

Magnetic ordering in dilute PdFe alloys

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Polarized neutrons are used to investigate dilute PdFe solutions with Fe concentrations from 0.5 to 8 at.%. The temperature dependence of the polarization of the neutrons passing through the samples and of the small-angle scattering of the polarized neutrons has three singularities connected with magnetic-ordering processes, viz., domain formation, critical fluctuations, and polarization of the palladium matrix. The dependence of T_c on the concentration of the Fe atoms follows an exponential law with an exponent $C^{-1/3}$. The parameters of this dependence, namely, the radius and the interaction potential of the impurity atoms, are determined. An estimate is presented of the correlation radius of the spins near T_c for $C=4$ at.% Fe. A tendency of the singularities of the depolarization to coalesce is noted when the average magnetization of the alloys is increased.

INTRODUCTION

The behavior of ferromagnets near the Curie temperature has been studied intensively of late (see, e.g., [1,2]). In magnetic systems undergoing a phase transition, the appearance of spontaneous magnetization and domain formation frequently occur in rather narrow temperature intervals. As a result, the existing measurement methods usually yield integral characteristics, where the two processes are not specifically separated. Therefore investigations of ferromagnets of the isomorphous type with variable magnetization are of great importance from the point of view of the physics of phase transitions. In this study, the objects of investigation were dilute PdFe alloys.

It is known that owing to the large paramagnetic susceptibility of palladium [3], small additions of a magnetic impurity (up to 0.05 at.% iron or cobalt) make it ferromagnetic. The average magnetic moment per atom of the alloy depends on the concentration of the magnetic impurity [4,5]. By thus varying the average magnetic energy of the sample [6] and by the same token influence the domain-formation process, accelerating or retarding its onset relative to the appearance of the spontaneous magnetization. This offers a possibility of separating the region of critical fluctuations of the magnetization.

THE EXPERIMENT

We investigated PdFe samples with nominal iron concentrations of 0.5, 1 (1.1), 2 (1.6), 4 (3.8), 6 (5.6), and 8 at.% (the figures in the parentheses are the chemical-analysis data). A beam of polarized neutrons with $\lambda = 2 \text{ \AA}$ was obtained with a setup (Fig. 1a) in which the polarizer and analyzer were $\text{Co}_{0.92}\text{Fe}_{0.08}$ single crystals placed in permanent magnets. Measurements with $\lambda = 4 \text{ \AA}$ were performed with the setup described by Okorokov [7]. The polarizations P_0 of the neutron beams incident on the sample were 0.91 and 0.86 for $\lambda = 2 \text{ \AA}$ and $\lambda = 4 \text{ \AA}$, respectively. Special shields have made it possible to reduce the field at the location of the sample practically to zero ($\sim 0.01 \text{ Oe}$) [8]. The samples were placed in a cryostat unit so constructed that the sample experienced no mechanical load but made thermal contact with the heat exchanger [8] (Figs. 1b and 1c). The measurements in magnetic fields were performed in a magnet that also served simultaneously as the heat ex-

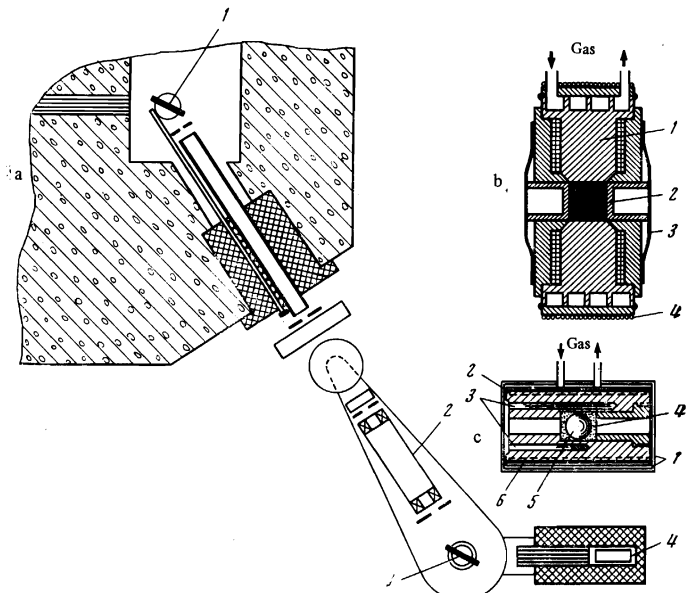


FIG. 1. Diagram of setup ($\lambda = 2 \text{ \AA}$). a) Setup: 1—polarizing crystal, 2—neutron spin-flip device, 3—analyzing crystal, 4—detector; b) unit used for sample in the magnetic measurements: 1—magnet (heat exchanger), 2—sample, 3—spring, 4—heater; c) sample unit for measurement in a field $H = 0$: 1—magnetic shields, 2—thermometers, 4—ampule with He, 5—sample, 6—heater.

changer. The temperature was varied or maintained with the aid of an electronic stabilization circuit with alloyed-germanium resistance thermometers. The temperature was monitored with a platinum resistance thermometer. The temperature was maintained with accuracy better than 0.01°K . The coolants were liquid helium, neon, and nitrogen.

All the polycrystalline samples used for the measurements in zero field were spheres with 10 mm diameter. The magnetic measurements were performed on a cube measuring $10 \times 10 \times 10 \text{ mm}$ with iron concentration 1 at.%. The blanks for the samples were prepared by repeated induction melting in vacuum. After preparation, the samples were annealed in vacuum at 1100°C for several days. The palladium was 99.99% pure (spectral analysis revealed no magnetic impurities), and the iron purity was 99.999%.

We measured, in a wide temperature range, the polarization of neutrons passing through the samples at

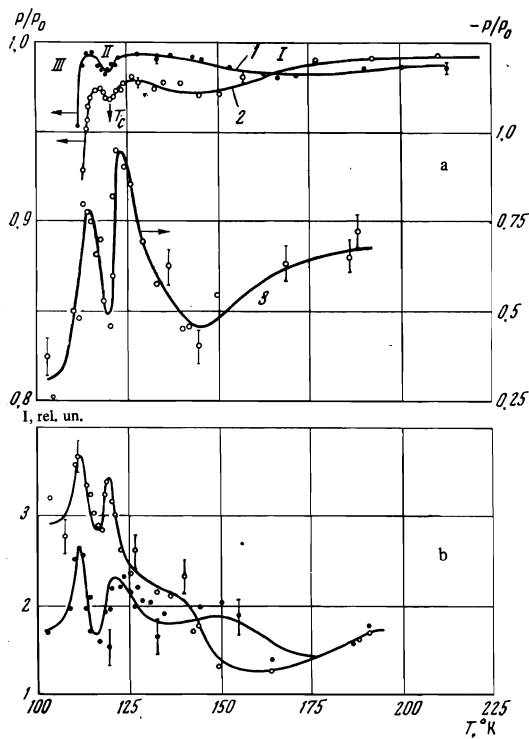


FIG. 2. a) $P(T)$ plot of neutrons with $\lambda = 2 \text{ \AA}$ (curve 1) and $\lambda = 4 \text{ \AA}$ (curve 2) passing through the sample and of the neutrons scattered at $\theta = 10'$ (curve 3); b) scattering intensity: $\circ - \theta = 10'$, $\bullet - \theta = 30'$.

all concentrations. In addition, we measured the temperature dependence of the small-angle scattering (scattering angles $10'$ and $30'$) of neutrons with $\lambda = 4 \text{ \AA}$ by a sample with concentration 4 at.% Fe.

RESULTS AND DISCUSSION

The measured polarizations of the neutrons passing through PdFe with 4 at.% Fe are shown in Fig. 2a. The curves for the remaining concentrations are similar (see Fig. 5).

The temperature dependence of the polarization exhibits three features; a broad minimum I at high temperatures, a narrow minimum II at lower temperatures, and finally a steep descent III. It is important to note here that the positions of the gently-sloping minimum I and of the steep descent III depend, at a given concentration, on the neutron wavelength λ , whereas the temperature position of II does not depend on λ . A temperature hysteresis of the steep polarization descent III is observed (Fig. 3a) all the way to the minimum II. With decreasing temperature, starting with the minimum II, rotation of the polarization vector sets in, owing to the onset of spontaneous magnetization.

All three characteristic regions are also manifest in the picture of the small-angle scattering of a beam of polarized neutrons. Regions I, II, and III correspond to maxima of the scattering intensity, and also to peculiarities in the behavior of the polarization of the scattered beam (Figs. 2a and 2b). When II is approached from the high-temperature side, the polarization of the scattered beam reverses sign, reaching a value $P \approx -P_0$, while the scattering intensity increases abruptly. The scattering in region II is mainly through small angles, for the peak II becomes almost completely smeared out even at an angle $30'$. According to Maleev^[9] this means that the scattering is quasielastic (the relation

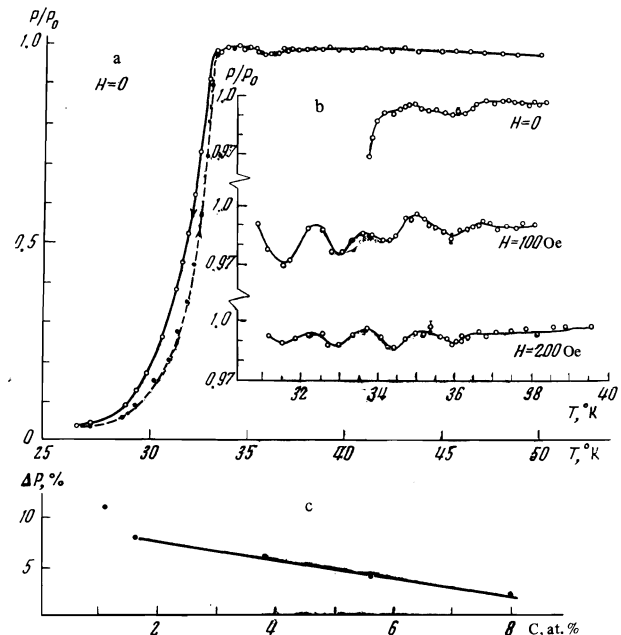


FIG. 3. Depolarization hysteresis: a) $P(T)$ plot for $H = 0$ ($C = 1 \text{ at.}\%$ Fe, $\lambda = 2 \text{ \AA}$); b) the same, for fields 100 and 200 Oe; c) dependence of hysteresis on C .

$(E_0 - E)^2/4E_0^2\theta^2 \ll 1$ is satisfied, where E_0 and E are the energies of the incident and scattered neutron, and θ is the scattering angle), and the scale of the scattering inhomogeneities $d \approx \lambda/\theta$ is large. The scattering in region III is through angles much larger than in II. Here, just as in region I, appreciable inelasticity can be noted (Fig. 2a).

It can be concluded from the experimental data that II is the region of critical fluctuations of the magnetization, and that the temperature corresponding to the minimum II is T_C of the given alloy. Region III, on the other hand, is due to a first-order transition with formation of domain-type regions. It appears that there are no domains here in the usual sense. These are more readily a kind of quasidomain (embryos of domains) that results from the increasing influence of the dipole-dipole interactions. It is precisely here that the depolarization hysteresis starts to be observed, and its value decreases with increasing average magnetization of the alloy and practically vanishes at $C = 8 \text{ at.}\%$ Fe (Fig. 3c). We chose as a measure of the hysteresis the polarization difference $\Delta P = P' - P''$, where P' is the polarization at the 0.5 level on going from high temperatures and P'' is the polarization on going from low temperatures to the temperature point corresponding to P' .

This behavior can be qualitatively attributed to the formation of quasidomains or clusters of magnetization. With decreasing magnetization of the alloy, other conditions being equal, the dimension of such clusters increases^[10], and consequently their inertia with respect to magnetization reversal increases.

Neutrons with polarization vectors parallel and perpendicular to their velocity are depolarized differently in region III. The measure of the difference is the factor $\frac{3}{2}$,^[8] which is evidence of formation of regions with inhomogeneous magnetization distribution^[11].

In fields $\sim 100\text{--}200 \text{ Oe}$ (Fig. 3b), the polarization of region III remains practically unchanged, and the ob-

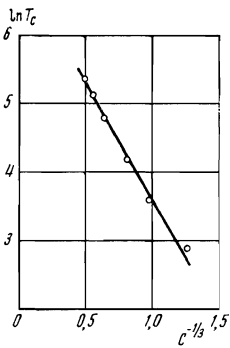


FIG. 4. Dependence of T_C on C .

served oscillations are due to the rotation of the neutron polarization vector as a result of its noncollinearity with the magnetization of the sample in the external field. However, the phase transition (region II) does not become smeared out. The presence of transverse magnetization fluctuations is manifest in T_C by weak depolarization. When the magnetic transitions is so represented, the magnetization in the region between II and III seems to be uniform.

There is still no unambiguous explanation of the minimum I. First, the dependence of its position on the neutron wavelength and the shift of the corresponding scattering maximum (Fig. 2) as a function of the angle indicate that a change takes place in the dimension of the scattering regions when the temperature changes. An impression is gained that whereas for the faster neutrons ($\lambda = 2 \text{ \AA}$) there are no longer any inhomogeneities that lead to depolarization, the slow neutrons ($\lambda = 4 \text{ \AA}$) still feel their presence. This is possible if the clouds of the polarization of Pd around the Fe atoms increase gradually with decreasing temperature until they overlap (homogeneous polarization). Then the apparent instant when the polarization clouds overlap will be different for neutrons with different wavelengths. This process causes an increase in the number of interacting impurities, since the interaction is via the d-holes of Pd. Second, one cannot exclude the existence of peculiar spin excitations (paramagnons) in this region^[12]; these lead to slow depolarization. The inelasticity revealed by the polarization of the scattered beam does not contradict this assumption.

We note also that in the corresponding temperature ranges anomalies were observed in the resistivity of the PdFe alloys ($C = 0.5$ and 1 at.% Fe)^[13]. It is difficult to find an unambiguous answer without a detailed angular and energy analysis of the scattering.

Assuming that the interaction between the impurities is described by a potential $V(r) = V_0 e^{-r/R}$, the following relation between T_C and the impurity concentration was obtained in^[14]:

$$\ln T_C = \ln V_0 - 1/aC^b.$$

Our data agree satisfactorily with this relation (Fig. 4) if $a \approx R = 3.6 \text{ \AA}$ and $V_0 \approx ZV_0 = 1000^\circ \text{K}$, where R is the average interaction radius of the impurities and Z is the average number of impurities whose energy of interaction with the given impurity is of the order of T_C . According to the data of Low and Holden^[15], $R = 3 \text{ \AA}$, and a value $R = 5 \text{ \AA}$ is mentioned in^[16]. Korenblit and Shender^[14] cite a value $a = 4 \text{ \AA}$.

Let us estimate the spin-correlation radius from the depolarization of the transmitted beam. The depolariza-

tion due to critical fluctuations near T_C is small and amounts to 1–2% for concentrations lower than 6 at.% Fe. This depolarization can be connected^[11] with the total scattering cross sections σ_1 and σ_2 inside and outside of the angle spanned by the detector, respectively,

$$P = \frac{P_0 [1 - (\sigma_1 + \sigma_2) N_0 L] + \bar{P} \sigma_1 N_0 L}{1 - N_0 L \sigma_2}; \quad (1)$$

$$\sigma_1 = \frac{2}{3} \pi (r_0 \gamma)^2 S(S+1) (pr)^{-2} \ln \left[1 + \left(\frac{pa' \varphi_m}{r^2} \right)^2 \right], \quad (2)$$

$$\sigma_2 \approx \frac{4}{3} \pi (r_0 \gamma)^2 S(S+1) (pr)^{-2} \ln \frac{1}{\varphi_m}, \quad (3)$$

where P and P_0 are the polarizations of the transmitted and incident beams, \bar{P} is the average polarization of the neutrons scattered within the limits of the transmitted beam, φ_m is the angle spanned by the detector, r and a' are the parameters of the Ornstein-Zernike theory, and L is the sample thickness.

For the case of pure elastic scattering of neutrons with a polarization vector perpendicular to their velocity we obtain from formula (1)

$$\delta P_{\perp} / P_{0\perp} = \sigma_1 / \sigma_2 N_0 L. \quad (4)$$

For $\tau = (T - T_C) / T_C \approx 5 \cdot 10^{-3}$ we obtain from curve 2 (Fig. 2a) $\sigma_1 \approx 0.06 \text{ b}$. The cross section σ_2 , on the other hand, can be determined from the attenuation of the beam when the sample goes from the paramagnetic to the ferromagnetic state

$$\sigma_2 = \frac{\ln I_f / I_p}{N_0 L}. \quad (5)$$

The cross section was found to be $\sigma_2 \approx 2.3 \text{ b}$. We can then use (2) and (3) to determine a' / τ^{ν} . For the spin S we chose an average value based on the average magnetic moment $0.36 \mu_B$ per atom of the alloy^[17]. At $\varphi_m \approx 10^{-3}$ and at a Lande factor $g = 2$ we have $R_C = a' / \tau^{\nu} \approx 400\text{--}500 \text{ \AA}$. If it is assumed that the expression for $R_C = a' \tau^{-2/3}$ which follows from similarity theory is valid far from the transition ($T > T_C$), then the correlation length at $\tau \approx 1.5$ (room temperature) is $\sim 8\text{--}10 \text{ \AA}$.

The equality of the correlation length to the dimension of the matrix polarization near the impurity atom^[15] and to the distance between the Fe atoms ($\sim 8 \text{ \AA}$) when the latter are uniformly distributed in the alloy with $C = 4$ at.% Fe indicates that the palladium is practically completely polarized.

Let us examine the behavior of the singularities of the depolarization with changing impurity concentration (Fig. 5a). If the polarization of the samples with all concentrations is plotted as a function of τ , then all the singularities exhibit a tendency to merge into one with increasing average magnetization of the alloy. This raises a difficulty in the choice of T_C for alloys with large Fe concentrations, particularly for $C = 8$ at.% Fe. It is seen from Fig. 5c, however, that at $C = 8$ at.% Fe the temperatures T_C and T_m coincide (T_m is the temperature corresponding to the minimum of the region I). The error in the determination of T_m increases at low concentrations, owing to the increase in the width of the minimum (Fig. 5b).

Thus see that at large magnetizations a difficulty arises in the separation of the critical regions, and by the same token in the exact determination of T_C . This probably explains the discrepancy between the values of the critical depolarizations in^[1,11].

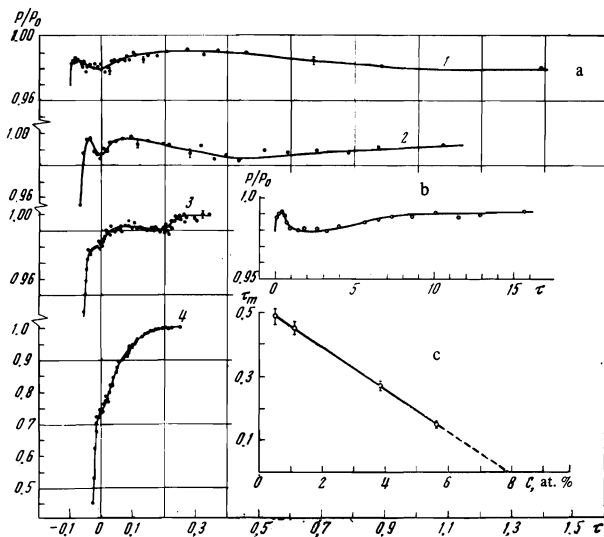


FIG. 5. a) Dependence of P on $(\tau = (T - T_c)/T_c)$: 1— $C = 0.5\%$, $T_c = 18^\circ\text{K}$, $T_m = 35^\circ\text{K}$; 2— $C = 4\%$, $T_c = 119.5^\circ\text{K}$, $T_m = 165^\circ\text{K}$; 3— $C = 6\%$, $T_c = 165^\circ\text{K}$, $T_m = 195^\circ\text{K}$; 4— $C = 8\%$, $T_c = 214^\circ\text{K}$, $T_m = 214^\circ\text{K}$. b) Dependence of P on τ for $C = 0.5$ at.% Fe. c) Shift of the region I; $\tau_m = (T_m - T_c)/T_m$.

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¹⁾We say "practically to zero" only because measurements performed by us in a field of approximately 0.3 Oe resulted in practically no noticeable change in the polarization curves.

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