

Molecular-field distribution and local magnetization in a dilute alloy of palladium cobalt and iron

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Magnetic ordering in a PdCo alloy with Co density 0.3 at.% is investigated using the Mössbauer effect in ^{57}Fe in the temperature range from 5 to 20 K. The distribution functions of the local magnetization and of the molecular field, $P(S_z)$ and $P(H_{\text{mol}})$ are obtained, as well as the temperature dependences of the average and local magnetizations.

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1. INTRODUCTION

Dilute alloys of palladium with iron and cobalt have been attracting much interest during the last two decades.^{1,2} A characteristic feature of these alloys is the formation of a swarm of polarized conduction electrons, called the giant moment [(10-12) μ_B], around the impurity 3-d atom. The overlap of these swarms causes to interactions between these impurities not to be oscillatory as, e.g., in AuFe,³ but of constant sign; this leads to a ferromagnetic ordering at very low impurity densities (less than 0.1 at.%). The properties of these alloys are determined to a considerable degree by the random arrangement of the impurities, as a result of which the behavior of the magnetic atoms depends on their local environment. Microscopic methods of investigating these alloys, such as the Mössbauer effect and NMR, become particularly important.

We have investigated in the present study, using the Mössbauer effect on ^{57}Fe , the process of magnetic ordering in a PdCo alloy with 0.3 at.% Co. The ^{57}Fe nucleus is the result of the decay of the parent nucleus ^{57}Co . The vanishingly small amount of radioactive cobalt in our alloy, as well as the similar behavior of iron and cobalt in alloys with palladium,¹ allow us to regard the iron atoms as nondistorting magnetic probes. The proportionality of the hyperfine field at the iron nuclei to the spin projection S_z (Ref. 4) makes it possible to obtain the distribution function $P(S_z)$ of the local magnetization, the distribution function $P(H_{\text{mol}})$ of the molecular fields, as well as the temperature dependences of the average and local magnetizations.

2. EXPERIMENTAL PROCEDURE

A PdCo alloy with 0.3 at.% Co was prepared from palladium of 4N purity and chemically pure cobalt. The ingot was rolled into a foil 20 μm thick, on which a drop of radioactive cobalt-chloride solution was spread. The diffusion was carried out in a hydrogen stream at 800 °C for 24 h. The sample was next dehydrogenated at 700 °C in a vacuum of 10^{-4} Torr for 3 h and finally homogenized at $T = 1050$ °C in an atmosphere of argon for 24 h. The activity of the obtained source was ~ 1 mCi. The fact that the γ -quantum counts registered from both sides of the foil were equal was evidence of the uniform distribution of the ^{57}Co over the sample thickness. The estimated density of the impurities introduced into the

sample from the radioactive solution does not exceed 0.01 at.%, so that the active and inactive samples had practically the same magnetic-impurity content.

The Mössbauer measurements were made in the temperature range from 5 to 20 K. The temperature was measured with a germanium semiconductor thermometer. The γ radiation was recorded with a miniature resonant counter based on FeAl (Ref. 5), mounted on the Mössbauer vibrator.

3. RESULTS AND THEIR DISCUSSION

1. Distribution of hyperfine fields

The Mössbauer spectra of the PdCo alloy with 0.3 at.% Co are shown in Fig. 1. The shapes and temperature dependences of the spectra are similar to those published for the PdFe alloys.⁶ In the paramagnetic

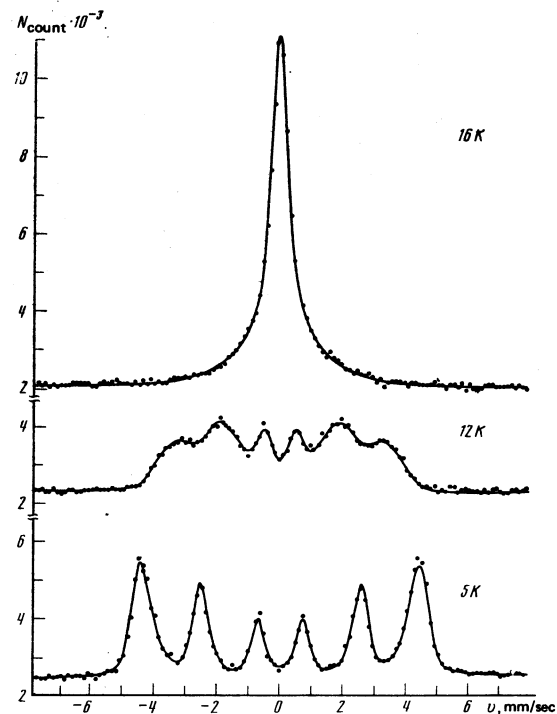


FIG. 1. Mössbauer spectra of $\text{Pd}_{0.97}\text{Co}_{0.3}$ (^{57}Fe) at various temperatures. Solid line—result of computer reduction of the spectra.

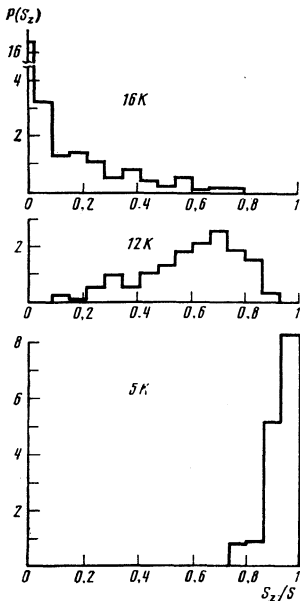


FIG. 2. Distribution of local magnetization $P(S_z)$.

region, the width of the observed single line, at $T = 20$ K, is $\Gamma = 9.49$ mm/s. With decreasing temperature, smeared spectra appear and indicate the presence of some distribution of the hyperfine fields at the iron nuclei. At 5 K the hyperfine fields for most nuclei are close to the saturation field H_{sat} , whose strength, measured by us for samples with 0.1–0.3 at. % at temperatures ~ 40 mK, is 292 ± 5 kOe.

The spectra were reduced with a computer by least squares. The spectrum was represented in the form

$$y(\nu) = N_{\infty} + \int_0^{H_{\text{sat}}} P(H) L_6(H, \nu) dH, \quad (1)$$

where $y(\nu)$ is the count in the channel, N_{∞} is the background, $P(H)$ is the distribution function of the hyperfine field, and $L_6(h, \nu)$ is the experimental Mössbauer spectrum, consisting of six lines and corresponding to one value of the hyperfine field H . The experimental-line shape was best described by a sum of three Lorentz lines, therefore $L_6(H, \nu)$ in (1) is a Lorentz operator.

The integral (1) can be represented as a sum of integrals over small intervals ΔH_i , in each of which $P(H)$ is assumed constant. Then

$$y(\nu) = N_{\infty} + \sum_i P(H_i) \int_{H_i}^{H_i + \Delta H_i} L_6(H, \nu) dH. \quad (2)$$

The integral (2) can be calculated exactly and has a simple mathematical form. A good description of the spectra was attained when the number of intervals was 16. We varied $P(H_i)$, N_{∞} , and the isomeric shift. The obtained $P(H)$ distribution curves are shown in Fig. 2. As indicated above, these curves constitute the distribution function $P(S_z)$ of the local magnetization. From this function we can obtain such important characteristics of the alloys in question as the average and local magnetizations as well as the distribution of the molecular fields.

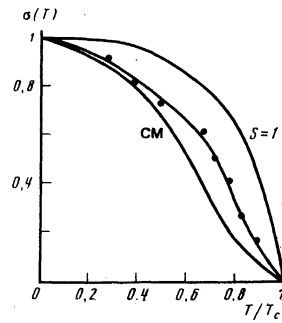


FIG. 3. Average magnetization $\sigma(T)$; $T_c = 18$ K. Upper curve—magnetization calculated in molecular-field approximation for a spin $S = 1$; lower curve—magnetization obtained from the cluster model (CM) in Ref. 2 for an impurity concentration 0.025 at. %.

2. The average magnetization $\sigma(T)$

In the form presented, $\sigma(T)$ is an integral of the distribution function of the spin projection:

$$\sigma(T) = \frac{1}{S} \int_0^1 P(S_z, T) S_z dS_z. \quad (3)$$

The obtained $\sigma(T)$ dependence is shown in Fig. 3. It is seen that the $\sigma(T)$ curve lies lower than the magnetization-function curve drawn in the same figure and calculated in the molecular-field approximation for a spin $S = 1$. A similar deviation was observed for the alloy PdFe with 0.4 at. % Fe in Ref. 6, where the reduced magnetization of the alloy was calculated on the basis of a molecular-field distribution function containing parameters obtained from the Mössbauer spectra. It was shown in Ref. 4 that the curve shown in Fig. 3 for $S = 1$ describes well the course of the magnetization in Pd_{97.35}Fe_{2.65} alloy with a large iron concentration. The different temperature dependences of the magnetization in alloys with different concentrations is due to the increase of the fluctuations of the exchange interaction with increasing dilution of the alloy.

The lower curve of Fig. 3 was obtained theoretically in Ref. 2, where the alloy magnetization was calculated by percolation theory and was assumed to be proportional to the number of atoms entering into an infinite cluster. The qualitative agreement between our data and the theoretical curve² confirms the applicability of the cluster model to the description of the macroscopic properties of dilute alloys.

3. Curves of local magnetization $S_z(T)$

The large set of hyperfine fields acting on the iron nuclei presupposes different hyperfine-field temperature dependences for atoms located in different magnetic environments, or the existence of different local-magnetization curves $S_z(T)$. These curves can be obtained from the plots of the $P(S_z, T)$ distributions in the following manner. Assume that the $S_z(T)$ curves corresponding to different atoms do not intersect. Then the quantity

$$I = \int_0^{S_z^i(T)} P(S_z, T) dS_z, \quad (4)$$

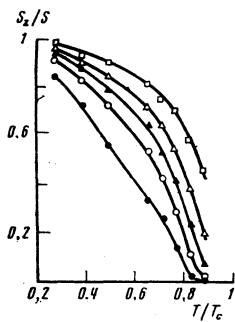


FIG. 4. Plots of local magnetization $S_z(T)$: \bullet) $I = 0.1$, \circ) $I = 0.3$; \blacktriangle) $I = 0.5$; \triangle) $I = 0.7$; \square) $I = 0.9$; $T_c = 18$ K.

which is proportional to the number of Fe impurity atoms with spin projections $S_z < S_z^i(T)$, where S_z^i is the spin of an arbitrarily chosen atom, should not depend on the temperature. Choosing an arbitrary value S_z^i on the plot of the $P(S_z, T)$ distribution, and determining from the distributions at other temperatures T_2, T_3, \dots values $S_z^i, S_z^{i'}$, \dots such that the integrals

$$\int_0^{S_z^i} P(S_z, T_1) dS_z = \int_0^{S_z^{i'}} P(S_z, T_2) dS_z = \dots$$

are all equal, we obtain for the local magnetization $S_z(T)$ the temperature dependence corresponding to the chosen value of the integral I .

Figure 4 shows a set of local-magnetization curves with different values of the integral I . Obviously, the larger I corresponds to a larger molecular field at the iron atom. It is seen from Fig. 4 that the $S_z(T)$ curves have different functional forms. The spins located in weaker molecular fields are magnetized more slowly when the temperature is lowered. This behavior can be expected if it is assumed that these spins should be subjected to a magnetization action on the part of the more strongly magnetized spins. A similar behavior was observed for impurities coupled by weak exchange interaction with the surrounding ferromagnetic matrix.⁷ The presence of slowly magnetizing spins causes also the slower course of the average-magnetization curve $\sigma(T)$ in Fig. 3.

4. Molecular-field distribution $P(H_{mol})$

The properties of dilute alloys are frequently described with the aid of the molecular-field distribution function. This function can be obtained from the distribution $P(S_z)$ of the local magnetization, by using the relation

$$P(H_{mol}) = P(S_z) dS_z / dH_{mol}. \quad (5)$$

As the function $S_z(H_{mol})$ we chose the data obtained from measurements of the Mössbauer effect in PdFe alloys in external magnetic fields.^{8,9} The obtained molecular-field distribution curves at different temperatures are shown in Fig. 5.

The corresponding theoretical calculations for the alloys PdCo and PdFe were carried out on the basis of statistical models^{10,11} that lead to very complicated mathematical expressions for the molecular field dis-

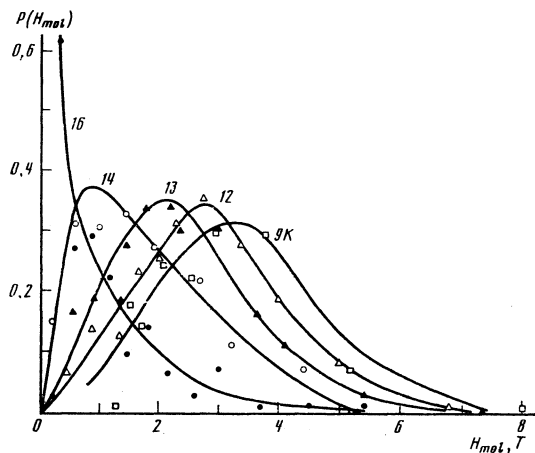


FIG. 5. Molecular field distribution $P(H_{mol})$ at various temperatures.

tribution (MFD) function. These expressions are simpler at zero temperatures, when all the impurity spins have reached saturation. Thus, in Ref. 1 were obtained MFD curves at $T=0$ were obtained for 0.2, 0.8, and 2.0 at. % numerically with a computer. The analytic form of $P(H_{mol})$ in two limiting cases, at the distribution end points, was investigated in Ref. 2. At nonzero temperatures it is necessary to take into account in MFD calculations the fluctuations of not only the arrangement of the impurity spins, but also of their values. We know of only one paper⁶ in which the MFD function was obtained for PdFe with 0.4 at. % iron was obtained at $T \neq 0$ by a self-consistent computer calculation. However, because of the incorrectly chosen statistical model and to the wrong form of the interaction potential between the impurities, the results of Ref. 6 must be regarded only as approximate (see the footnote on the second page of Ref. 12). In Ref. 1 it was assumed, to simplify the MFD calculations at $T \neq 0$, that

$$H_{mol}(r, T) = H_{mol}(r, T=0) \sigma(T). \quad (6)$$

This means that the form of the $P(H_{mol})$ distribution does not depend on temperature.

In contrast to all the cited references, we obtained the distributions of the molecular fields directly by reducing the experimental data, without resorting to any statistical models. It turned that no scale transformation can make congruent the curves of Fig. 5 that correspond to different temperatures, and the relative fluctuation of the molecular field ($\Delta H_{mol} / H_{mol}$) increases with increasing temperature. Thus, relation (6), which presupposes invariance of the form of the MFD, is incorrect. The reason is that the temperature dependence of $H_{mol}(T)$ in (6) is determined only by the value of $\sigma(T)$, and by the same token no account is taken of the essentially local character of the magnetization in a disordered dilute alloy (see Fig. 4). Unfortunately, our curves were not plotted accurately enough to permit a more detailed study of their functional form.

We note in conclusion that the method proposed by us for the construction of local-magnetization curves may be useful in the study of other disordered magnetic sys-

tems, for example amorphous magnets.

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