

# Determination of the spatial orientation of neutron polarization and investigation of magnetization near a phase-transition point

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A new method of analyzing the polarization of a neutron beam is described. It is based on a non-adiabatic projection of the polarization on the direction of the magnetic field and makes it possible to determine the spatial orientation of the polarization vector. A similar three-dimensional analysis of the polarization was used to investigate the magnetization of iron-yttrium iron garnet near the Curie temperature. The spontaneous magnetization  $M_s$  has a macroscopic spatial inhomogeneity, and the temperature dependence of  $M_s(T)$  has a variable critical exponent  $\beta$  up to  $\beta < 1$ .

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## 1. INTRODUCTION

Slow polarized neutrons have found wide use in the study of magnetic materials, because of their high sensitivity to the magnetic structure of crystals and to magnetic inhomogeneities (domains, critical fluctuations of the magnetic density, spin waves, etc.<sup>[1-3]</sup>). In measurements of this type, the information is obtained from the depolarization of a beam passing through the sample in the forward direction, or scattered through a certain angle. Since we shall deal henceforth with rotation of the polarization vector, we agree to use the term "depolarization" for the change in the absolute value of  $|\mathbf{P}|$ , and not merely the change of the polarization along a preferred direction, as is frequently done.

Beam depolarization is connected with magnetic inhomogeneities the characteristic dimension  $d$  of which is much smaller than the transverse dimension  $D$  of the beam passing through the sample ( $\lambda \ll d \ll D$ , where  $\lambda$  is the neutron wavelength), when averaging over the beam cross section yields only the change of  $|\mathbf{P}|$ . Magnetic inhomogeneities with dimensions  $d \gtrsim D$  lead to homogeneous precession of the polarization about the effective field of the sample, and in final analysis to rotation of the vector  $\mathbf{P}$  of the transmitted beam. In addition, an apparent rotation of  $\mathbf{P}$  (together with a change of the modulus  $|\mathbf{P}|$ ) is brought about also by depolarization of the beam by minute inhomogeneities in the case when the vector  $\mathbf{P}_0$  in the incident beam is not strictly parallel or perpendicular to the neutron velocity  $\mathbf{v}$ .<sup>[4]</sup> In experiments with polarized neutrons, as a rule, it is necessary to orient  $\mathbf{P}_0$  in a definite manner in the incident beam and determine the direction of  $\mathbf{P}$  of the scattered neutrons. In typical experiments, the relations between the Larmor precession frequencies  $\omega_L$  and the characteristic dimensions of the components of the apparatus are such that adiabatic, nonadiabatic, and mixed conditions for the passage of the neutron beam between the polarizer and the analyzer are possible. These conditions can be particularly specified, but can also arise by themselves, uncontrollably, owing to imperfections in the adjustment of the apparatus and a non-optimal choice of the magnetic fields. In the latter case measurement of only one projection of the polarization can lead to a distortion of the experimental results.

This paper is devoted to a new approach to the measurement of polarization, namely a three-dimensional analysis of the polarization, which makes it possible to

determine the spatial orientation of the polarization vector before and after the interaction with the object, thereby greatly increasing the amount of information gained from the experiment. In fact, the polarization vectors of the incident ( $\mathbf{P}_0$ ) and scattered ( $\mathbf{P}$ ) beams are connected by some matrix operator, which contains the spin-dependent part of the interaction of the neutrons with the object. For example, in the simplest case of the passage of neutrons through a uniform magnetic field, the matrix operator of the polarization rotation contains the direction of the precession axis  $\mathbf{m}$  (the field direction) and the precession angle  $\varphi$  (the magnitude of the field). By performing the experiment with two arbitrary (but known) directions of  $\mathbf{P}_0$  it is possible to reconstruct the rotation matrix and to find the values of  $\mathbf{m}$  and  $\varphi$ . This information cannot be obtained in the general case from measurements of only one polarization projection.

The described procedure of the vector analysis of the polarization can be used in all experiments where the neutron spin takes part in the interaction. In Sec. 4 we present certain results obtained by studying the spontaneous magnetization near the Curie temperature.

## 2. DESCRIPTION OF THE METHOD OF VECTOR ANALYSIS OF THE POLARIZATION

The vector-analysis method is based on projecting the vector  $\mathbf{P}$  on a specified magnetic-field direction, followed by guiding the neutron beam into the leading field of the apparatus without a change of the obtained projection.

The change of the polarization is described by the equation for the precession of the vector  $\mathbf{P}$  about  $\mathbf{H}$ :

$$d\mathbf{P}/dt = g[\mathbf{P} \times \mathbf{H}], \quad (1)$$

where  $g$  is the gyromagnetic ratio of the neutron and  $t$  is the time. If the direction of  $\mathbf{H}$  does not change, then the projection of  $\mathbf{P}$  on the field  $\mathbf{H}$  remains constant. This parallel component  $P_H$  is the one usually measured with magnetized crystals or mirrors. If the direction of the field  $\mathbf{H}$  varies with frequency  $\omega_0$  in terms of the coordinates of the moving neutron, then two limiting cases are of interest—adiabatic and nonadiabatic passage of the neutrons through the region of the field variation; these cases are determined by the ratio  $K = \omega_L/\omega_0$ . In the case of nonadiabatic passage ( $K \ll 1$ ) the spatial orientation of the neutron polarization does not change and, ac-

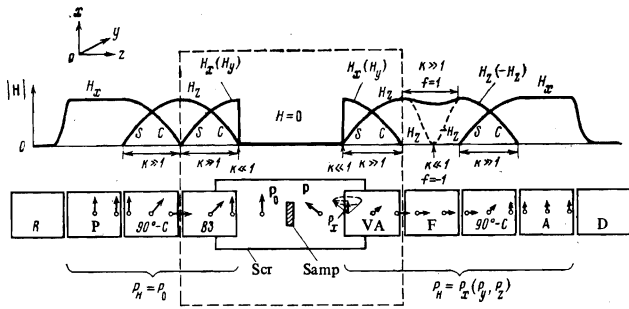


FIG. 1. Block diagram of the setup for the three-dimensional analysis of the polarization. Upper curves—distribution of the magnetic field  $H$  along the axis of the neutron beam. Symbols:  $S = \sin(\pi z/2L)$ ;  $C = \cos(\pi z/2L)$ ;  $K = \omega_L/\omega_0$ ;  $f$ —state of the flipper; R—reactor, P—polarizer mirror;  $90^\circ-C$ — $90^\circ$  field matching unit; VS—unit to set the direction of the polarization along the axes  $x$ ,  $y$ , and  $z$ ; Scr—magnetic screen; Samp—sample; VA—vector analyzer; F—spin flipper; A—mirror analyzer, D—neutron detector.

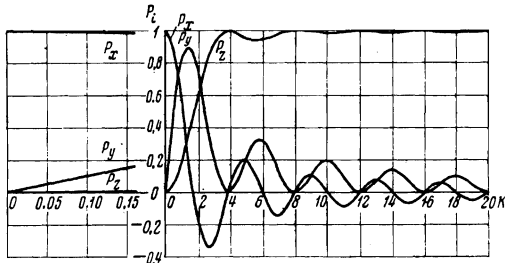


FIG. 2. Change of projections  $P_i$  following  $90^\circ$  rotation of the magnetic field as a function of the adiabaticity coefficient  $K = \omega_L/\omega_0$ .

According to (1), what is conserved subsequently (at constant  $H$ ) is the projection  $P_H$ . If  $K \gg 1$ , the polarization vector changes its direction in space and follows the direction of the magnetic field.

A general block diagram of the setup for the vector analysis of the polarization, together with the distribution of the magnetic field along the neutron beam, is shown in Fig. 1. The three-dimensional analyzer proper includes the following units: VS—unit that sets the direction of the polarization  $P_0$ , Scr—magnetic screen with the sample Samp, VA—analyzer of the projection of the vector  $P$ . The input and output of this block (the dashed box of Fig. 1) are connected with the remainder of the apparatus through magnetic-field matching units  $90^\circ - C$ , in which the magnetic field is smoothly rotated through  $90^\circ$ , thus ensuring the adiabaticity condition. In front of the analyzer mirror is a spin-flipper to rotate the neutron spin  $180^\circ$  relative to the field. This flipper can be placed also in front of the sample if it is necessary to reverse the polarization in the incident beam. The field scale in the figure is arbitrary. Actually the field in the polarizer is 500 Oe, and in the units  $90^\circ - C$ , VS, VA, and F it amounts to several dozen oersteds; homogeneous leading magnetic fields of arbitrary length can be produced between the units shown in the figure.

In the region with  $H = 0$ , the direction of  $P$  is fixed in space in accord with Eq. (1), so that  $dP/dt = 0$  at  $H = 0$ . The entrance into the region VA, which has a length  $L$  along the beam, is nonadiabatic ( $K \ll 1$ , projection takes place). The field  $H$  is then rotated uniformly through  $90^\circ$  over the base  $L$  until it coincides with the leading field of the apparatus (in this case with the input field of the flipper), thereby ensuring the adiabaticity conditions ( $K \gg 1$ ) and maintaining  $P_H$  constant.

Figure 2 shows the ratios of the projections of the vector  $P$  after the neutrons traverse a field region of length  $L$  in which the direction of the magnetic field is rotated through  $90^\circ$ , as functions of  $K = \omega_L/\omega_0 = 2gHL/\pi v$ . These relations follow from the solution of Eq. (1). It is seen from the figure that in addition to the limiting case ( $K \rightarrow \infty$ ) there are discrete regions ( $K = 3.8$ ,  $K = 7.8$ , etc.) in which the  $90^\circ$  rotation is effected with good accuracy ( $P_z > 0.99$ ) for a nonmonochromatic beam  $\Delta v/v \lesssim 15-20\%$ . These values of  $K$  can be used if necessary to decrease the dimension of the region  $L$ . In this case the quantity  $\Delta v/v$  given above characterizes either the possible nonmonochromaticity of the beam, or the accuracy requirements that  $H$  and  $L$  must satisfy for a monochromatic beam.

The input field of the VA unit can be directed along any of the three axes of the chosen coordinate system. The obtained polarization projections  $P_{x,y,z}$  likewise correspond to these directions. The described VA unit can be used to set the vector  $P_0$  at the input in the region of  $H = 0$ , i.e., the VS unit is analogous to the VA unit rotated  $180^\circ$  relative to the beam axis. By specifying and measuring  $P_0$  and then placing the investigated sample in the region  $H = 0$  and measuring  $P$  in the transmitted beam, we can obtain the information connected with the depolarization and rotation of the polarization vector in the sample.

### 3. CONSTRUCTION OF VECTOR ANALYZER

The vector-analyzer designs developed in our laboratory took into account the conditions necessary for the guidance of the neutron beam through the system of magnetic fields. Two designs were tested: an electromagnet with an iron yoke and a system of solenoids.

In the first analyzer (Fig. 3a) the field  $H_x$  was produced with an electromagnet having a cylindrical yoke, the pole pieces of which were shaped to ensure the following field distribution along the  $z$  axis:

$$H_x = H_0 \cos(\pi z/2L). \quad (2)$$

The magnet was mechanically rotated about its axis (the  $z$  axis), so that any field direction  $H_x$  could be set in the  $xy$  plane. The solenoid producing the field  $H_z$  was secured in the magnet yoke. The direction of the field  $H_z$  coincided with the  $z$  axis and corresponded to the distribution

$$H_z = H_0 \sin(\pi z/2L). \quad (3)$$

Conditions (2) and (3) ensured preservation of the sum-mary field  $H_0 = (H_x^2 + H_z^2)^{1/2}$  on the base  $L$  and  $90^\circ$  rotation of the field direction until it coincided with that of the leading field of the apparatus.

The second design consisted of three mutually perpendicular solenoids (Fig. 3b). The fields  $H_{x,y}$  in the directions  $x$  and  $y$  were produced by solenoids wound on

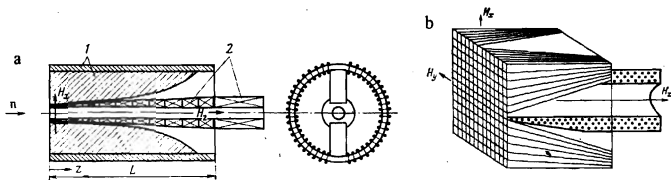


FIG. 3. Construction of three-dimensional analyzer: a) electromagnet: 1—magnet yoke, 2—solenoids; b) analyzer in the form of a system of solenoids.

corresponding perpendicular faces of a cube in such a way that the field distribution satisfied the condition (2). To decrease the stray field, the cube with the rectangular solenoids was placed snugly in a Permalloy magnetic screen. This design has many advantages over the first, mainly when it comes to automating the measurements.

The analyzer length and the field were  $L = 100$  mm and  $H = 10$  Oe. The residual field in the screen was  $H_{scr} \approx 10^{-3}$  Oe, so that at the real field distribution of the analyzing apparatus along the  $z$  axis it was possible to estimate the nonadiabaticity of the entry of  $\mathbf{P}$  into the analyzing apparatus at  $K \approx 0.01$ .

The measurement of the projections of the vector  $\mathbf{P}$  consists in the following: when measuring the projection  $P_x(P_y)$ , the field  $H_x(H_y)$  along the  $x(y)$  axis respectively is turned on, as well as the field  $H_z$ , and when measuring the projection  $P_z$  the fields  $H_{x,y}$  are turned off and the vector  $\mathbf{P}$  is projected on the field  $H_z$ .

The flipper used by us to reverse the neutron spin during the measurement of the degree of polarization has been described in [1,5]. It is based on the same principle of nonadiabaticity of the passage of the neutrons through the boundary separating two oppositely-directed fields. The value of the field near the boundary is close to zero, and this ensures  $K \ll 1$ . The flipper is constructed in the form of two flat coils located on the ends of the cylindrical magnetic screen and producing fields along the beam axis, with subsequent rotation of the exit field towards the direction of the field  $H_{lead}$ . The efficiency of such a flipper is  $f > 0.995$  for a beam with monochromaticity  $\Delta v/v \approx 15\%$  and wavelength  $\lambda_{max} = 4 \text{ \AA}$  at the maximum of the spectrum.

The main task in the construction of the instrument is the choice and matching of the magnetic fields so as to produce the necessary conditions for the passage of the neutron beam. The results of the adjustment can be estimated by determining the ratio of the projections and the conservation of  $|\mathbf{P}|$  as  $\mathbf{P}_0$  is varied in accordance with a definite law at the entrance into the analyzing devices or, for example, by measuring  $P_{x,y,z}$  as a function of the angle  $\varphi$  of rotation of the analyzing field  $H_{x(y)}$  in the  $xy$  plane (i.e., by rotating the coordinate frame through an angle  $\varphi$  about the  $z$  axis) with the direction of  $\mathbf{P}_0$  unchanged. Figure 4 shows a typical calibration curve  $P_{x,y}(\varphi)$  for opposite directions of the fields  $H_{x,y}$ , with  $P_z(\varphi)$  constant at 0.02. The calibration curves make it

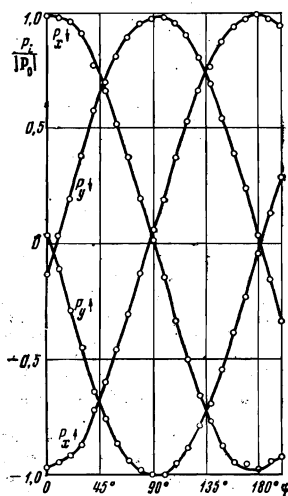


FIG. 4. Calibration curves plotted as the electromagnet-analyzer was rotated through an angle  $\varphi$  around the  $z$  axis. The arrows show the direction of the magnetic field.

possible to take into account the systematic errors connected with the small ( $\sim 2^\circ$ ) adiabatic rotation of the polarization vector at the entrance into the analyzing apparatus and with the deviation from orthogonality (on the order of  $1^\circ$ ) of the fields  $H_x$  and  $H_y$ . If the polarization projections are measured with a statistical accuracy on the order of 1%, the angular uncertainty of the vector  $\mathbf{P}$  is determined by a cone with apex angle  $\theta \leq (2/P)^\circ$ .

#### 4. USE OF THE VECTOR ANALYSIS PROCEDURE

The vector analysis of the polarization makes it possible to separate the depolarization of the beam as it passes through the sample from the rotation of the polarization at any given direction  $\mathbf{P}_0$  in the incident beam.

The first experiments with vector analysis of the polarization consisted of measuring the anisotropy of the depolarization of the neutron beam [6] relative to the neutron-velocity vector  $\mathbf{v}$  in isotropic samples. According to [4], owing to the connection between the polarization  $\mathbf{P}$  with the scattering vector  $\mathbf{e}$  in elastic magnetic scattering (this connection is  $\mathbf{P} = -\mathbf{e}(\mathbf{e} \cdot \mathbf{P}_0)$ ), the component  $P_\perp$  perpendicular to  $\mathbf{v}$  should change more strongly than the component  $P_\parallel$  parallel to  $\mathbf{v}$ , and the magnitude  $A$  of this anisotropy should be

$$A = (\ln P_\perp - \ln P_{0\perp}) / (\ln P_\parallel - \ln P_{0\parallel}) = 3/2. \quad (4)$$

To verify (4) we investigated samples of nickel pressed into tablets, with different effective nickel contents. The external field was  $H_{scr} < 0.04$  Oe, and  $\lambda \approx 4 \text{ \AA}$ . Experiments with different directions of  $\mathbf{P}_0$  in the incident beam have shown that the experimental values are in good agreement with the theoretical value  $A = 1.5$ . It is important to note that under the influence of such an anisotropy of the depolarization, in the case when the initial vector  $\mathbf{P}_0$  has components both parallel and perpendicular to the velocity, the vector  $\mathbf{P}$  is effectively rotated towards the  $\mathbf{v}$  axis. This must be taken into account in the real experiment, so as to separate correctly the processes that lead to a true rotation of  $\mathbf{P}$  and a true depolarization  $|\mathbf{P}|/|\mathbf{P}_0|$ .

At the present time, much attention is being paid to investigations of the behavior of the spontaneous magnetization  $M_S$  in the immediate vicinity of  $T_c$ . The critical exponent  $\beta$  ( $M_S \sim |\tau|^\beta$ , where  $\tau = (T - T_c)/T_c$ , which has in accord with similarity theory a value  $\beta = 1/3$ , ranges from 0.3 to 0.75 in different experiments and in different materials. This multiply-valued character of the experimental data can be attributed to the difficulty of the magnetic measurement methods, since application of an external magnetizing field distorts the phase transition. Measurements with neutrons offer advantages in this respect, since they can be performed locally and in zero magnetic fields. When the neutrons pass through the sample, the polarization vector precesses about the direction of the induction  $\mathbf{B}$ , and under certain conditions that ensure homogeneity of the field over the thickness of the sample it is possible to assess the magnitude and direction of  $\mathbf{B}$ . The precession angle is equal to  $\varphi = gB\delta/v \equiv \varphi_0 B\delta$ , where  $\delta$  is the sample thickness and  $\varphi_0 \approx 11 \text{ deg-G}^{-1}\text{-cm}^{-1}$  for neutrons with  $\lambda = 4 \text{ \AA}$ . This ensures a sensitivity  $\Delta B = 1 \text{ G}$  at  $\delta = 0.25 \text{ cm}$ . To find the value of  $M_S$ , which is proportional to  $\varphi$ , and the direction  $\mathbf{m} = \mathbf{M}_S/M_S$ , we use relations that follow from a geometric analysis of the rotation of  $\mathbf{P}$ . According to (1), the unit vectors  $\mathbf{n}_1 = (\mathbf{P}_1 - \mathbf{P}_{01}) / (|\mathbf{P}_1 - \mathbf{P}_{01}|)$  and

$\mathbf{n}_2 = (\mathbf{P}_2 - \mathbf{P}_{02}) / (|\mathbf{P}_2 - \mathbf{P}_{02}|)$ , where  $\mathbf{P}_{0i}$  and  $\mathbf{P}_i$  are the unit vectors of the polarization of the incident and transmitted neutron beams ( $i$  is the number of the experiment), lie in a plane perpendicular to  $\mathbf{m}$ , i.e.,  $\mathbf{n}_1 \perp \mathbf{m}$  and  $\mathbf{n}_2 \perp \mathbf{m}$ . Hence

$$\mathbf{m} = [\mathbf{n}_1 \times \mathbf{n}_2] / |[\mathbf{n}_1 \times \mathbf{n}_2]|, \quad (5)$$

and the precession angle  $\varphi$  is determined from the relation

$$\varphi = 2 \arcsin \frac{|\mathbf{P}_i - \mathbf{P}_{0i}|}{2[1 - (\mathbf{P}_{0i} \cdot \mathbf{m})^2]^{1/2}}, \quad (6)$$

i.e., by performing two experiments with different values of  $\mathbf{P}_0$  we can determine the magnitude and direction of the magnetic moment. In the case when the field is not uniform inside the beam and over the thickness of the sample, the problem of finding the magnitude and direction of the field has no unique solution, since the rotation operators do not commute. The combined rotation then characterizes a certain effective field  $\mathbf{M}_{\text{eff}}$ , the direction and magnitude of which depend on the direction of the incident polarization vectors  $\mathbf{P}_{0i}$ . Such an experiment can yield information on the ratio of the characteristic dimensions of the magnetic inhomogeneities and the beam cross section.

By way of illustration, Fig. 5 shows plots of the projection of the polarization vector of a beam passing through the sample when the sample is scanned by a neutron beam of 1 mm diameter near  $T_c$ , in the case of  $\text{Y}_3\text{Fe}_5\text{O}_{12}$  (external field  $10^{-3}$  Oe, neutron wavelength  $\lambda \approx 3 \text{ \AA}$ , displacement along the sample in steps of 0.8 mm). It is seen from the figure that the magnetization has a large-scale inhomogeneity over the sample cross section (characteristic dimension  $\sim 4$  mm). However, experiments with different values of  $\mathbf{P}_0$  do not yield self-consistent quantitative results, meaning that a smaller-scale inhomogeneity ( $< 1$  mm) is present. A quantitative reduction of the data shows that  $\mathbf{M}_{\text{eff}}$  varies in direction over a wide range, and in a range of 20 G in magnitude, in the temperature interval  $T_c - T \approx 1^\circ\text{C}$ . The temperature dependence of the induction  $B = 4\pi\mathbf{M}_{\text{eff}}$  is shown in Fig. 6 together with the results of one of the measurements of the polarization components  $P_{x,y,z}$ . We see that the anomalously large rotations of the vector  $\mathbf{P}$  and the minimum of  $|\mathbf{P}|$  correspond to an abrupt increase of the spontaneous magnetization (if it is assumed that  $M_S \sim M_{\text{eff}}$ ), and this spontaneous magnetization appears at a higher temperature. The presence of a magnetization "tail" for the case when the sample is

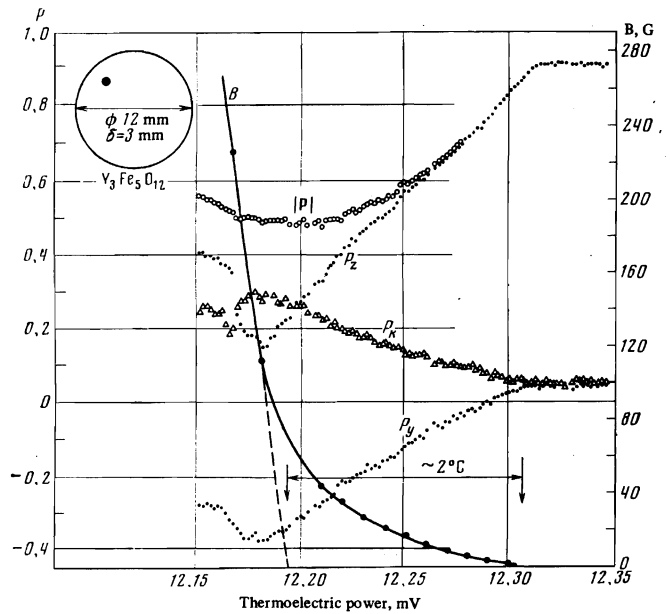


FIG. 6. The induction  $B$ , calculated from the polarization components  $P_{x,y,z}$ , as a function of the temperature measured with a copper-constantan thermocouple. The point where the measurement was made on a disk-shaped sample is marked by the dot in the circle.

placed in a magnetic field is not strange, since the critical fluctuations of the magnetization are oriented by the applied field. The magnetization observed by us cannot be induced by the residual field in the screen ( $\sim 10^{-3}$  Oe). The causes of this behavior of  $M_S(T)$  are not yet clear. They can be connected both with imperfections in the crystal and with the influence of the dipole forces. At a temperature close to  $T_c$ , weak but longer-range dipole forces can prevail over the exchange forces.<sup>[7]</sup> In this case, at a large distance from  $T_c$ , where the dipole interaction can be neglected, agreement with the similarity laws will be observed, but the temperature obtained by extrapolating these laws to  $M_S = 0$  (to determine  $T_c$ ) may not coincide with the temperature at which the spontaneous magnetization, which is determined by the dipole-dipole interaction, sets in. At the present time it is not clear in the theory how  $M_S(T)$  should behave when account is taken of the dipole forces, but it is clear that it can differ from the behavior determined by the pure exchange interaction. In any case, the  $M_S(T)$  dependence observed by us should lead to an increase of the critical exponent  $\beta$  on approaching  $T_c$ , up to  $\beta > 1$ , if  $T_c$  is taken to be the temperature of the start of the rotation of the polarization vector. It becomes understandable therefore why different exponents  $\beta$  are obtained in different experiments, as already noted above.

In addition to studying the macroscopic behavior of the magnetization, the vector-analysis procedure can be used to investigate neutron polarization in critical scattering in magnets, magnon scattering, helicoidal structures, etc. A joint analysis of the intensity and polarization of the scattering yields more complete information on the scatterer, sometimes information not obtainable with ordinary crystal-diffraction spectrometers. Thus, in the case of inelastic scattering, using the connection  $\mathbf{P} = -\mathbf{e}(\mathbf{e} \cdot \mathbf{P}_0)$  of the polarization with the scattering vector, spectrometry becomes possible at energy transfers  $\sim 10^{-5}$  eV.<sup>[8,9]</sup>

In conclusion, the authors are deeply grateful to G. M.

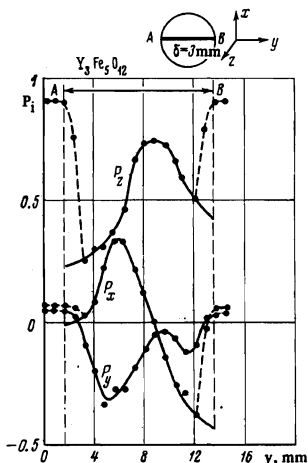


FIG. 5. Polarization of the transmitted beam over the section AB of the sample near the Curie point. The dashed curves show the sections where the edge effects appear.

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