Observation of the pendellösung fringe effect in magnetic scattering of neutrons

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The inclination method revealed that the intensity of magnetic, nuclear, and mixed (nuclear–magnetic) neutron scattering by a "perfect" weakly ferromagnetic $FeBO_3$ crystal oscillates when the optical thickness of a sample is varied. The positions of the oscillations (pendellösung fringes) were in agreement with the predictions of the dynamical theory. An external magnetic field influenced the contrast of the oscillations, and the direction of the field affected the positions of the oscillations in the magnetic and mixed scattering cases. The structure factors were determined to within ~1% from the pendellösung fringe positions.

INTRODUCTION

When radiation interacts with a perfect crystal, a wave field appears in this crystal and a special feature of this field is a periodic "transfer" of energy from a primary to a diffracted wave and back again as the radiation penetrates a crystal.[1,2] The interference between these waves gives rise to a number of dynamical phenomena, one of which is the pendellösung fringe effect manifested by oscillations of the scattered-radiation intensity (under Laue diffraction conditions) as a function of the radiation wavelength or the thickness of a crystal. This effect has been observed many times and has been studied thoroughly in the scattering of x rays and neutrons.[3–6] Since these dynamical effects are universal in nature, they should be exhibited also in the case of magnetic neutron scattering.

The different aspects of neutron diffraction in "perfect" magnetically ordered crystals have been considered theoretically by many authors,[7–10] but experimental attempts to detect the pendellösung fringe effect in magnetic scattering of neutrons have been unsuccessful because the sample quality has been inadequate.[11,12] Using the anisotropy of the distribution of defects in an imperfect yttrium iron garnet crystal (cut in such a way as to weaken the influence of defects on the scattering) and applying the technique of polarized neutrons, the authors of Ref. 13 observed for one of the polarizations a maximum in the dependence of the scattered-radiation intensity on the wavelength. This maximum agreed qualitatively with predictions of the dynamical theory.

Our experimental investigation of the pendellösung fringe effect was carried out using thin (of the order of the extinction length) magnetic crystals, for example those of $FeBO_3$, which behave as "perfect" crystals in the scattering of x rays and Mössbauer γ photons.[14,15]

EXPERIMENTAL METHOD AND SAMPLES

Our experiments were carried out in the wavelength range 2.0–2.4 Å using a MOND single-crystal neutron deflectrometer and the IR-8 reactor at the Kurchatov Institute of Atomic Energy. A monochromatic neutron beam was formed using a double monochromator[16] with pairs of Zn (002) or pyrolytic graphite (022) crystals. The contribution of the higher orders of neutron reflection was reduced to ~2% by quartz and pyrolytic graphite filters.

The pendellösung fringes were observed by the inclination method.[6] A study was made of the dependence of the scattered-radiation intensity on the optical thickness of a crystal, which was varied by rotating a sample about the normal to the reflecting plane by a method described in Ref. 17. In view of the thinness of the crystal, several oscillations were observed when it was tilted by angles up to 80° so as to increase the optical thickness by a factor of 6–7. It was known[18] that an increase in the angle of inclination reduced the contrast of the pendellösung fringes owing to various experimental factors (vertical and horizontal divergence of the beam, its nonmonochromaticity, and inhomogeneous distribution of the crystal thickness), so that for angles of inclination in excess of 60° the fringes could disappear completely. We therefore carried out experiments on "perfect" plane-parallel Si crystals, which demonstrated (Fig. 1) that a reduction of the angular divergence of the beam could increase the oscillation contrast at high angles of inclination. For example, when the vertical divergence was 5.5′ and the horizontal was 9′, the contrast of the oscillations with the serial numbers of the order of 20 amounted to half the theoretical value for angles of inclination close to 80°.

Our experiments were carried out on a $FeBO_3$ crystal grown at the Physics Institute of the Czechoslovak Academy of Sciences in Prague by a method described in Ref. 15. Our sample was a plate with the (111) large face, of dimensions 5×9 mm, and ~70 μm thick (the thickness fluctuations were 15 μm). The rocking curves, determined for different parts of a crystal using an x-ray diffractometer, showed that the widths of these curves (8″ ± 0.5″) were close to the theoretical value (7.5″). In several control experiments we used an $FeBO_3$ crystal 120 μm thick which was less perfect (30″).

Crystalline $FeBO_3$ is a weak ferromagnet ($T_n = 348$

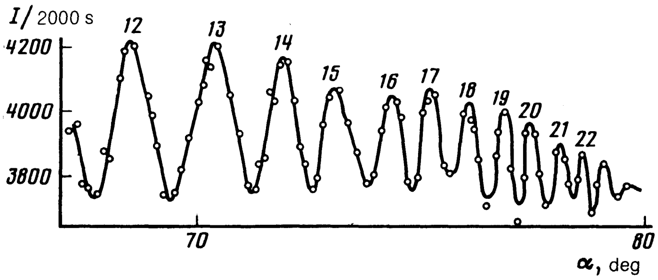

FIG. 1. Dependence of the intensity I of neutrons scattered by a "perfect" Si (220) crystal on the angle α of inclination from the normal to the reflecting plane. The horizontal divergence of the beam was 9′ and the vertical divergence was 5.5′; $\lambda = 2.0$ Å.

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K) with the (111) easy magnetization plane. Its structure and properties were described in Ref. 19. In some of our experiments the crystal was converted to a single-domain state by the application of a saturating field of 60 Oe directed parallel to the easy-magnetization plane. The structure factor for the magnetic scattering of neutrons could be altered by a suitable selection of the angle between the direction of the magnetic field, which governed the direction of the antiferromagnetic vector perpendicular to the field, and the scattering vector.[20]

The influence of fluctuations of the thickness on the scattering pattern was reduced by limiting the beam with a slit of ~1 mm width at the entry.

THEORETICAL DEPENDENCE

An analysis of the atomic and magnetic structures of iron borate shows that three different types of reflection are possible when neutrons are scattered by this compound: purely magnetic (scattering by the magnetic moments of the iron atoms), purely nuclear (when the magnetic contribution is negligible), and mixed (when the magnetic scattering occurs on the iron atoms and the nuclear scattering on the oxygen atoms). In the case of the first two types of scattering the integral reflection coefficient is of the form[1]

$$R=\frac{\lambda^2|F|}{2V\sin(2\theta_B)}\left(\frac{\cos\gamma_h}{\cos\gamma_0}\right)^{1/2}\int_0^{2\pi t_0/t_e} J_0(x)\,dx, \qquad (1)$$

where λ is the wavelength; F is the structure factor; V is the cell volume; θ_B is the Bragg angle; t_0 is the thickness of the crystal; t_e is the extinction length; γ_h and γ_0 are the angles between the normal to the surface of the sample and the directions of the incident and the reflected beams; and $J_0(x)$ is a Bessel function of zeroth order.

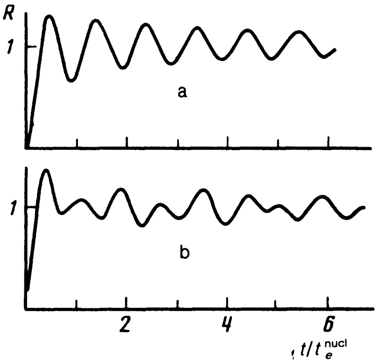

FIG. 2. Theoretical dependence of the integral reflection coefficient for the nuclear and magnetic scattering of neutrons on the thickness of an antiferromagnetic crystal, expressed in terms of the extinction length (a), and of the corresponding reflection for mixed scattering of neutrons by a FeBO$_3$ (210) crystal on the thickness of this crystal, expressed in terms of the extinction length of the nuclear scattering of neutrons in the presence of an external magnetic field applied along the $[10\bar{1}]$ direction (b).

Using Δ for the angle between the normal to the reflecting plane and the main plane of the investigated plate, we can describe the dependence of the integral reflection coefficient on the experimentally determined angle of inclination α of the crystal by substituting

$$\begin{aligned}\cos\gamma_0&=\sin\theta_B\sin\Delta+\cos\theta_B\cos\Delta\cos\alpha,\\ \cos\gamma_h&=-\sin\theta_B\sin\Delta+\cos\theta_B\cos\Delta\cos\alpha\end{aligned} \qquad (2)$$

In the mixed scattering case the integral reflection coefficient is the sum of the integral reflection coefficients of

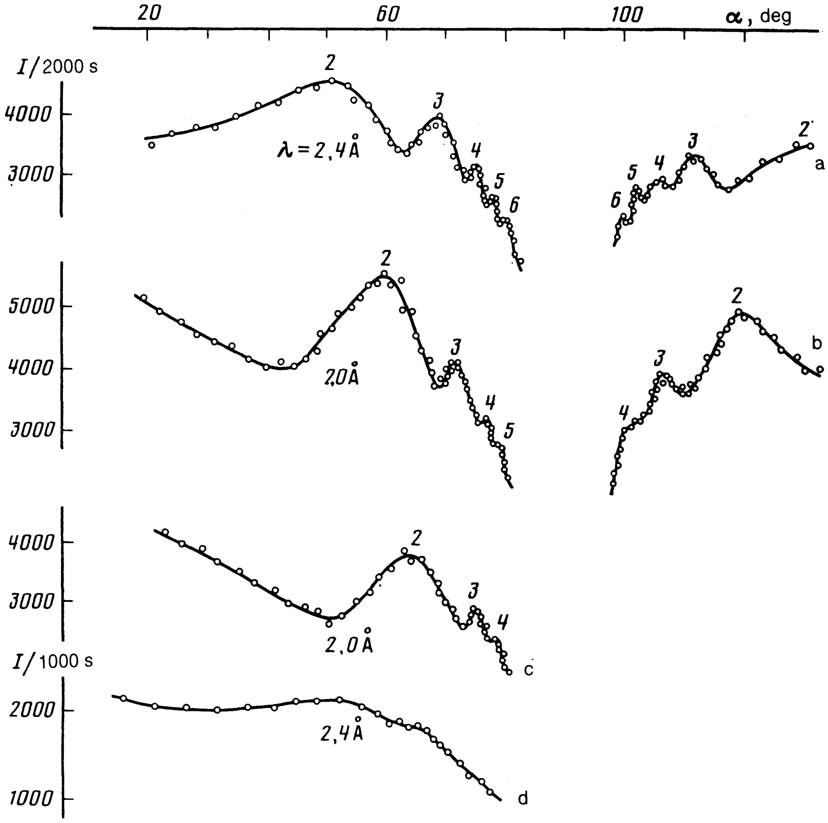

FIG. 3. Dependence of the intensity I of the magnetic reflection of neutrons on the angle of inclination of a FeBO$_3$ (100) crystal. The experimental results labeled *a* and *b* were obtained in a field directed parallel to the $[\bar{2}11]$ axis to ensure the maximum value of the magnetic structure factor. In the case of curve *c* the magnetic structure factor was reduced by rotating the direction of the field at an angle of 50° in the (111) plane. In the case of curve *d* the experiments were carried out in the absence of a field.

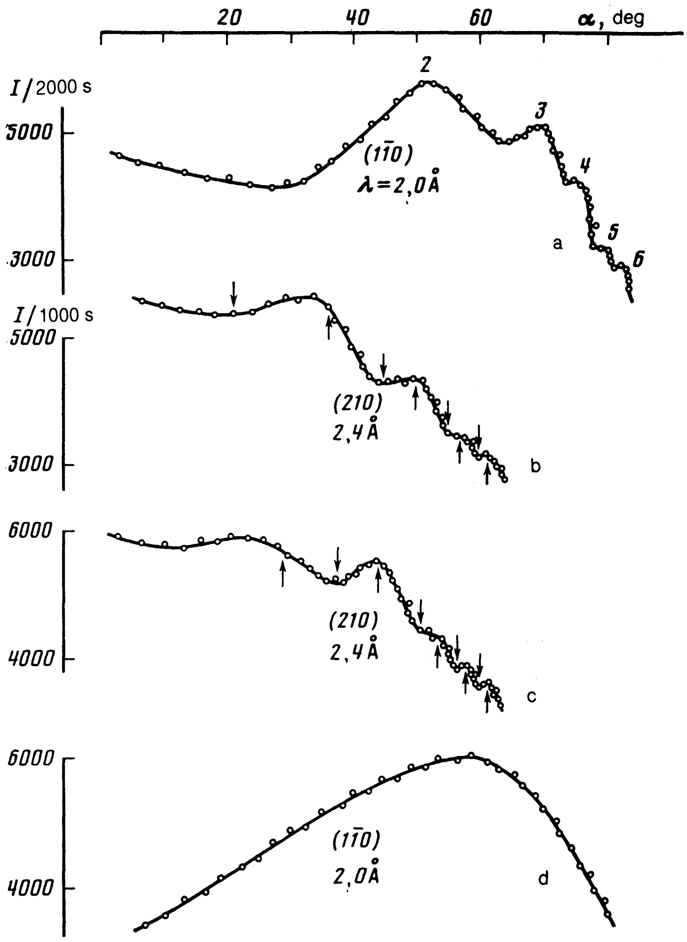

FIG. 4. Dependences of the intensity of the nuclear (1$\bar{1}$0) (*a*) and mixed (210) scattering of neutrons on the angle of inclination of a "perfect" $FeBO_3$ crystal. The direction of the field was [10$\bar{1}$] and [$\bar{1}2\bar{1}$] for the curves labeled *b* and *c*. The curve labeled *d* represents the dependence of the intensity of the nuclear scattering of neutrons by an imperfect $FeBO_3$ crystal on the angle of inclination. The arrows identify the calculated positions of the oscillations.

neutrons of two polarizations diffracted by an antiferromagnetic crystal independently of one another[10] governed in one case by the sum of the nuclear and magnetic structure factors, and in the other case by the difference between these factors.

It follows from Eq. (1) and the results of the calculations presented in Fig. 2 that in the pure nuclear and pure magnetic cases the intensity of the reflected radiation should depend in an oscillatory manner on the optical thickness of a crystal, in a manner governed by the nuclear or magnetic extinction length, whereas in the mixed reflection case there should be a more complex pattern of "beats" governed by the simultaneous influence of both (nuclear and magnetic) extinction lengths.

EXPERIMENTAL RESULTS

Figures 3–5 give the results of neutron experiments carried out by the inclination method in the case of the magnetic (100), nuclear (1$\bar{1}$0), and mixed (210) reflections. In all cases a thin "perfect" $FeBO_3$ crystal exhibited oscillatory dependence of the reflected-radiation intensity on the thickness and up to 5–6 oscillations were observed clearly. The positions of the maxima and minima were in good agreement with the calculations carried out using Eq. (1). The contrast of the oscillation pattern was less than theoretical, and it rapidly decreased with increasing optical thickness.

The angular positions of the pendellösung fringes were determined more accurately when the angle of inclination was greater than 90°. Oscillations with identical serial numbers were located symmetrically on both sides of this angle, and the contrast was approximately the same. The number of the observed oscillations decreased as the neutron wavelength was reduced from 2.4 to 2.0 Å (Figs. 3a and 3b), and also as the magnetic structure factor decreased (Figs. 3b and 3c). This is due to a change in the direction of the magnetic field in the plane of the crystal, resulting from the increase in the extinction length. The integral reflection coefficient exceeded the theoretical value by a small amount (20%) so that the difference between the pendellösung fringe contrast and the theoretical value was not due to defects, but to different thicknesses of the sample, as shown in Ref. 18. In the demagnetized state there were practically no oscillations in the magnetic scattering (Fig. 3d), which could be attributed naturally to the influence of magnetic

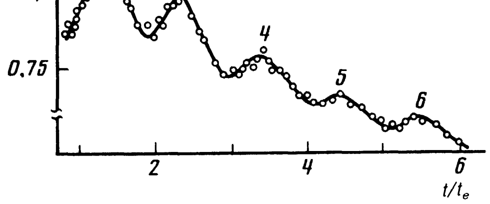

FIG. 5. Dependence of the intensity of the magnetic reflection of neutrons by a $FeBO_3$ (100) crystal on the ratio of the thickness of the crystal to the extinction length.

domains. In the nuclear scattering case the contrast of the oscillations decreased by 20 %, which corresponded to the influence of the magnetic domain structure on the quality of a $FeBO_3$ crystal determined by x-ray diffraction.[21]

The intensity of the neutron scattering of a $FeBO_3$ crystal with more defects but with plane-parallel surfaces increased smoothly (without oscillations) as a function of the angle of inclination and then fell because of the absorption (Fig. 4d), which was typical of the kinematic scattering in a mosaic crystal.[20] The integral reflection coefficient was then approximately three times the theoretical value for a perfect crystal.

CONCLUSIONS

Our experiments demonstrated directly the occurrence of the dynamical magnetic scattering of neutrons, manifested by oscillations of the dependence of the intensity of the reflected neutrons on the thickness of the sample, in agreement with the dynamical theory in the case of the purely magnetic and mixed reflections. The observation of these oscillations for colder neutrons could be used in precision determination of the magnetic form factors and of the distribution of the spin density in a crystal. In particular, in our experiments the absolute value of $F^{\rm mag}_{(100)} = 1.23\times 10^{-12}$ cm was deduced from the oscillation period to within $\pm 1\%$, i.e., the error was smaller than usually attained in the intensity measurements.

Clearly, the range of investigated samples can be fairly wide if we use sufficiently thin crystals, as was done in the present study. We were thus able to observe analogous dynamical effects in thin α-Fe_2O_3 crystals.

Finally, a study of the contrast of the oscillations obtained for different reflections provides new opportunities for the investigation of the magnetic and atomic defects and of their interaction with one another.

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