

proportional to  $\langle s^2 \rangle$ —the mean squared atom displacement due to the presence of defects in the crystal. The presence of the residual factor (7) should lead to the appearance of resonant dips against the background of intensity maxima. The absorption of a paired state by a defect can be called a local Mössbauer effect.

No account was taken in the calculated spectrum of Fig. 5a of the presence of defects in the crystals. It is seen that the experimental points in that figure fit the theoretical line well. The absence, within the measurement accuracy limits, of residual resonant absorption due to disturbances of the crystal lattice is evidence of the high perfection of the employed crystal. The solid line in Fig. 5b was calculated for  $e^{-L} = 0.9$ . The static Debye-Waller factor should have this value in order to obtain the picture produced with the aid of the standard.

## CONCLUSION

Thus, the iron borate crystals provide the most favorable conditions for the investigation of the suppression of a nuclear reaction in pure nuclear diffraction of Mössbauer  $\gamma$  quanta. In these crystals, if the conditions of the total SE are maintained, it is possible to vary widely the polarization structure of the superposition state of the  $\gamma$  quantum in the crystal. In addition, artificial controllable violation of the SE by varying the magnetization of the crystal is easily realized.

The sensitivity of the Laue-diffraction spectra to res-

onant residual absorption can be used to study the static and dynamic violations of the periodicity of the crystal lattice.

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# Magnetic anisotropy of PdFe alloy near the Curie point $T_c$

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Magnetoelastic effects that occur under the action of a uniaxial load and data on the depolarization and intensity of a neutron beam passing through the specimen are used to determine the range of existence of a quasidomain structure ( $\tau = (T_c - T)/T_c \approx 10^{-2}$ ). It is shown that the texture parameter is related to the magnetic-anisotropy and magnetostriction constants. From its temperature dependence, the dependence of the anisotropy constant on  $\tau$  follows in the form of the power series. Possible reasons for such a dependence are discussed.

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## 1. INTRODUCTION

By analysis of data on depolarization of neutrons in nickel,<sup>1</sup> Maleev and one of the authors<sup>2</sup> arrived at the conclusion that the observed large depolarization at a phase transition near the Curie point  $T_c$  could not be explained by critical fluctuations alone, since in the immediate vicinity of  $T_c$  there is a scattering of neutrons with spin reversal that is excessive in comparison with the critical. It is contained within a narrow cone of angles, determined by the aperture of the detector, and leads to large depolarization of the trans-

mitted beam. This excessive small-angle scattering, apparently caused by unusual quasidomains, means that the transition to the ferromagnetic state is actually a transition of the first kind that is nearly of the second.

It is known that the chief role in the formation of domains is played by competition between the anisotropy, exchange, and demagnetization energies. In cubic ferromagnets, with lowering of the temperature near  $T_c$  the anisotropy energy ( $E_K \sim M_s^4$ ) increases faster than the magnetostatic ( $E_{ms} \sim M_s^2$ ), and therefore one must actually

expect, within a small temperature interval near  $T_c$ , occurrence of a quasidomain structure similar to ordinary domains. It is obvious that for large magnetization of the material (large magnetic moment of the atom), this interval must be very close to the region of critical phenomena, and therefore difficult to observe in its pure form.

By study of the depolarization of neutrons in PdFe alloys,<sup>3</sup> whose magnetization depends on the concentration of iron atoms, it was possible to observe both the region of critical fluctuations and the region of existence of domains. The domain region was identified in these experiments by a number of indicators, such as a peculiar hysteresis of the depolarization dependent on the magnetization, a large change of the depolarization from zero to a maximum value within a narrow temperature interval, a peak of the small-angle scattering that was more intense than the high-temperature peak of the critical scattering, and, finally, manifestation of magneto-elastic effects: a dependence of the depolarization on the direction of application of a uniaxial load,<sup>4</sup> which, because of the occurrence of magnetostriction, acts in an orientating fashion on the macroscopic magnetization of the quasidomains.

The intermediate region, domain formation, is of interest. The present paper is devoted to investigation of it in PdFe alloys suitable for this purpose. Here we deal, in particular, with the nature of the magnetization distribution, and also the temperature dependence of the magnetic-anisotropy energy near the Curie temperature. Taking into account that the linear magnetostriction and the magnetocrystalline anisotropy play an important role in the formation of a domain structure, we made use of magnetoelastic effects in this investigation.

## 2. METHOD OF OBTAINING AND ANALYZING DATA

For elastic scattering of neutrons on isotropically distributed inhomogeneities, a characteristic kinematic effect is observed.<sup>2</sup> It consists in the fact that neutrons initially polarized along the velocity are depolarized more weakly, by a factor 1.5, than neutrons polarized transversely to it; that is, the following experimentally confirmed relation holds<sup>4,5</sup>:

$$\alpha = \Delta P_{\perp} / \Delta P_{\parallel} = 3/2, \quad (1)$$

where  $\Delta P$  is the depolarization; the symbol after  $\Delta P$  denotes the orientation of the polarization vector  $P_0$  with respect to the neutron velocity  $V$ .

If the distribution of the inhomogeneities in a ferromagnet of the type with domains near  $T_c$  is characterized by uniaxial anisotropy of some type (for example, in Ref. 4 the anisotropy was produced by external pressure<sup>1</sup>), then relation (1) breaks down. As was shown in Ref. 6, the depolarization in such a ferromagnet depends on the mutual orientation of three vectors:  $P_0$ ,  $V$ , and the anisotropy axis  $n$ . The total cross section of scattering of neutrons on inhomogeneities also depends on the mutual orientation of  $V$  and  $n$ .

The depolarization and the cross section are ex-

pressed analytically in terms of quantities describing the domain structure and the kinematics of the scattering, and they depend on the angle  $\psi$  between the direction of the beam and the axis  $n$  (see formulas (24), (27), and (30) of Ref. 6). Measurement of the depolarization of all three components of the vector  $P$  at present poses no problem, but in measurements of the total magnetic-scattering cross section we encounter definite difficulties. In experiments on transmission of polarized neutrons (the usual polarization analysis), one could determine it from the weakening of the transmitted beam. But this requires detectors with a very small aperture, which are actually not used because of the low intensity of the beams and the limited dimensions of the specimens. Correct calculation of the background of nonmagnetic scattering and of the absorption of neutrons in the specimen is an obstacle of fundamental character, which already greatly complicates the situation. As a result, from the experimentally measured quantities there remains only the depolarization, formulas for which simplify in the limiting cases  $\psi = 0$  and  $\psi = 90^\circ$ , easily realized experimentally.

For large depolarization ( $\Delta P \sim P_0$ ), usually observed in a thick specimen, and under the condition that  $P_{0x} \parallel V$  and  $P_{0y}$ ,  $P_{0z} \perp V$ , we have:

a) The case  $n \parallel V$ :

$$\begin{aligned} \ln(P_x/P_{0x}) &\equiv f = -A_{\parallel}(1-x_{\parallel})/3, \\ \ln(P_y/P_{0y}) &\equiv g = -A_{\parallel}(1+x_{\parallel})/2, \\ \ln(P_z/P_{0z}) &\equiv h = -A_{\parallel}(1+x_{\parallel})/2, \end{aligned} \quad (2)$$

b) The case  $n \perp V$ :

$$\begin{aligned} \ln(P_x/P_{0x}) &\equiv f' = -A_{\perp}(1/3 + 1/6 x_{\perp} - 1/2 C_1), \\ \ln(P_y/P_{0y}) &\equiv g' = -1/2 A_{\perp} [1 - 3/4 x_{\perp} + 1/8 (C_0 - C_1) - 1/4 d_1], \\ \ln(P_z/P_{0z}) &\equiv h' = -1/2 A_{\perp} [1 - 1/4 x_{\perp} - 1/8 (C_0 + 2C_1) + 1/4 d_1]. \end{aligned} \quad (3)$$

Here

$$A_{\parallel} = 2N_0 \sigma_{0\parallel} L, \quad A_{\perp} = 2N_0 \sigma_{0\perp} L, \quad \sigma_{0\parallel} \sim M_s^2(T) R_{\parallel}, \quad \sigma_{0\perp} \sim M_s^2(T) R_{\perp};$$

$L$  is the specimen thickness,  $\chi_{\parallel, \perp}$  is the degree of departure of the magnetization distribution from an isotropic one (the degree of uniaxiality),  $M_s(T)$  is the saturation magnetization at temperature  $T$ ,  $R_{\parallel, \perp}$  is the mean linear dimension of a domain along the neutron trajectory,  $N_0$  is the density of magnetic cells, and  $C_0$ ,  $C_1$ , and  $d_1$  are Fourier components of the expansion of the mean statistical form-factor of the domains, which are an averaged characteristic of its angular anisotropy in the plane perpendicular to the beam. The symbols  $\parallel$  and  $\perp$  after the parameters reflect their dependence on the angle  $\psi$  (the symbol  $\parallel$  corresponds to  $\psi = 0$ ,  $\perp$  to  $\psi = 90^\circ$ ). In formulas (2) and (3), the initial polarization  $P_{0i}$  and the measured  $P_i$  are supposed directed along the same axis of a coordinate system with its  $X$  axis along  $V$  ( $i = X, Y, Z$ ).

Any one of the parameters ( $\chi$ ,  $C$ ,  $R$ ) may be used to study the magnetic properties of the material, but we shall be interested only in the parameter  $\chi$ . It depends on the mean square of the cosine of the angle  $\theta_1$  between the magnetization  $M$  and the axis  $n$ ,

$$x = 1/2 (3 \overline{\cos^2 \theta_1} - 1) \quad (4)$$

and it varies over the range  $-1/2 \leq x \leq 1$ . If the vectors  $M$  are located in the plane perpendicular to  $n$ , then

$x = -\frac{1}{2}$ ; when the magnetization  $M$  is oriented along  $n$ , then  $x = 1$ . The direction of alignment of  $M$  under the action of external uniaxial loads is determined by the relation between the signs of the load and of the magnetostriction coefficient  $\lambda_s$ .

In general it is clear that  $x$  as a texture parameter must be a function of the anisotropy energy  $E_K$  and of an energy connected with the source of the texture; for example, the magnetoelastic energy  $E_{me} \approx \lambda_s \sigma$  under the action of a stress  $\sigma$ . Therefore by analysis of the depolarization (2) and (3) at various temperatures of the specimen, it is possible to obtain the temperature dependence of  $x$  and hence the temperature dependence of the ratio of these energies. When  $\psi = 0$ , the parameter  $x_{||}$  is easily determined from the system (2) through the experimentally measured values of  $f$ ,  $g$ , and  $h$ . The situation is different with  $x_{\perp}$ . As is seen from (3), the number of unknowns is larger than the number of equations relating them to the quantities  $f'$ ,  $g'$ , and  $h'$ . But if the domains are magnetized uniformly,<sup>6</sup> then

$$x_{||} = x_{\perp} = x, \quad C_1 = xC_0. \quad (5)$$

Furthermore, the coefficient  $d_1$  is defined as the fourth Fourier component of the form-factor of the domains, and therefore it probably is smaller than all the others and may be neglected. Under these conditions, data on the depolarization alone are sufficient for determination of  $x_{\perp}$ .

It should be noted that these conditions are well satisfied<sup>7</sup> only when  $T \ll T_c$ ; near  $T_c$ , equations (5) in general are not valid, and furthermore nothing is known about  $d_1$ . Nevertheless, by supposing that  $d_1 = 0$  and  $C_1 = x_1 C_0$ , one can establish from the results of the experiment whether this is actually the case. If it turns out that  $x_{||} = x_{\perp}$ , then the supposition is correct, and therefore the domains are magnetized uniformly. But if  $x_{||} \neq x_{\perp}$ , then they are probably nonuniform.

### 3. EXPERIMENT AND RESULTS

From considerations of convenience, the specimen are prepared from a PdFe alloy with iron concentration 3.9 at.%, obtained by induction melting in an inert atmosphere. It was a cube with edge 1 cm. After machining, the specimen was annealed for three days in a vacuum.

The experiment consisted of measurement of the depolarization of a neutron beam as a function of the specimen temperature for various mutual orientations of  $P_0$ ,  $V$ , and  $n$  and was done on a setup<sup>3</sup> provided with special apparatus for orientation of the vector  $P_0$  with respect to  $V$ , with subsequent analysis of the polarization after the specimen.<sup>8</sup> The specimen, in the thermostat unit,<sup>7</sup> was placed inside this apparatus, the magnetic field in which did not exceed 0.05 Oe.

The anisotropy was produced by uniaxial compression, by means of gaseous helium. Although the construction of the thermostat made it possible to produce anisotropy in two mutually perpendicular directions by means of two pairs of pistons, in the present experiment a single pair, horizontal, was used in order to

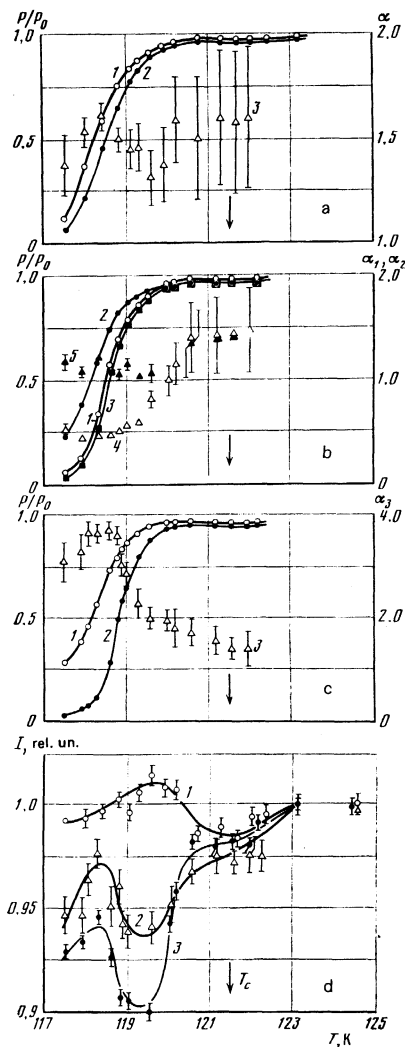


FIG. 1. Temperature dependence of the polarization and intensity of the neutron beam transmitted through the specimen; temperature dependence of the parameters  $\alpha$  and  $\alpha_i$ : a)  $\sigma = 0$ ; 1— $P_0 \parallel V$ ; 2— $P_0 \perp V$ ; 3— $\alpha$ . b)  $n \perp V$  ( $\sigma = 0.6 \text{ kg/mm}^2$ ); 1— $P_0 \parallel V$ ; 2— $P_0 \perp V$ ; 3— $P_0 \perp n \perp V$ ; 4— $\alpha_1$ ; 5— $\alpha_2$ . c)  $n \parallel V$  ( $\sigma = 0.6 \text{ kg/mm}^2$ ); 1— $P_0 \parallel V$ ; 2— $P_0 \perp V$ ; 3— $\alpha_3$ . d) 1— $\sigma = 0$ ; 2— $n \parallel V$  ( $\sigma = 0.6 \text{ kg/mm}^2$ ); 3— $n \perp V$  ( $\sigma = 0.6 \text{ kg/mm}^2$ ).

avoid possible systematic error in the loading as a result of different rigidities of the diaphragms in the different pairs. In this case, for performance of the experiment with  $\psi = 90^\circ$  the thermostat was rotated about the vertical axis  $90^\circ$  with respect to the position corresponding to  $n \parallel V$ . The necessary specimen temperature was attained by means of cooling of the gas and heaters and was stabilized with accuracy  $\sim 0.01^\circ$ .

Before going on to a discussion of the results, we shall make a few remarks of general character.

1. The parameter  $x$  was determined solely from the experimental variations of the polarization  $P$  of the transmitted beam with the specimen temperature  $T$  (Fig. 1) for  $T < T_c$ , as was discussed in Section 2:  $x_{\perp}$  from the  $P(T)$  curves of Fig. 1b,  $x_{||}$  from the  $P(T)$  curves of Fig. 1c. It is quite obvious that the values of  $x_{||}$  and  $x_{\perp}$  depend on the character of the initial ( $\sigma = 0$ ) magnetization distribution in the specimen. Trustworthy

data are obtained when it is isotropic. But if the specimen is initially textured and if the orientation of the texture axis with respect to it is unknown, then spurious results are unavoidable.

A criterion of isotropy is the equality (1); therefore we conducted a control experiment without loading (Fig. 1a), which showed that within the temperature interval investigated,  $\alpha = 1.5$ ; that is, in the free specimen no anisotropy is observed. At the same time, when  $\sigma = 0.6 \text{ kg/mm}^2$ , below 121 K, both for  $n \parallel V$  and for  $n \perp V$  this parameter deviates from 1.5 and has a temperature dependence. Since in the PdFe alloy the magnetostriction coefficient  $\lambda_s$  is negative in the transition region,<sup>4</sup> as a result of compression the magnetization  $M$  aligns itself along  $n$ , and this in turn, for appropriate orientations of  $n$  and  $V$ , determines values of  $\alpha_i$  larger or smaller than  $3/2$ . We note that for  $n \perp V$ , two parameters  $\alpha$  can be defined:  $\alpha_1 = g'/f'$  and  $\alpha_2 = h'/f'$  [see (3)].

2. The measurements always began with the high temperatures (in the paramagnetic phase). This made it possible to avoid hysteretic effects.

3. The loads did not exceed the elastic limit of pure palladium,  $3.2 \text{ kg/mm}^2$ .<sup>9</sup> We cite palladium in the absence of data on PdFe alloys.

#### A. Concerning the quasidomain structure

The large aperture of our detector ( $\sim 20'$ ) made it impossible to measure accurately the total magnetic-scattering cross section but permitted observation of the temperature behavior of the intensity of the transmitted beam, represented in Fig. 1d by curves 1, 2, and 3 for  $\sigma = 0$ ,  $n \parallel V$ , and  $n \perp V$  respectively. From these curves and from the behavior of  $\alpha_i$ , the Curie temperature of this alloy was determined,  $T_c = 121.5 \text{ K}$ ,<sup>7</sup> and the range of formation of quasidomains was identified. In fact, the increase of intensity below 121 K (curve 1 in Fig. 1d) means that the scattering becomes more small-angle; nuclei of the ferromagnetic phase appear, i.e., an unusual domain structure in which the dimensions of the domains and of the walls are of the same order of magnitude. The resulting magnetization in them is small because of the nonuniform distribution. This explains the small (up to 5%) depolarization at the temperature corresponding to the maximum of the intensity (Curve 1 in Fig. 1d), at 119.7 K (according to estimates in Ref. 7, the size of these quasidomains is comparable with the dimensions of the specimen and is  $\delta \sim 0.4 \text{ cm}$  when the mean magnetization is  $\sim 1 \text{ G}$ ).

Loading, like uniaxial anisotropy, enhances the contrast between a domain and a wall; the magnetization distribution in some quasidomains becomes nearly uniform. The depolarization at the same temperature increases to  $\sim 10\%$ , and the broad maximum (Curve 1, Fig. 1d) changes shape, since together with coarse inhomogeneities there now appear fine ones as well (probably walls), which scatter neutrons outside the aperture.

The nature of the magnetization distribution can be judged from the  $\chi(T)$  relation (Fig. 2). From  $T = T_c$  to  $T = 119 \text{ K}$  a difference between  $\chi_{\parallel}$  and  $\chi_{\perp}$  is observed;

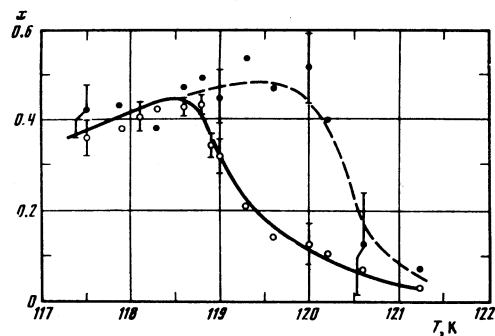


FIG. 2. Temperature dependence of the anisotropy parameter ( $\sigma = 0.6 \text{ kg/mm}^2$ ):  $\circ - \chi_{\parallel}$ ;  $\bullet - \chi_{\perp}$ .

this is a consequence of the nonuniform magnetization of domains that are under stress. But the loading apparently changes the the initial (at  $\sigma = 0$ ) distribution insignificantly, since the depolarization increases approximately by a factor 2, and therefore the change of  $|B|$  according to the formula<sup>10</sup>  $P = P_0 \exp(-g^2 B^2 \delta L / 3V^2)$  is small. This is actually possible if the external pressure is small, since the resulting magnetoelastic energy  $E_{me} \sim \lambda_s \sigma$  makes a negligible contribution to the total energy of the system.

It is seen from Fig. 2 that at lower temperatures  $\chi_{\parallel}$  and  $\chi_{\perp}$  are equal; and since  $\sigma = \text{const}$  at all temperatures, the difference between  $\chi_{\parallel}$  and  $\chi_{\perp}$  probably disappears because of a change of the magnetization distribution in the quasidomains: that is, from a nonuniform one at high temperatures it changes to a uniform one at low.

Thus for  $\tau = (T_c - T)/T_c \approx 10^{-2}$  a nonuniform quasidomain structure exists, and a temperature  $\sim 119.7 \text{ K}$  corresponds to the beginning of its formation. In the region of decrease of  $P(T)$ , coarse inhomogeneities have been observed in nickel<sup>11</sup> and in yttrium-iron garnet.<sup>12</sup>

#### B. Concerning magnetic anisotropy

During the process of lowering of temperature from  $T = 119.7 \text{ K}$ , a gradual reorganization of the quasidomain structure occurs. This is evident both from the behavior of the intensity curves (Fig. 1d) and from that of the parameter  $\chi$  (Fig. 2). This reconstruction is caused by a change of the ratio between those forms of energy that determine the nature of the domain structure; between the magnetic anisotropy energy  $E_K$  and the magnetoelastic energy  $E_{me}$ .

At present there is no rigorous microscopic theory of the quasi-domain structure, from which it would be possible to obtain a functional relation between the experimentally measurable parameters of this structure, the behavior of the magnetization and the constants  $\lambda_s$  and  $K$  ( $K$  is the dimensional anisotropy constant) near  $T_c$ . Therefore in the analysis of the results, it is necessary to use the simple qualitative picture characteristic of ordinary domains.<sup>13</sup>

Under small loads, when the anisotropy energy  $E_K$  dominates over the magnetoelastic  $E_{me}$ , and if the magnetization does not change in modulus but undergoes

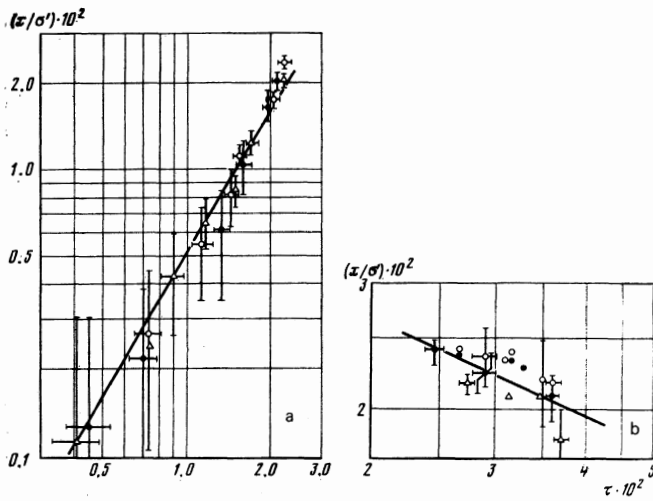


FIG. 3. Dependence of  $x/\sigma'$  on  $\tau = (T_c - T)/T_c$  ( $\sigma' = 33\sigma$ ): a) for  $\tau < 2.2 \cdot 10^{-2}$ ; b) for  $\tau > 2.2 \cdot 10^{-2}$ . The values  $\circ$  are for  $\sigma = 0.3$  kg/mm<sup>2</sup>;  $\bullet$ —for  $\sigma = 0.6$  kg/mm<sup>2</sup>;  $\triangle$ —for  $\sigma = 0.9$  kg/mm<sup>2</sup>.

only small rotations within the domains, the analysis leads to the following functional relation (see Appendix I):

$$x = \lambda_s / \sigma / K, \quad (6)$$

where  $\lambda_s$  is the coefficient of isotropic saturation magnetostriction in a cubic ferromagnet. Proportionality of the parameter  $x$  to the load is confirmed by an experiment conducted at three values of  $\sigma$ . The results are shown in Fig. 3, from which it is evident that the points for the ratio  $x/\sigma$  at all temperatures  $\tau$  are located on a single straight line within the limits of experimental error.

We shall analyze the experimental data in the light of the relation (6). In Fig. 3 it is represented by the power law

$$x/\sigma \propto \lambda_s / K \propto \tau^m. \quad (7)$$

For  $x$  two clearly expressed regions of variation are observed (Figs. 2 and 3): in the interval between  $T = 121.5$  K and  $T = 118.7$  K it increases, below  $T = 118.7$  K it falls. Therefore the exponent  $m$  has for these regions the values  $1.7 \pm 0.2$  and  $-0.6 \pm 0.2$ , respectively. Allowing for this, one can write

$$x(\tau) \propto \frac{\tau^{1.7}}{a + b\tau^{2.3}}, \quad (8)$$

where  $a$  and  $b$  are constants.

What possible temperature behavior of the constants  $\lambda_s$  and  $K$  would lead to such a result? Unfortunately there are no data on the  $\lambda_s(\tau)$  relation near  $T_c$  for PdFe. In a number of ferromagnets of cubic symmetry (for example, Ni, Fe, and their alloys) near the transition, as well as at low temperatures,  $\lambda_s$  is proportional to  $M_s^2$ .<sup>14</sup> In some amorphous magnetic materials, PtFe and PdNi alloys,  $\lambda_s$  is also proportional to  $M_s^2$ .<sup>15, 16</sup> We shall assume the same relation for PdFe also: that is,  $\lambda_s \propto M_s^2 \propto \tau^{2\beta}$ , in accordance with similarity theory ( $M_s \propto \tau^\beta$ ). Then if in (7) we go over to a dimensionless constant  $K' = K/M_s^2 = K/\tau^{2\beta}$  convenient for comparison with theory (Appendix II), we have, with allowance for

(8),

$$K' \propto \frac{\tau^{-1.7}}{A + B\tau^{-2.3}}. \quad (9)$$

Actually, this is the function,  $1/x(\tau)$ , and it corresponds to the two versions of the change of  $K$  with increase of  $\tau$ : 1) either a nonmonotonic change without change of sign with a minimum value at  $\tau \approx 2 \cdot 10^{-2}$ , 2) or a monotonic decrease with change of sign at the same  $\tau$  (Figs. 2 and 3).

It is possible that the result (9) depends on an incorrect assumption about the dependence of  $\lambda_s$  on  $M_s$ . But this seems unlikely. In fact, let  $K$  follow a monotonic variation  $K \propto M_s^4 \propto \tau^{4\beta}$ , which is characteristic of a cubic ferromagnet. According to a number of papers,<sup>17-19</sup> the index  $\beta \approx 0.43$  for an alloy close in iron concentration to that studied. Starting from our results (Fig. 4) on measurement of the magnetization in weak magnetic fields, estimates of  $\beta$  give the same value, if we take as the geometric position of the  $M_s(T)$  points the line on which the high-temperature break lies. As a result it is found that  $\lambda_s$  must change at an improbable rate near  $T_c$ : faster than  $M_s^6$ . Such a variation of the magnetostriction has not been observed in any material. At the same time, there are well-known theoretical bases for realization of the first and second versions.

As we know, fluctuations of the order parameter may exert a considerable influence on the magnetic anisotropy of a crystal near  $T_c$ . Then, depending on the val-

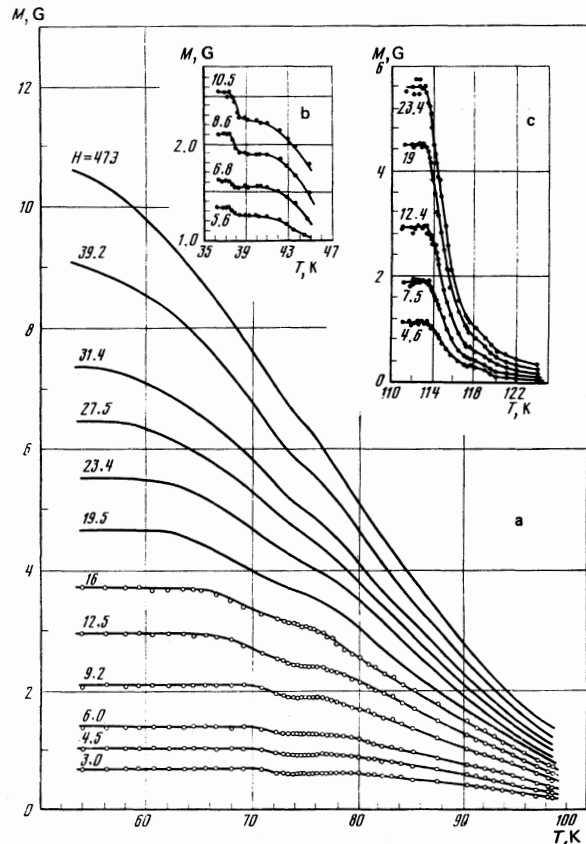


FIG. 4. The  $M(T, H)$  relation for PdFe alloys: a) with 1.8 at.% Fe; b) with 1.1 at.% Fe; c) with 3.8 at.% Fe.

ue of the energy of dipole-dipole interaction and the value of the of the constant of anisotropy of the exchange interaction  $f_1$ , various modes of behavior of the anisotropy constant  $K$  on change of temperatures are possible. If the dipole forces and the anisotropy constants  $K$  and  $f_1$  are small, then the critical fluctuations make the magnetic subsystem of the crystal effectively isotropic;<sup>20-22</sup> that is,  $K \rightarrow 0$  for  $\tau \rightarrow 0$ . But if we are in a range of temperature where the dipole-dipole interaction is already effective, then here, on the contrary, for arbitrarily small initial anisotropy  $K$  an increase of it with decrease of  $\tau$  should be observed.<sup>23, 24</sup> And finally, if our system is characterized by a considerable anisotropy of the exchange interaction  $f_1$ , then in the dipole-fluctuation region, for a definite sign of  $f_1$ , a change of sign of the anisotropy constant  $K$  is possible.<sup>25</sup>

Having this in mind, we can treat the results obtained by either of the following methods. We assume to begin with that the exchange anisotropy  $f_1$  is small or has such a sign that no change of sign of  $K$  should be observed. Then not too close to  $T_c$ , the fluctuations of the magnetization lead to a decrease of  $K$  with decrease of  $\tau$ . But as a certain temperature  $\tau^*$ , a "turning on" of the dipole-dipole forces will occur, and this will cause a transition of the system to the dipole-fluctuation mode, which expresses itself in a change of the character of the  $K(\tau)$  dependence for  $\tau < \tau^*$ : the anisotropy  $K$  begins to increase. The values of the critical index of the anisotropy in both temperature ranges are calculated in Appendix II. Thus it is possible to explain the non-monotonic behavior of  $K$  on change of temperature.

But another method of explanation is also possible. Suppose that we are in the dipole range and that the anisotropy of the exchange interaction  $f_1$  has an appreciable value and an appropriate, in the sense indicated above, sign. Then, as was shown in Ref. 25, with approach to  $T_c$  the anisotropy constant  $K$  will first decrease, then pass through zero and begin to increase, but with the opposite sign.

For both the methods of explanation proposed above, it is very important what value the characteristic energy of dipole-dipole interaction has and, accordingly, how large (or small) the dipole-fluctuation region is. Numerical estimates show that the dipole forces become important at  $T_c - T \approx 2$  K.<sup>21</sup> But it is just at these values of  $T$  that the anomalies described are observed; in particular, a change of character of the  $K(\tau)$  relation or a change of sign of  $K$ .

Obviously such a coincidence must be considered a strong argument in favor of the proposed treatment of the experimental data. At the same time, the substantial discrepancy between the theoretical (Appendix II) and experimental (9) values of the critical index of the anisotropy scarcely deserves to be considered a factor that destroys the agreement between theory and experiment, because the accuracy of determination of this index, both in the theory and in the experiment, is very low. A change of the axis of easy magnetization in PdFe can explain the unusual form of the temperature variation of the magnetization near  $T_c$ , measured by

the kink method in small magnetic fields (Fig. 4). It is known that for ordered ferromagnets it has a kink at a temperature  $T_k$  when the internal field becomes negligibly small, and therefore below this temperature there exists an unsaturated phase (see, for example, Ref. 26). The magnetization at temperature  $T_k$  corresponds to the saturation magnetization.

For PdFe, two kinks are observed. The appearance of the second, at a lower temperature, is perhaps due to vanishing of the anisotropy constant.

The question may arise: why are  $M(H, T)$  curves with characteristic singularities (Fig. 4) observed in a polycrystal? This can be explained as follows. For a quasicrystal that has appeared below  $T_c$ , it is advantageous, from the point of view of exchange energy, to capture many crystallites, transforming the specimen in the magnetic sense to a poor "single crystal" (this is indicated by the estimates of domain size given above). Then a change of the axis of magnetization of such a single crystal will also give a change of the magnetization in a field.

Experimental information on the behavior of anisotropy constants in the neighborhood of a transition is, for ferromagnets, scanty. For PdFe alloys it is absent; therefore it is difficult to carry out even a qualitative comparison. According to (9), the anisotropy in PdFe at and above  $T_c$  should be finite. In this connection, we point out recent papers<sup>27, 28</sup> in which depolarization and scattering of neutrons also suggested the existence of anisotropy above  $T_c$  in iron. In PdCo alloys, it follows from experimental data on the ferromagnetic-resonance linewidth that there is probably a minimum of the magnetic anisotropy near  $T_c$ .<sup>29</sup> These results agree qualitatively with the temperature dependence of the anisotropy that follows from analysis of our data, and therefore they are evidence in favor of the functional relation (6) and the assumed behavior  $\lambda_s \propto M_s^2$ . We note that study of the temperature dependence of the experimentally determined ratio  $\lambda_s/K$  is of independent interest, because the linear magnetostriction is intimately connected with the magnetic anisotropy, and they both play an important role in domain formation.

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## APPENDIX I

We shall consider the character of the magnetic texture in a polycrystalline ferromagnetic specimen (of PdFe type), consisting of fine, randomly oriented crystallites possessing cubic anisotropy. Texture arises under the influence of external uniform loading. For an individual crystallite, the uniform magnetization  $\mathbf{M}$  must be directed along one of the easy axes, in accordance with the requirement of minimization of the anisotropy energy, of the form<sup>13</sup>

$$E_a = -\eta(M_x^2 M_y^2 + M_x^2 M_z^2 + M_y^2 M_z^2), \quad \eta = \text{const}, \quad (\text{I.1})$$

so that without loading, on the average over the specimen there is an isotropic distribution of the magnetization vectors of the domains. Application of a load gives, because of magnetostriction, an additional contribution of magnetoelastic energy,

$$E_{me} = -\frac{1}{2} \lambda_s \sigma \cos^2 \theta_1, \quad (I.2)$$

where  $\lambda_s$  is the coefficient of isotropic saturation magnetostriction in a cubic ferromagnet ( $\lambda_{100} = \lambda_{111} = \lambda_s$ ),  $\sigma$  is the value of the stress, and  $\theta_1$  is the angle between the magnetization  $M$  and the direction  $n$  of the stress.

The stress acts like a uniaxial anisotropy and leads to the appearance of a magnetic texture: a preferred orientation of the magnetization of the domains with respect to a distinguished direction  $n$  in the specimen.

At small loads ( $\lambda_s \sigma \ll \eta M^2 = K$ ), when the cubic anisotropy energy (I.1) dominates over the magnetoelastic (I.2), the equilibrium position of the magnetization vector in the domains must be close to the easy axes, for simplicity the cube edges ( $\eta > 0$ ). If we describe the direction of the stress by spherical angles  $\theta$  and  $\Phi$  and the equilibrium position  $m$  of the magnetization by angles  $\vartheta$  and  $\varphi$  (see Fig. 5), then in this case the change of the total free energy

$$E = \frac{1}{2} \eta M^4 (m_x^4 + m_y^4 + m_z^4) - \frac{1}{2} \lambda_s \sigma (mn)^2 \quad (I.3)$$

is principally determined, for small deviations of  $m$  from the easy axes, by the change solely of the component of magnetization along the easy axis, in the form

$$\Delta E = K \left[ \frac{1}{2} \theta^4 \sin^2 2\varphi + \theta^2 (1 - \frac{1}{2} \theta^2) \right] - \frac{1}{2} \lambda_s \sigma \cos^2 \Psi \approx K \theta^2 - \frac{1}{2} \lambda_s \sigma \cos^2 (\theta - \vartheta), \quad (I.4)$$

where

$$\cos \Psi = \cos \theta \cos \vartheta + \sin \theta \sin \vartheta \cos (\Phi - \varphi) = \cos (\theta - \vartheta),$$

since the minimum of (I.4) is attained when  $\varphi = \Phi$ .

In accordance with the requirement of minimization of the free energy (I.4), the magnetization in each individual crystallite must be directed at an angle

$$\vartheta = \frac{3}{4} \frac{\lambda_s \sigma}{K} \sin 2\theta. \quad (I.5)$$

On the other hand, the parameter of anisotropy or of magnetic texture, by definition,<sup>6</sup> is [see also (4)]

$$x = \frac{1}{2} [3 \langle (mn)^2 \rangle - 1], \quad (I.6)$$

where the averaging of the projection of the domain magnetization  $m$  on the distinguished axis  $n$  extends over all the crystallites in the specimen. In view of the random orientation of the crystallites, such an averaging is equivalent to an averaging over the direction of the vector  $m$ , so that, in accordance (I.6) and (I.5), we have

$$\langle (mn)^2 \rangle = \cos^2 \theta + \frac{3}{4} \frac{\lambda_s \sigma}{K} \sin^2 2\theta. \quad (I.7)$$

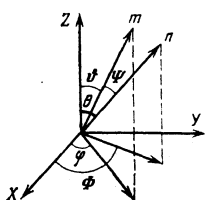


FIG. 5.

Using  $\cos^2 \theta = 1/3$  and  $\sin^2 2\theta = 8/15$ , we get from (I.6) and (I.7)

$$x = \frac{1}{2} \lambda_s \sigma / K. \quad (I.8)$$

Thus the magnetic-texture parameter in a polycrystalline cubic ferromagnet is linearly related to the stress and is determined by the ratio of the magnetostriction and anisotropy constants.

We note that for small loads, a relation of the same form holds also for a polycrystal with uniaxial crystallites, since the change of the free energy at small deviations actually coincides with formula (I.4) on appropriate change of the anisotropy constants. Equiprobable orientation of the axes of such crystallites makes the specimen, in first approximation, equivalent to a polycrystalline specimen consisting of cubic crystals, also with equiprobable orientation of axes. In fact, the free energy of a uniaxial crystal is

$$E = -K_1 \cos^2 (\Psi - \theta) - \frac{1}{2} \lambda_s \sigma \cos^2 \theta. \quad (I.9)$$

Taking into account that the loading leads to a small deviation of the magnetization from the easy axis, i.e., that the angle  $\vartheta = \Psi - \theta$  is small ( $\lambda_s \sigma / K_1 \ll 1$ ), we have

$$E = -K_1 \cos^2 \vartheta - \frac{1}{2} \lambda_s \sigma \cos^2 (\Psi - \vartheta). \quad (I.10)$$

On minimization of (I.10) with respect to  $\vartheta$  and retention only of terms linear in  $\vartheta$ , we have after averaging over  $\Psi$

$$\cos^2 \theta = \frac{2}{5} \frac{\lambda_s \sigma}{K_1} + \frac{1}{3}, \quad (I.11)$$

whence we arrive, by use of (I.6), at the result (I.8).

## APPENDIX II

The critical thermodynamics of a cubic ferromagnet, in the temperature range where the dipole forces are not important, is described by the following equations of the renormalization group<sup>22</sup>:

$$\partial g_1 / \partial t = g_1 - \nu / 11 g_1^2 - \nu / 11 g_2^2, \quad (II.1)$$

$$\partial g_2 / \partial t = g_2 - \nu / 11 g_1 g_2 - \nu / 11 g_2^2, \quad (II.2)$$

$$t = -\ln \kappa, \quad g_1 \sim \gamma_1 / \kappa, \quad g_2 \sim \gamma_2 / \kappa,$$

where  $\kappa \sim |\tau|^\nu$  is the reciprocal of the correlation radius,  $\nu = 0.7$  is a critical index, and  $\gamma_1$  and  $\gamma_2$  are the coefficients of the invariants  $\sum_i M_i^4$  and  $\sum_{i \neq j} M_i^2 M_j^2$  in the expansion of the free energy of the crystal. The difference  $\delta' = g_2 - g_1$  can serve as a measure of the anisotropy of the system. Since the initial value of  $\delta'$  is small, its variation with temperature is described sufficiently accurately by the renormalization-group equation linearized with respect to  $\delta'$ , which is easily obtained from (II.1):

$$\partial \delta' / \partial t = \delta' (1 - 12 / 11 g_1). \quad (II.3)$$

From (II.3) it is evident that near the fixed point  $g_1 = g_2 = 1$  the value of  $\delta'$  decreases with decrease of  $|\tau|$  according to the law

$$\delta' \propto \kappa^{1/11} \propto |\tau|^{0.06}.$$

But if the system has not yet passed into the asymptotic region and  $g_1 \approx g_2 > 1$ , then the rate of decrease of  $\delta'$  may be faster.

The equations of the renormalization group for a cubic crystal in the dipole fluctuation range have the form<sup>24</sup>

$$\begin{aligned} \partial g_1 / \partial t &= g_1^{-12} / {}_{17}g_1^2 - {}_{17}g_1^2 - {}_{17}g_1 g_2^{-3} / {}_{17}g_2^2, \\ \partial g_2 / \partial t &= g_2^{-3} / {}_{34}g_1^2 - {}_{34}g_1^2 - {}_{17}g_1 g_2^{-13} / {}_{34}g_2^2. \end{aligned} \quad (\text{II.4})$$

In this case the linearized equation for  $\delta'$  can be written in the following form:

$$\partial \delta' / \partial t = \delta' (1 - {}_{17}g_1). \quad (\text{II.5})$$

As is evident from (II.5), near a "Heisenberg" fixed point the anisotropy increases according to the law

$$\delta' \propto \tau^{-2/17} \propto |\tau|^{-0.12},$$

which, however, may change on departure from this point.

<sup>1</sup>In this case it would be more accurate to interpret uniaxial anisotropy as magnetic texture; that is, as the preferred orientation of the magnetic phases.

<sup>2</sup>The value of the dipole energy  $\omega_{\text{dip}} = 4\pi(g\mu)^2/R^3$  for PdFe (4 at.% Fe), with  $R = 7 \text{ \AA}$ ,  $\mu = 5\mu_B$ ,  $g = 2$ , is  $\omega_{\text{dip}} \approx 2^\circ$ .

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