

HYPERFINE INTERACTIONS IN AN ORDERING Pd<sub>3</sub>Fe ALLOY

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The temperature dependence of the specific heat of the alloy Pd<sub>3</sub>Fe in the disordered and ordered states has been measured at temperatures between 0.2 and 1°K. The values of the electron (γ) and hyperfine (δ) specific heat coefficients have been determined. The values of γ remained practically constant in the ordering process. The value of δ changed from 0.010 to 0.030 mJ-deg/mole, which corresponds to a change in the effective magnetic field at the Pd<sup>105</sup> nuclei from 170 to 290 kOe. These changes are compared with data for the change in the local magnetic moments of the Pd atoms and in the mean alloy magnetic moments obtained by the neutron diffraction method. From this comparison, it is concluded that in the case of the alloy Pd<sub>3</sub>Fe the main role in the formation of H<sub>eff</sub> at the Pd<sup>105</sup> nuclei is played by the Fermi contact interactions, both with uncompensated core s-electrons and with polarized conduction electrons.

ONE of the most pressing problems in the study of hyperfine interactions in transition metals and alloys is the elucidation of the correlations between the effective magnetic field at the nucleus and the local magnetic moments of the atoms, and also the study of the role of the conduction electrons in these interactions. Concentrated Pd-Fe alloys present us with a good opportunity for studying this problem.

In the process of atomic ordering, which is observed in these alloys in concentration regions close to the compositions AB and AB<sub>3</sub>, the character of the distribution of neighboring atoms changes, leading, as a rule, to considerable changes in the local magnetic moments. It is important to note that, in the case of Pd<sub>3</sub>Fe, these changes proceed without change of the crystal lattice parameters of the alloy. It was shown earlier<sup>[1]</sup> that in the alloy Ni<sub>3</sub>Mn, isomorphous with the alloy Pd<sub>3</sub>Fe, a very sharp change in H<sub>eff</sub> at the Mn<sup>55</sup> nuclei is observed during ordering.

In the present work we have investigated the effective magnetic field at the Pd<sup>105</sup> nuclei in the alloy Pd<sub>3</sub>Fe in the disordered and ordered states by measuring the nuclear specific heat at extremely low temperatures.

In general, the specific heat of a ferromagnetic metal can be represented in the form of a sum of terms:

$$C = C_{lat} + C_{sp} + C_{el} + C_{hyp}$$

where C<sub>lat</sub> = αT<sup>3</sup> is the Debye lattice specific heat, C<sub>sp</sub> = βT<sup>3/2</sup> is the spin-wave specific heat, C<sub>el</sub> = γT is the conduction-electron specific heat and C<sub>hyp</sub> = δT<sup>-2</sup> is the specific heat due to the hyperfine interactions, if the temperature is far enough from the Schottky anomaly maximum. Since the crystal lattice of the alloy Pd<sub>3</sub>Fe has cubic symmetry and is not deformed in the ordering process, the electric quadrupole hyperfine interaction gives no contribution to the specific heat and the quantity C<sub>hyp</sub> is due entirely to the magnetic interactions. Therefore, in accordance with<sup>[2]</sup>, for ferromagnetic materials we can write, for a gram-molecule of material,

$$C_{hyp} = \frac{1}{3} \frac{R}{(kT)^2} \frac{I+1}{I} (\mu_n H_{eff})^2 + \dots$$

where I is the spin of the interacting nucleus and μ<sub>n</sub> is the nuclear magnetic moment in nuclear magnetons.

In the case of the alloy Pd<sub>3</sub>Fe, the contribution of interactions with the Fe<sup>57</sup> nuclei to the total hyperfine interaction energy will be small, and since, for the composition Pd<sub>3</sub>Fe, the Pd atoms are in equivalent crystallographic positions, it is possible to calculate unambiguously the magnitude of the effective magnetic field at the Pd<sup>105</sup> nuclei from the experimentally determined values of the coefficient δ.

The measurements were made on a polycrystalline sample of the alloy Pd<sub>3</sub>Fe (75.3 at.% Pd and 24.7 at.% Fe from chemical analysis data), smelted in an induction furnace in an atmosphere of purified Ar. The starting materials were spongy Pd of 99.83% purity and carbonyl iron of 99.999% purity.

The disordered state of the alloy was attained after annealing at 1000°C for 15 hours and rapid quenching in water. To obtain the ordered state, the alloy was annealed for 215 hours at 600°C. The presence or absence of long-range order in the alloy was monitored by the presence or absence of superlattice reflections (100) in the Debye crystallograms. A quantitative determination of the degree of long-range order in the alloy was not made.

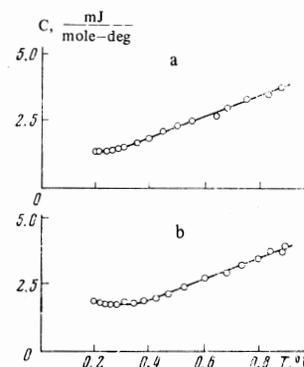


FIG. 1. Dependence on temperature of the specific heat C of the alloy Pd<sub>3</sub>Fe: a) —disordered state b) —ordered state.

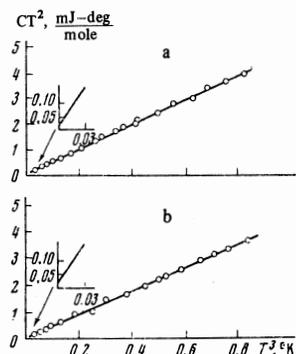


FIG. 2. Dependence of  $CT^2$  on  $T^3$  for the alloy  $Pd_3Fe$ : a) —disordered state, b) —ordered state.

	Disordered alloy	Ordered alloy
$\alpha$ , mJ/mole-deg <sup>2</sup>	4.3	4.4
$\delta$ , mJ-deg/mole	0.010	0.030
$H_{eff}$ , kOe	170	290

The specific heat was measured in a calorimeter with adiabatic demagnetization of a block of potassium chromium alum, connected to the sample by means of a thermally superconducting link. The temperature measurements were made by means of an "Allen Bradley" carbon thermometer, which was calibrated against the magnetic susceptibility of the potassium chromium alum and against the He vapor pressure on a mirror in the ampoule. A detailed description of the experimental setup and of the technique of the measurements is given in<sup>[3]</sup>.

The temperature dependence of the specific heat of (a) the disordered and (b) the ordered  $Pd_3Fe$  alloys is shown in Fig. 1. In the lowest temperature region an increase of specific heat with lowering of temperature is observed; this is the beginning of the Schottky anomaly due to the magnetic hyperfine interaction.

In Fig. 2 the dependence of  $CT^2$  on  $T^3$  is shown for the disordered and ordered states of the alloy respectively. As can be seen, the experimental points in both cases are well positioned on straight lines. This makes it possible to approximate the temperature behavior of the specific heat by the expression  $C = \gamma T + \delta T^{-2}$ . The values of the coefficients in the electron and hyperfine specific heats were calculated by the least squares method and are given in the table. Here also we give values of the effective magnetic fields at the  $Pd^{105}$  nuclei, calculated from the values of  $\delta$ .

The magnitude of the coefficient in the electron specific heat does not change, within the limits of experimental error, in the ordering process. An analogous situation was observed earlier<sup>[4]</sup> in an investigation of the specific heat of the alloy  $Cu_3Pd$ , which is isomorphous with the alloy  $Pd_3Fe$ . This is evidence that the change in the effective radius of the Brillouin zone on formation of the superlattice in the ordering process gives essentially no change in the relative positions of the Fermi level and the electron density-of-states curve.

During the ordering, substantial changes occur in the coefficient  $\delta$  in the hyperfine term in the specific heat. It is appropriate to compare these changes with

changes in the local Pd magnetic moment and in the mean moment of the alloy, measured by neutron diffraction methods. Campbell<sup>[6]</sup> has investigated experimental data on measurements of the local magnetic moments of impurities in Fe and of the effective magnetic fields at the nuclei of these impurities; in the empirical relationship

$$H_{eff} = a\mu_{loc} + b\mu_{mean}$$

for Pd-group element impurities in Fe, he has proposed the following values for the coefficients:  $a = -250 \text{ kOe}/\mu_B$  and  $b = -150 \text{ kOe}/\mu_B$ .

Using the magnetic moment values obtained by the neutron diffraction method and the above values of the coefficients, one can obtain the magnitude of  $H_{eff}$  at the  $Pd^{105}$  nuclei in the alloy  $Pd_3Fe$ . For the disordered state,  $H_{eff} = 217 \text{ kOe}$ , and for the ordered state,  $H_{eff} = 275 \text{ kOe}$ . These values are in fair agreement with our experimental data, given in the table. Here it must be remarked that the degree of atomic order may be somewhat different in our samples from that in the samples of<sup>[5]</sup>.

If we assume that the local moment of the Pd atoms is due to the electrons of an uncompensated d-shell, we can compare our data on the variation in the magnitude of the effective field at the  $Pd^{105}$  nuclei with change in the local moment,  $\Delta H_{eff}/\Delta\mu_{loc} = 667 \text{ kOe}/\mu_B$ , with the value, obtained in<sup>[7]</sup> by a nuclear resonance method, of the effective field due to one d-spin,  $H_{eff}/\mu_B = 689 \text{ kOe}/\mu_B$ .

These comparisons indicate that in the case of Pd and its alloys the resultant magnitude of  $H_{eff}$  is due to contributions from interactions with s-electrons of uncompensated inner shells and also to the contribution of interactions with the conduction electrons; these contributions can be assumed to be additive. Taking account of only one of these contributions gives significantly worse agreement with the experimental results.

In conclusion the authors express their deep gratitude to E. I. Kondorskiĭ for valuable discussions of the work.

<sup>1</sup> P. N. Stetsenko and Yu. I. Avksent'ev, Proceedings of the International Conference on Magnetism, Nottingham, 1964, p. 217 (Published by Institute of Physics and the Physical Society).

<sup>2</sup> W. Marshall, Phys. Rev. 110, 1280 (1958).

<sup>3</sup> P. N. Stetsenko and Yu. I. Avksent'ev, Prib. Tekh. Eksp. 6, 178 (1965) [Instrum. Exp. Tech. (USSR) 1492 (1966)].

<sup>4</sup> P. N. Stetsenko and Yu. I. Avksent'ev, Ann. Acad. Sci. Fenn., Ser. A VI (Phys.) 210, 226 (1966).

<sup>5</sup> J. W. Cable, E. O. Wollan, W. C. Koehler, and M. K. Wilkinson, J. Appl. Phys. 33, 1340 (1962).

<sup>6</sup> I. A. Campbell, Proc. Phys. Soc., 89, 71 (1966).

<sup>7</sup> J. A. Seitchik, A. C. Gossard, and V. Jaccarino, Phys. Rev. 136, 1119A (1964).