some effects is the same. Thus we can refer to the corresponding articles in this volume /6,7/.
The reason for this feature is, that the time-independent Schroedinger equation is formally quite similar to the Helmholtz-equation one obtains from Maxwell's equations. The analog to the spatially dependent polarizability in the X-ray case is the spatially dependent potential in the neutron case. For more detailed comparisons we refer to review papers on neutron dynamical diffraction /8,9,10/.
The phenomenological pecularities of the neutron case are

1. With few exceptions, the absorption for neutrons is very low, it is particularly negligible for Si-crystals of usual laboratory thickness.
2. The neutron velocity is low ($\sim$ 1000 m/s for thermal neutrons) as

compared to X-rays.

3. The neutron is a spin-$\frac{1}{2}$ particle with magnetic moment and experiences considerable magnetic interaction.

This latter property leads to nontrivial extensions of the X-ray dynamical theory results /11/, if perfect magnetic crystals are employed. Another extension concerns the incorporation of inelastic neutron scattering into dynamical diffraction theory /12/. The theoretical work mentioned above has in general been restricted to the 2-beam case, but there has already been some work done on the neutron 3-beam case /13/. The present paper will be restricted mainly to those aspects which have found experimental verification.

Two large and important fields of applications, i.e. neutron topography and neutron interferometry are not covered here since they are the topics of two other papers of this volume /14,15/.

2. Laue-Case Reflectivity Experiments

Knowles /16/ was the first one to demonstrate neutron dynamical diffraction effects by showing that the intensity of neutron-capture γ-radiation varies in a characteristic way with a variation close to the exact Bragg-angle of the direction of incidence of the incoming neutrons for $CdSO_4$-crystals. This effect is related to the anomalous transmission phenomenon which is generally encountered in X-ray dynamical diffraction. More experimentation on neutron anomalous transmission was done by Sippel et al. /17/ using InSb-crystals and by Shilstein et al. /18/ using CdS.

A rather interesting phenomenon which is characteristic for neutrons and has no X-ray counterpart is anomalous spin-incoherent scattering. This effect was demonstrated by Sippel et al. /19/ for a KH_2PO_4-crystal. In that crystal the planes containing the H-atoms are arranged between the planes containing the K and P atoms. Thus, by varying again the angle of incidence in an angular region close to the exact Bragg-angle the amplitudes of the wave-fields at the positions of the H-atoms can be varied. This leads to a strong variation of the transmitted intensity because H shows a very strong spin-incoherent scattering which reduces the coherent scattering.

The validity of the integrated reflectivity predictions of dynamical diffraction theory for nonabsorbing crystals has also been shown first by Sippel et al. /20/.

3. The Bragg-Case Double-Crystal Diffractometer

For the absorption-free case the reflectivity of a perfect crystal in symmetric Bragg-geometry is obtained to be unity over an angular range

$$\Delta\theta = \frac{2|F_N(\vec{G})|\lambda^2}{\pi v_c \sin(2\theta_B)} \tag{2}$$

where $F_N(\vec{G})$ is the appropriate structure factor, λ is the neutron wave length, v_c is the volume of the crystal unit cell and θ_B is the Bragg-angle. The reflection width is typically a few seconds of arc, the reflection curve was measured in detail by Kikuta et al. /21/.
One employs this high angle resolution feature for small angle scattering experiments in the double-crystal spectrometer where two perfect crystals in parallel arrangement are used with the sample between them, the angular changes of the beam due to the sample being measured. This arrangement has been used, among others, in experiments on a limit of the neutron charge /22/, on single-slit diffraction of neutrons /23/ and refraction of neutrons at magnetic boundaries /24,25/. In the backscattering spectrometer /26/ the double-crystal arrangement provides an energy resolution of about 10^{-7} eV.

4. Ray-Optical Aspects

In the following we restrict our considerations to the absorption-free symmetric Laue-case in the 2-beam approximation. Then the solutions to the Schroedinger equation are the two wave fields (see e.g. /9/)

$$\psi_{1,2} = u_{1,2}(0) \exp(i\,\vec{K}_{1,2}.\vec{r}) + u_{1,2}(\vec{G}) \exp[i\,(\vec{K}_{1,2} + \vec{G}).\vec{r}] \tag{3}$$

with

$$\vec{K}_{1,2} = \vec{k} + \frac{\pi}{\Delta}(-y \pm \sqrt{y^2+1})\hat{n} - \frac{k\,V(0)}{2E\cos\theta_B}\hat{n} \tag{4}$$

$$y =: (\theta_B - \theta)\frac{E}{|V(\vec{G})|}\sin(2\theta_B), \quad \Delta = \lambda\frac{E}{|V(\vec{G})|}\cos\theta_B \tag{5}$$

$V(\vec{G})$ is the Fourier-component of the neutron-crystal interaction potential and Δ is the pendellösung length. The amplitudes u are also functions of y and given elsewhere /9/. The angle Ω of the mean neutron current with the lattice planes is given through

$$\Gamma \equiv \frac{\tan\Omega_{1,2}}{\tan\theta_B} = \frac{\mp y}{\sqrt{1+y^2}} \qquad (6)$$

This equation defines the Borrmann-fan, which is excited if a fine entrance slit is used (Fig.1). The intensity distribution within the Borrmann-fan,

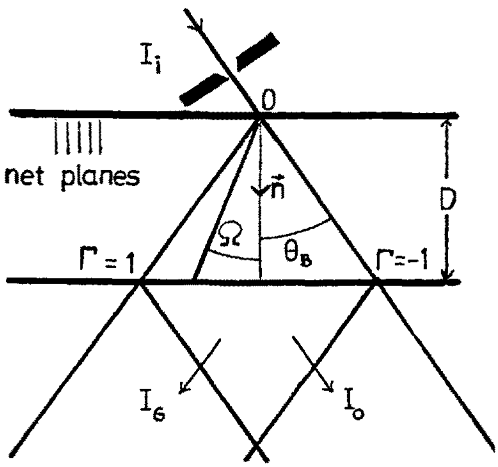

Fig.1: Borrmann-fan neutron propagation in the symmetric Laue-case

the so-called "Kato-profile" was measured in detail by Shull and co-workers /27/. With no coherence between the +y and -y components we obtain

$$I_G(\Gamma) = I_i/2 \sqrt{1-\Gamma^2} \quad , \qquad (7)$$

which, if smeared out with the experimental resolution gives the result of Fig.2, which was obtained with white radiation and poor collimation. If the collimation is improved, the wave length becomes well defined and thus interferences between +y and -y components are observable which are the characteristic spherical wave interference oscillations as shown in Fig.3. This feature was used by Shull et al. for a precision determination of the scattering lengths of Si and Ge.

An interesting question concerns the optical length of the path given by Equ.6, which is different from its geometrical length because the momentum eigenstates $\hbar\vec{K}_{1,2}$ and $\hbar(\vec{K}_{1,2}+\vec{G})$ are not parallel to the path. Thus one defines an infinitesimal zig-zag path (Kato /28/) with components along the momentum eigendirections, whose length is

$$D_{opt} = D/\cos\theta_B \qquad (8)$$

independent of y in the symmetrical Laue-case. This path length was measured in a neutron time-of-flight experiment /29/.

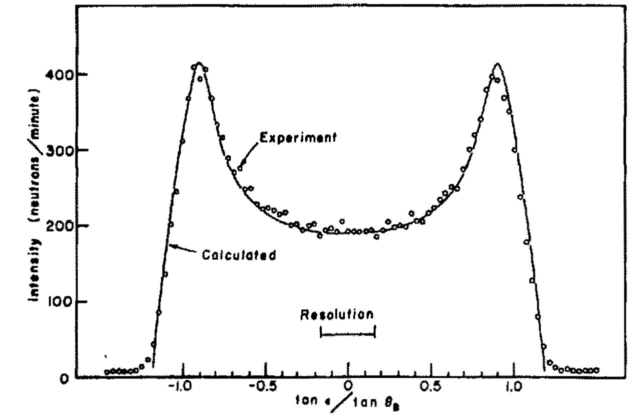

Fig.2: Distribution of the intensity I_G released from the backface of an arrangement as shown in Fig.1 (after Shull /11/).

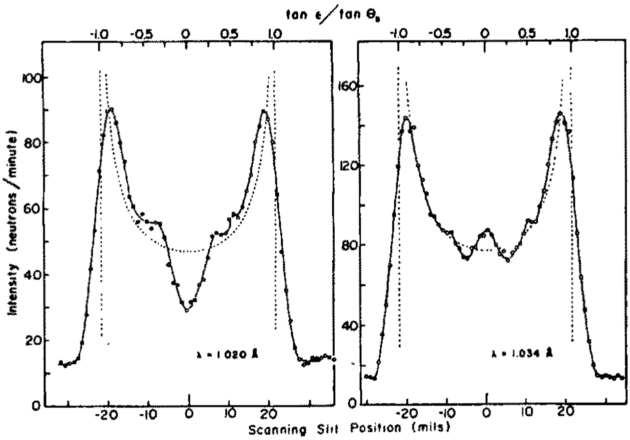

Fig.3: As Fig.2, but with better incident radiation collimation, such that spherical wave interferences occur (after Shull /11/).

5. Applications of Ray Optics

5.1 Diffraction Focussing of Neutrons

There are different types of focussing effects to be expected in dynamical diffraction /30/. One of them occurs if two crystals of equal thickness are arranged parallel in symmetric Laue-geometry, which is most easily seen from symmetry considerations (Fig.4). This double Laue-crystal arrangement is very sensitive to the relative angular position of the crystals /31,32/, because the double crystal rocking curve exhibits a narrow peak of width

$$\delta\theta = \lambda/W \tag{9}$$

on top of the broader peak whose width is approximately that of Equ.2. As W is the width of the beam between the crystals $\delta\theta$ is typically in the range of a few 10^{-3} arc sec (Fig.5).

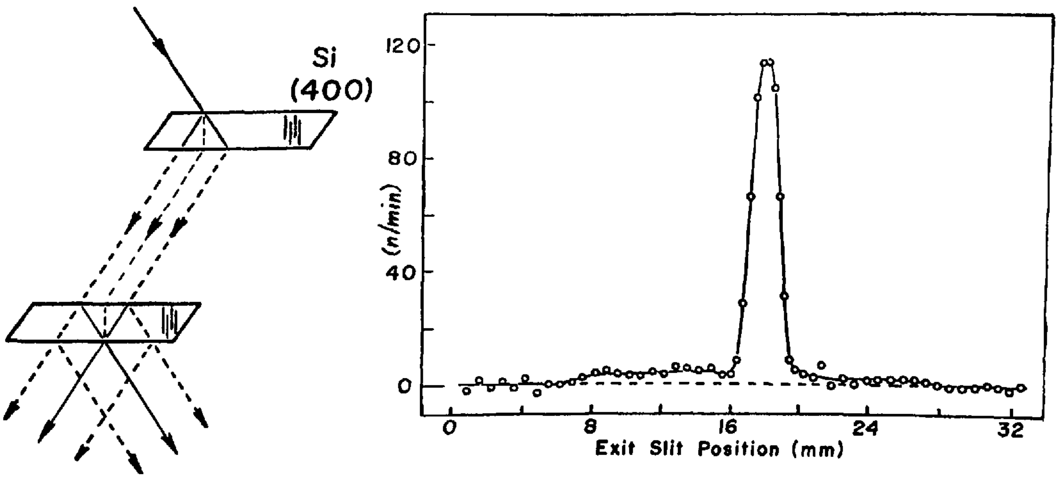

Fig.4: Diffraction Focussing of Neutrons:
Ray paths in a double Laue-case arrangement showing focussing action (left) and intensity released from the backface of the second crystal into the double diffracted direction as scanned with an exit slit (right) /31/.

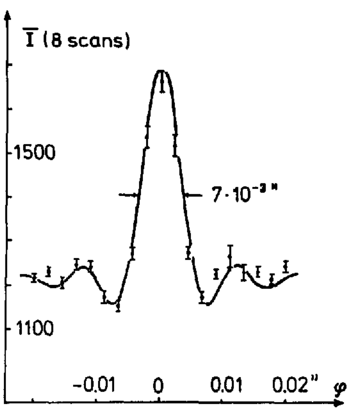

Fig.5: The very narrow central peak occuring in the rocking curve of two Laue-crystals with equal thickness /32/.

The physical reason for this effect is the property that such a double crystal arrangement shows interference action in the central focus /31/ if the crystals are exactly parallel. This constructive interference is deteriorated by optical path length changes due to angular changes, which leads directly to Equ.9. An alternative interpretation can be given in terms of the oscillatory type Laue-case reflection curves /32/.

5.2 The Angle Amplification Feature

For small y we obtain from Equs.5 and 6

$$\Omega \approx (\theta - \theta_B) \frac{E}{|V(\vec{G})|} 2 \sin^2 \theta_B \qquad (10)$$

which for e.g. Si (400)-reflection and $\lambda = 1.865$ Å gives

$$\Omega \approx 4.4 \cdot 10^5 \ (\theta - \theta_B). \qquad (11)$$

Thus, a small change of the incoming radiation direction results in a large change of in-crystal neutron propagation direction. This feature was first used by Authier /33/ for X-rays and later exploited by Kikuta et al. /21/ to measure the small directional changes of a neutron beam due to prism refraction (Fig.6). In that experiment neutron absorbing slits in front and on the backface of the first crystal plate act as a "crystal-collimator" selecting only those neutrons which are travelling through the crystal along paths close to the lattice planes. These neutrons travel in the second crystal plate again in the same directions. If now the beam is deflected on its way between the two crystals, its path in the second crystal plate splits according to the equations above (Fig.6).
The same phenomenon occurs if not the direction but the wave length of the neutrons is changed on their way between the crystal plates because a change of wave length results in a change of Bragg-angle according to

$$\delta\theta_B = \frac{\delta\lambda}{\lambda} \tan \theta_B . \qquad (12)$$

This feature was utilized /34/ in a direct measurement of the wave length change of a neutron in a magnetic field (Fig.7)

$$\frac{\delta\lambda}{\lambda} = \mp \frac{\mu B}{2E} . \qquad (13)$$

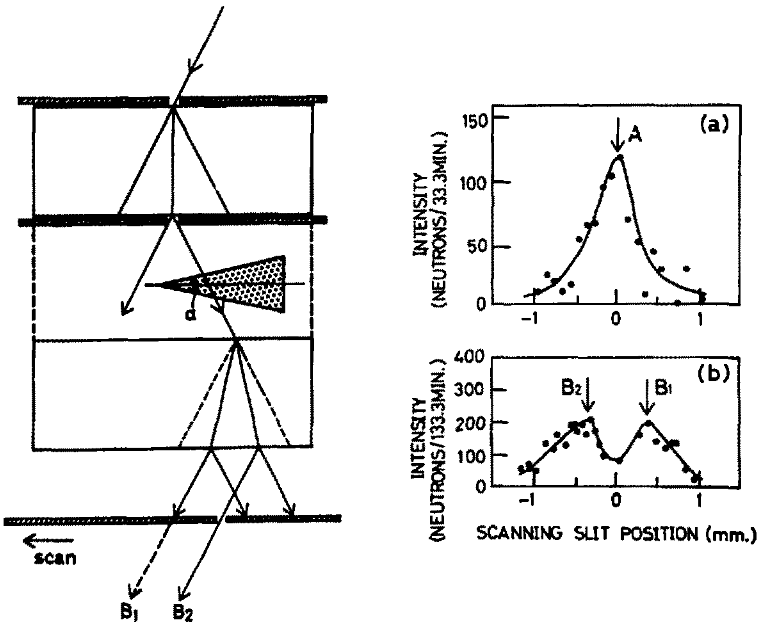

Fig.6: Measurement of small directional changes utilizing the angle amplification feature. Principle of the setup (left) and backface intensity profiles (right) without (a) and with (b) a 0.032 arc sec refracting prism in beam.

The results demonstrate a sensitivity of about 10^{-8} eV. This experiment can also be regarded as the first verification of dynamical diffraction theory predictions because the experimental situation corresponds to the case of a purely nuclear reflection in a ferromagnet. Additionally, by simultaneous magnetic field action and prism refraction a separation of the spin states could be achieved within the crystal /34/.

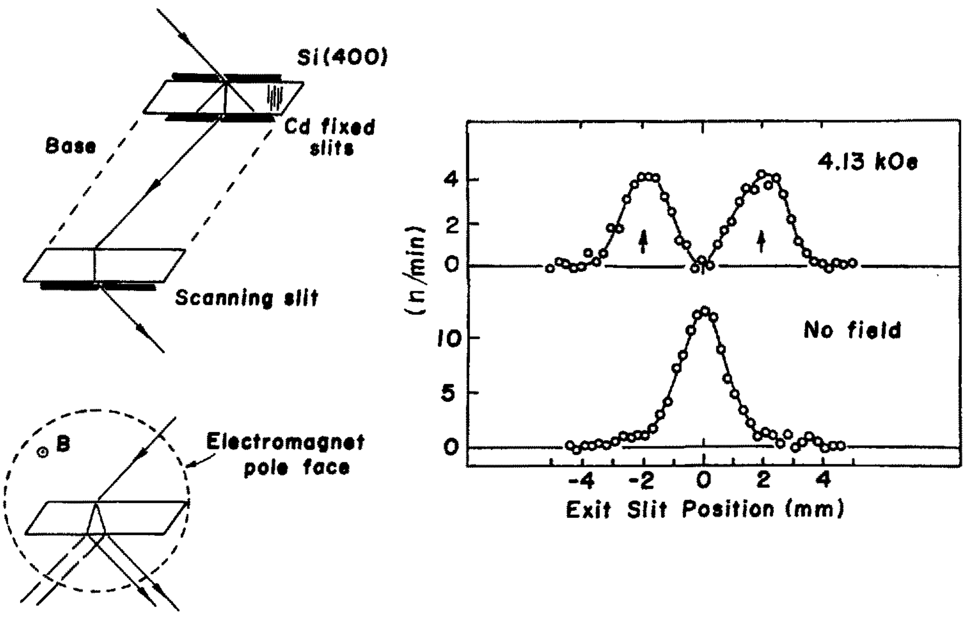

Fig.7: Measurement of the wave length change of a neutron in a magnetic field /34/. Principle of the arrangement (top left) and ray path in the second crystal plate with magnetic field applied (bottom left). The experimental results (right) demonstrate the expected separation effect.

6. Concluding Comments

The neutron's magnetic dipole interaction introduced to dynamical diffraction opens an exciting field of new experimental possibilities. The extension of the experiments mentioned in the last paragraph will be to use inhomogeneous rather than homogeneous magnetic fields applied to, say, a nonmagnetic crystal.

For such an experiment and for a similar one where the potential gradient is due to the gravitational interaction of the neutron /35/ an extension of dynamical diffraction theory to the case of a force acting on the particle while in the crystal is needed, a situation not encountered in the X-ray case. It turns out that approaches very similar to diffraction by a crystal with a gradient in lattice spacing /7/ can be used. For example one obtains in the Laue-case curved in-crystal neutron paths /36/ with curvatures immensly larger than in a free-space Stern-Gerlach experiment. Creation of new wavefields can be expected if the gradient of the mean potential V(O) does not fulfil the inequality

$$\Delta_o\ \vec{G}.\mathrm{grad}\ (V(O)) \ll k(V(\vec{G})).$$

anymore. It seems feasible that such field gradients can be achieved experimentally. This offers the possibility of directly testing the different approaches used in topography /7,37/.
The field of new effects expected is even more extended if the polarisation of the neutron is taken into account explicitely. So, e.g. there exist spin rotation phenomena if the two polarisation states travel as different wave fields /38/, which effect is related to the coherence phenomena in neutron polarisation /39/. Finally, the application of magnetic fields to perfect nonmagnetic crystals /40/ or the use of perfect ferromagnets /41/ in off-set Bragg-geometry permits the construction of new types of highly efficient neutron polarizers. Here, with inhomogeneous magnetic fields an increase in reflectivity is to be expected.

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References

/1/ A.Steyerl: Propagation of Neutron Beams (this volume)
/2/ J.Sivardiere: Formalism of Wave-Matter Interaction (this volume)
/3/ G.L.Squires: Interactions of Neutrons with Matter (this volume)
/4/ H.Rauch, W.Treimer, U.Bonse: Phys.Lett. 47A (1974) 271
/5/ M.L.Goldberger, F.Seitz: Phys.Rev. 71 (1947) 294
Yu.Kagan, M.Afanasev: Soviet Phys. JETP 22 (1966) 1032; Z.Naturforsch. 28a (1973) 1351
P.Hiismäki: Acta Cryst. A25 (1969) 377
R.Lenk, H.Solbrig: phys.stat.sol. B46 (1971) 273
H.Solbrig: phys.stat.sol. B47 (1971) 143 and B51 (1972) 555
/6/ A.Authier: Kinematical and Dynamical Theories of X-ray Diffraction (this volume)

/7/ F.Balibar: Geometric and Diffraction Approaches in X-Ray Diffraction (this volume)

/8/ F.Eichhorn: Dynamische Neutroneninterferenzen und ihre Anwendungen, in: Dynamische Interferenztheorie (O.Brümmer, H.Stepanik, Eds.), Akademische Verlagsgesellschaft, Leipzig, 1976

/9/ H.Rauch and D.Petrascheck: Dynamical Neutron Diffraction and Its Application, in: Neutron Diffraction (H.Dachs, Ed.) Vol.6 of Topics in Current Physics, Springer Verlag, Berlin, 1978

/10/ V.F.Sears: Canad.J.Phys. 56 (1978) 1261

/11/ C.G.Shull: J.Appl.Cryst. 6 (1973) 257
C.Stassis, J.A.Oberteuffer: Phys.Rev. B10 (1974) 5192
J.Sivardiere: Acta Cryst. A31 (1975) 340
H.H.Schmidt, P.Deimel: J.Phys.C: Solid State Phys. 8 (1975) 1991
S.K.Mendiratta, M.Blume: Phys.Rev. B14 (1976) 144
V.A.Belyakov, R.Ch.Bokun: Soviet Phys. Solid State 18 (1976) 1399
O.Schärpf: J.Appl.Cryst. 11 (1978) 626 and 631

/12/ S.K.Mendiratta: Phys.Rev. B14 (1976) 155

/13/ W.Treimer: Phys.Lett. 68A (1978) 162 and Z.Naturforsch. 33a (1978) 1432

/14/ W.Graeff: X-ray and Neutron Interferometry (this volume)

/15/ M.Schlenker: Neutron Topography (this volume)

/16/ J.W.Knowles: Acta Cryst. 9 (1961) 61

/17/ D.Sippel, K.Kleinstück, G.E.R.Schulze: phys.stat.sol. 2 (1962) K104

/18/ S.Sh.Shilshtein, V.A.Somenkov, V.P.Dokashenko: JETP Lett. 13 (1971) 214

/19/ D.Sippel, F.Eichhorn: Acta Cryst. A24 (1968) 237

/20/ D.Sippel, K.Kleinstück, G.E.R.Schulze: Phys.Lett. 14 (1965) 174

/21/ S.Kikuta, I.Ishikawa, K.Kohra, S.Hoshino: J.Phys.Soc.Japan 39 (1975) 471

/22/ C.G.Shull, K.W.Billman, F.A.Wedgewood: Phys.Rev. 153 (1967) 1415

/23/ C.G.Shull: Phys.Rev. 179 (1969) 752

/24/ S.Sh.Shilshtein, V.A.Somenkov, M.Kalanov: Sov.Phys.JETP 36 (1973) 1170
N.O.Elyutin, A.O.Bubleinik, V.A.Somenkov, S.Sh.Shilshtein: JETP Lett. 18 (1973) 186

/25/ O.Schärpf, H.Strothmann: Vol.II, p.713 in Proc.Conf.Neutron Scattering, Gatlinburg 1976, publ. by U.S.Dept. of Commerce, CONF-760601-P2

/26/ M.Birr, A.Heidemann, B.Alefeld: Nucl.Instr.Meth. 95 (1971) 435

/27/ C.G.Shull: Phys.Rev.Lett. 21 (1968) 1585
C.G.Shull, J.Oberteuffer: Phys.Rev.Lett. 29 (1972) 871

C.G.Shull, W.M.Shaw: Z.Naturforsch. 28a (1973) 657

/28/ N.Kato: J.Appl.Phys. 39 (1968) 2225

/29/ C.G.Shull, A.Zeilinger, G.L.Squires, M.A.Horne: to be published, preliminary result presented at "Int.Workshop on Neutron Interferometry", Grenoble 1978

/30/ V.L.Indenbom, I.Slobodetskii, K.G.Truni: Sov.Phys.JETP 39 (1974) 542
E.V.Suvorov, V.I.Polovinkina: JETP Lett. 20 (1974) 145
A.M.Afanasev, V.K.Kon: Sov.Phys.Solid State 19 (1977) 1035
V.V.Aristov, V.I.Polovinkina, I.M.Shmytko, E.V.Shulakov: JETP Lett. 28 (1978) 4

/31/ A.Zeilinger, C.G.Shull, M.A.Horne, G.L.Squires: Proc.Int.Workshop on Neutron Interferometry, Grenoble 1978, U.Bonse and H.Rauch (Eds.), Oxford University Press, 1979

/32/ U.Bonse, W.Graeff, H.Rauch: Phys.Lett. 69A (1979) 420

/33/ A.Authier: Compt.rend.Acad.Sci.Paris 251 (1960) 2502

/34/ A.Zeilinger, C.G.Shull: Phys.Rev. B (in press)

/35/ S.A.Werner: private communication

/36/ A.Zeilinger: paper to be presented at Int.Conf. Polarized Neutrons in Condensed Matter Res., Zaborow 1979

/37/ C.Malgrange: X-ray Topography: Principles (this volume)
F.Balibar, Y.Epelboin, C.Malgrange: Acta Cryst. A31 (1975) 836

/38/ A.Zeilinger: Proc.Int.Workshop on Neutron Interferometry, Grenoble 1978, U.Bonse and H.Rauch (Eds.), Oxford University Press 1979

/39/ F.Mezei: Coherent Approach to Polarized Neutrons (this volume)

/40/ S.Funahashi: Nucl.Instr.Meth. 137 (1976) 99

/41/ M.Hart: X-ray Optics (this volume)