Magnetic field effects on dynamical diffraction of neutrons by perfect crystals

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Experiments have been performed on the effect of magnetic fields on dynamical diffraction phenomena of neutrons, using a perfect Si crystal in symmetric Laue geometry. A crystal collimator was used to define the incoming beam. The small change of neutron wavelength when entering a modest magnetic field results in large changes of neutron-propagation direction within the crystal. Energy changes as small as 10^{-8} eV were detectable in the system by observing these directional changes. By combining prism refraction with the magnetic field action, the polarized states could be separated within the crystal and the polarization of these separated beams was studied.

I. INTRODUCTION

The experiments hitherto carried out on the dynamical diffraction of neutrons have not made explicit use of neutron spin because of the unavailability of large perfect magnetic crystals. Thus it has not been possible to test experimentally the wealth of new effects discussed and proposed by several authors[1,2] which are to be expected if the magnetic interaction of the neutron with the scattering centers comes into play in dynamical diffraction. These effects are associated with the splitting of each branch of the dispersion surface into two branches. This results in a doubling of the number of wave fields in the crystal. In most experimentally interesting cases, these wave fields are polarized. It is to be expected that separate dispersion surfaces for each spin state will lead to different neutron-propagation directions in the crystal, the difference being related to the Zeeman splitting.

As has been pointed out recently,[3] some magnetic dynamical neutron-diffraction effects can be expected if diffraction by perfect nonmagnetic crystals placed in suitable magnetic fields is studied. The present paper reports the experimental verification of some of the effects predicted.

II. MEASUREMENT OF VERY SMALL WAVE LENGTH CHANGES BY LAUE-CASE DYNAMICAL DIFFRACTION

We first discuss the concepts necessary for the present study of the dynamical diffraction of neutrons in the symmetric Laue-case with no magnetic field present. The notation we use is that of a recent review article.[1] It is assumed that only one reciprocal-lattice point besides the origin is close to the Ewald sphere, which implies that the two-beam approximation can be used. In this case an incoming plane neutron wave gives rise to two wave fields within the crystal associated with the two branches of the dispersion surface. Furthermore, each wave field is a coherent superposition of two plane-wave components. Neglecting small refraction corrections, one of these plane-wave components travels in the direction of the incoming plane wave (the "forward" direction) and the other one travels in the Bragg diffracted direction. The coherent superposition of the forward and Bragg diffracted waves, which in general are of different amplitude, results in a propagation direction of the wave field which is given by

$$\tan \Omega_{1,2} = \Gamma_{1,2} \tan \theta_B . \tag{1}$$

Here the subscripts 1 and 2 refer to the two wave fields, $\Omega_{1,2}$ is the angle of the propagation direction of the respective wave field within the crystal relative to the lattice planes, θ_B is the Bragg angle, and the dimensionless parameter Γ is given by

$$\Gamma_{1,2} = \mp y/(1+y^2)^{1/2} . \tag{2}$$

Here, y is related to the deviation angle $\delta\theta$ of the incoming-wave direction from the exact Bragg angle θ_B through

$$y = -\delta\theta (E/|V_G|) \sin(2\theta_B) , \tag{3}$$

where E is the neutron energy and V_G is the appropriate Fourier component of the crystal-neutron interaction potential, including the Debye-Waller factor. Since in neutron diffraction by crystals the neutron energy is always much larger than the potential V_G, these last equations imply that a perfect crystal acts like an angle amplifier. For very small $\delta\theta$ we obtain

$$\Omega \approx \delta\theta (E/|V_G|) 2 \sin^2 \theta_B , \tag{4}$$

which, e.g., for the Si(400) reflection and a neutron wavelength of $\lambda = 1.865$ Å (as in our experiment) implies $\Omega = (4.4\times10^5)\delta\theta$. This angle amplification feature of dynamical diffraction can be used to measure very small changes of the incident neutron direction. As the two wave fields travel in different directions within the crystal, they become separated if an entrance slit is used. The spatial separation of the beams leaving the crystal is a measure of $\delta\theta$. The first experimental demonstration of this effect was reported by Kikuta *et al.*,[4] who used it to measure the small angular change in the direction of a neutron beam caused by prism refraction.

In principle, one would have to take into account the diffraction of the neutrons by the finite-size entrance slit. In ordinary optics diffraction phenomena can be neglected if the characteristic dimensions of obstacles are on a scale which is large compared to the wavelength of the radiation. This is the domain of geometrical optics. In perfect-crystal diffraction the corresponding characteristic dimension is the pendellösung length, i.e., the primary extinction length. This fact was studied in detail by Indenbom and Chukhovskii.[5] Therefore, if a slit is placed in front of a crystal much wider than the pendellösung length, diffraction effects at the slit can be neglected, as is the case in the present study.

In the present paper, the applicability of the principles discussed above to the measurement of small changes in neutron wavelength rather than neutron-propagation direction is demonstrated. To analyze this experimental possibility, we recall that a change $\delta\lambda$ of the neutron wavelength λ implies a change in the Bragg angle of

$$\delta\theta_B = (\delta\lambda/\lambda)\tan\theta_B . \tag{5}$$

If we assume, as an example, that the neutron wave initially fulfilled the exact Bragg condition, then the last equation implies that after a wavelength change of $\delta\lambda$ with no simultaneous direction change, the neutron direction deviates from the exact Bragg angle θ_B by the angle $-\delta\theta_B$. This in turn results in a change in the neutron-propagation direction within the crystal by means of Eq. (3).

To demonstrate the sensitivity to small energy changes, we discuss again neutrons with a wavelength of $\lambda = 1.865$ Å incident on a Si crystal in symmetric (400) Laue geometry, and assume that the energy of the neutrons differs by 10^{-8} eV from that required to fulfill the Bragg condition exactly. This energy difference corresponds to a relative wavelength difference of $\delta\lambda/\lambda = 2.13\times10^{-7}$. This implies that the two wave fields propagate at angles of 5.1° with respect to the lattice planes. As will be seen from our experimental results, such a change can be measured with the experimental conditions used.

III. WAVELENGTH CHANGE OF A NEUTRON DUE TO ZEEMAN SPLITTING

A neutron interacting with a stationary magnetic field experiences a change of momentum which is spin dependent. This can be understood by referring to the Hamiltonian of a neutron in a magnetic field $\vec{B}$:

$$\mathcal{H} = \vec{p}^2/2m - \vec{\mu}\cdot\vec{B} , \tag{6}$$

where $\vec{p}$ is the neutron momentum operator, m is the neutron mass, and $\vec{\mu}$ is the neutron magnetic-moment operator

$$\vec{\mu} = \mu\vec{\sigma} , \tag{7}$$

where μ is the magnetic moment of the neutron ($\mu = -6.0311\times10^{-12}$ eV/G) and $\vec{\sigma}$ is the Pauli vector. If the magnetic field B is not time dependent, then the neutron momentum change obtained from Eq. (6) reflects the way field gradients are oriented with respect to the neutron-propagation direction. In the Stern-Gerlach experiment, the field gradients are oriented normal to the neutron-propagation direction, which results in the well-known spatial separation of the two spin states. The most recent experiment of this kind with neutrons was performed by Hamelin *et al.*[6] In contrast, if the field gradients are parallel to the neutron-propagation direction, they produce a longitudinal change of neutron momentum resulting in a wavelength change. This leads to effects like the refraction of neutrons by magnetic fields as measured by Just *et al.*[7]

In the present paper we report an explicit measurement by crystal diffraction of this wavelength change, which is (for $\mu B \ll E$)

$$\lambda_\pm = \lambda(1 \mp \mu B/2E) , \tag{8}$$

where λ is the neutron wavelength outside the field, and ($\pm$) refer to the two spin states parallel and antiparallel to the magnetic induction. In our experiment we let the neutron which initially fulfilled the exact Bragg condition enter on its way to the crystal a magnetic field with a gradient parallel to its momentum. The field is then constant over the region where diffraction in the crystal takes place. For this case we obtain by combination of Eqs. (3), (5) and (8),

$$y_\pm = \mp(\mu B/|V_G|)\sin^2\theta_B . \tag{9}$$

As a numerical example, a magnetic field of only 1.66 kOe (0.166 T) results in an energy change of 10^{-8} eV and thus gives a neutron-propagation direction of 5.1° from the lattice planes (again, for the 1.865-Å-wavelength neutrons used and Si (400) diffraction). It is worthwhile to notice from Eq. (9) that the effect depends on the ratio of the interaction potentials of the neutron with the magnetic field and with the crystal, and only indirectly via the Bragg criterion on the neutron energy.

IV. EXPERIMENT DESCRIPTION AND RESULTS

The experiment was performed using the monolithic two-crystal neutron interferometer which was described earlier.[8] This crystal system shown in Fig. 1 was designed for phase interferometry, although this feature was not exploited in the present study. The incoming radiation of wavelength $\lambda = 1.865$ Å was obtained by asymmetric (400) Laue diffraction from a mosaic Ge crystal. In the interferometer crystal the symmetric Si (400) reflection was used. The interferometer crystal consists of two crystal plates of equal thickness (9.21 mm) which are connected rigidly by a common base; this results in good angular definition and stability. The first crystal acted as a "crystal collimator" as proposed by Shull.[9] An entrance slit and an exit slit, each of width 1.40 mm as measured along the crystal face, were attached to this crystal plate in order to select the central part of the radiation distributed over the Borrmann fan, i.e., those neutrons which traveled through the crystal in directions close to the lattice planes as shown in Fig. 1(a).

When this radiation enters the second crystal plate, it propagates again along the crystal planes which are exactly parallel to those in the first plate. Thus the intensity profile as a function of position on the back face of the second crystal shows a single peak as measured by scanning with a 1.40-mm wide slit (bottom curve in Fig. 2).

The interferometer was arranged in the air gap of an electromagnet in such a way that the point where the neutrons enter the second crystal plate was in the center of the circular-pole faces of 50.8-mm diam. [see Fig. 1(b)]. This ensures that the neutrons, on their way from the first to the second crystal plate, see a field gradient which is oriented along their path, thereby producing no angular deviation. Since the neutron path between the crystals was 51.7 mm long, this geometry implies that the first crystal plate was also subjected to a sizable magnetic fringe field surrounding the electromagnet gap. Measurements of this fringe field at the entrance and exit slits on the first crystal showed it to be 28.7% and 36.0%, respectively, of the homogeneous field present at the second crystal. All experimental results were related to an effective field B_{eff}, defined as the difference between the mean value of the magnetic field at the first crystal plate and the stronger homogeneous field at the second crystal plate. The measurement of the neutron-propaga-

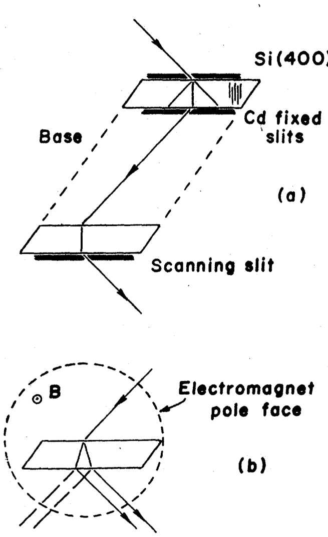

FIG. 1. (a) Two-crystal system used with the first crystal plate acting as a collimator. The full Borrmann fan is indicated. (b) Neutron propagation in the second crystal plate when a magnetic field is applied.

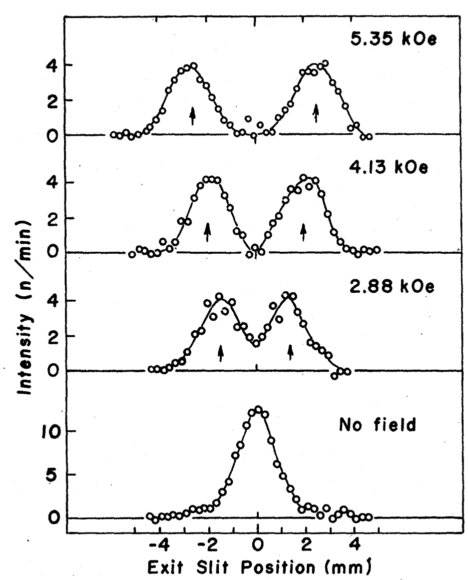

FIG. 2. Neutron intensity released from the backface of the second crystal in the double-diffracted direction as scanned with an exit slit. The parameter is the effective magnetic field difference between the second and first crystal plates.

tion direction was then performed for various values of B_{eff} by scanning an exit slit across the rear face of the second crystal, with the results shown in Fig. 2. Using Eq. (9) with values of B_{eff} of 2.88, 4.13, and 5.35 kOe, one calculates the corresponding values of y to be 0.163, 0.233, and 0.302, respectively. The expected peak positions for these field values are indicated by the arrows in Fig. 2, and these are in good agreement with the observations.

It is seen that the intensities measured in the experiment are very low because of the very high selectivity of the first crystal in preparing central radiation. The intensity shown has been corrected for background radiation, which amounted to about 2 neutrons/min. It was found in the course of the experiment that broadening and shifts in peak position were encountered because of changing ambient temperature conditions in the electromagnet assembly. Temperature differences between the two crystals show up very sensitively on the peak positions, and had to be minimized by thermal shielding and ventilation within the electromagnet. In illustration of this sensitivity, we calculate that a temperature difference of 0.1°C between the two crystal plates will cause by linear contraction a peak splitting equivalent to the half-width of the peak. In spite of these experimental problems, the agreement with theoretical prediction as shown in Fig. 2 is very good.

The magnetic fields used in the experiment correspond to neutron-energy changes of 1.74×10^{-8}, 2.49×10^{-8}, and 3.23×10^{-8} eV, respectively. The resolution limit of the present system is therefore about 10^{-8} eV. This can be compared with the resolution of the backscattering technique, which has been used to detect very small energy changes,[10] and which is in the range of about 10^{-7} eV. Thus, the use of the backscattering technique proposed by Funahashi[11] to measure the Zeeman splitting of the energy would require much higher fields than those used in the present work.

V. POLARIZATION EFFECTS

The wavelength change of Eq. (8) is numerically equal for the two neutron spin-state groups but of opposite sign. This implies that each spin state follows the same two paths in the second crystal plate, and hence the split peaks in Fig. 2 are unpolarized if the incoming neutrons are. Some specific polarization effects are to be expected in this case if polarized incident neutrons are used. This arises because the two spin-state neutron groups travel along a given common path as different wave fields and hence experience different phase shifts.[3] Because of intensity limitations, no attempt was made to measure effects of this kind in the present study.

However, there are polarization effects to be expected even with unpolarized incident neutrons. A separation of the spin states within the second crystal plate can be achieved by refracting the neutron beam by a nonmagnetic prism along its path from the first to the second crystal plate, thus causing the same directional change $\delta\theta$ for both spin states. In this case the change in y caused by refraction and by the magnetic field have the same sign for one spin state and opposite sign for the other one. This implies that the neutron-propagation directions within the second crystal plate are different for the two spin states. A special case arises if the y values induced by the magnetic field and by the prism are numerically equal. For this case, neutrons of one spin state arrive at the second crystal under exact Bragg conditions and travel along the lattice planes, while the splitting effect for the other spin-state group is doubled on the y scale. Thus we expect three polarized beams, spatially separated, to leave the second crystal plate, with the polarization of the central peak being opposite to the polarization of the two side beams (Fig. 3). The direction of polarization of these beams depends upon the direction of the prism apex angle, and thus an inversion of the prism changes the sign of $\delta\theta$ and the role of the two spin-state groups is interchanged.

The composite action of both prism refraction and magnetic field splitting has been studied with an aluminum prism of apex angle $\phi = 35°$ arranged in symmetric transmission. The neutron deviation angle through the prism is given by

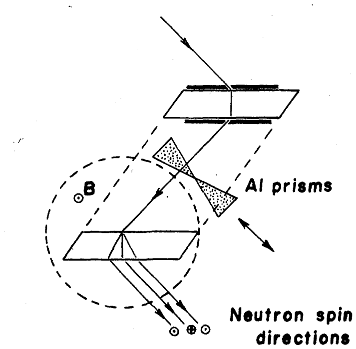

FIG. 3. Experimental arrangement used to separate the spin states by simultaneous prism refraction and magnetic field action. The spin directions indicated correspond to the prism position shown.

$$\delta\theta = 2(n-1)\tan(\phi/2), \tag{10}$$

where n is the Al index of refraction. In our experiment this angle is calculated to be $\delta\theta = 7.25 \times 10^{-7}$ rad $= 0.150$ arc sec. According to Eq. (3) this results in a y change of 0.329. The top curve in Fig. 4 shows the measured intensity profile with the prism in place and no magnetic field on the crystal. The arrows show the expected peak positions. The lower curve in this figure shows the intensity profile with the prism in place and a magnetic field of $B_{\text{eff}} = 5.45$ kOe. The arrows again indicate the theoretically expected peak positions, which are in reasonable agreement with observation. The increased width of the central peak in this curve is explainable by the fact that the y values of the Al prism ($y = 0.329$) and of the magnetic field ($y = 0.308$) did not match exactly. This results in the central peak in the experiment being composed of two peaks at slightly different positions.

As noted above, the peaks in the lower curve of Fig. 4 are expected to be completely polarized. To demonstrate this, the polarization of the central peak was studied by transmission through a polycrystalline iron plate (thickness 9.5 mm) magnetized by a field of 13.5 kOe. The low intensity available in the beam precluded the customary use of a polarization analyzing crystal. The transmitted intensity was measured with the neutron polarization being inverted by alternate insertion of oppositely directed aluminum prisms into the beam between the two crystal plates, as shown in Fig. 3. For this experiment the value of B_{eff} was adjusted so as to be exactly matched to the prism relative to their deflection effects.

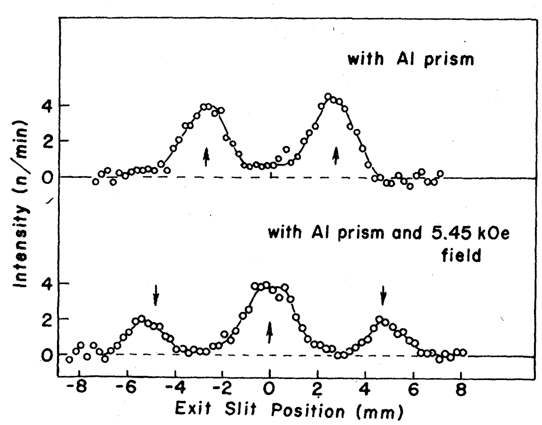

FIG. 4. Intensity released from the backface of the second crystal plate as a function of exit slit position when a 35° Al prism refracts the neutrons (top) and when simultaneously a magnetic field of $B_{\text{eff}} = 5.45$ kOe is applied (bottom).

A guide field was used to maintain the beam polarization between the two separated electromagnets. It was found that the intensity transmitted through the Fe plate was higher with the prism configuration as shown in Fig. 3. This implied that the neutron-spin direction was opposite to the field direction in the two electromagnets, in agreement with that expected.

Following a lengthy series of measurements with alternate prism orientation inverting the sense of neutron polarization, the polarization ratio (the ratio of intensities upon polarization reversal) of the central peak as transmitted through the iron analyzing plate was determined to be 1.36 ± 0.05. A separate experiment was then performed to calibrate the polarization sensitivity of the iron plate by using a completely polarized beam from a Co-Fe polarizing crystal (of the same wavelength and with half-wavelength absorption by a samarium filter). This gave a polarization ratio of 1.405 ± 0.005 and when combined with the above central-peak observation, the neutron polarization in the central peak was calculated to be $(96 \pm 9)\%$ after small correction for the $\lambda/2$ component. Thus the degree of polarization in the separated beams from the silicon crystal has been established as being very high and of the expected sense.

VI. CONCLUSIONS

The experiments reported here demonstrate some of the effects to be expected from the dynamical theory of magnetic neutron diffraction. In particular, diffraction by a nonmagnetic crystal in an external field as investigated here is analogous to using a purely nuclear diffraction peak (with only nuclear contribution to the structure factor) in a ferromagnetic crystal. The feature that the direction of neutron propagation within the crystal can be changed through large angles by applying a magnetic field of moderate strength is to be contrasted with the minute directional changes caused by magnetic field gradients or by magnetic refraction at boundaries. This feature, interesting by itself, was used to determine directly the wavelength change of the neutron due to Zeeman splitting in a magnetic field. The corresponding energy resolution in the present experiment was about 10^{-8} eV. It is to be expected that this sensitivity can be improved by another order of magnitude if thicker crystals, larger wavelengths, and higher-order reflections (lower V_G) are used. Whether or not this high-energy resolution feature can be exploited in future inelastic scattering experiments is open to consideration.

It has also been demonstrated that a polarized beam can be produced by the application of a magnetic field to a nonmagnetic crystal. It would be interesting to extend the present study to the case where field gradients, rather than homogeneous fields, are applied to the crystal. In this case there would be a force acting on the neutron while it propagates through the crystal, a situation not dealt with by the existing dynamical diffraction theory. In such an experiment curved trajectories of the neutron within the crystal are to be expected with, again, much larger curvature than in a conventional Stern-Gerlach experiment. These experiments and more detailed verification of the predictions of the polarization behavior will preferably be performed at high-flux beam reactors, thus avoiding the severe intensity limitation of the present study.

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