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†Excerpt from Ph.D. Thesis, University of California, San Diego, March, 1968. See also D. Wohlleben, *Bull. Am. Phys. Soc.* **13**, 363 (1968).

¹H. E. Nigh, S. Legvold, F. H. Spedding, and B. J. Beaudry, *J. Chem. Phys.* **41**, 3799 (1964).

²H. R. Child and W. C. Koehler, *J. Appl. Phys.* **37**, 1353 (1966).

³The result of an extensive study of the susceptibility of pure Sc metal will be published elsewhere.

⁴J. Crangle, *Phys. Rev. Letters* **13**, 569 (1964).

⁵H. Nagasawa and T. Sugawara, *J. Phys. Soc. Japan* **23**, 711 (1967).

⁶A. Blandin and F. Friedel, *J. Phys. Radium* **20**, 166 (1959).

⁷D. K. Finnemore, D. C. Hopkins, and P. E. Palmer, *Phys. Rev. Letters* **15**, 891 (1965).

⁸W. C. Thoburn, S. Legvold, and F. H. Spedding, *Phys. Rev.* **110**, 1298 (1958).

⁹F. Popplewell, A. M. Harris, and K. S. Tebble,

Proc. Phys. Soc. (London) **85**, 347 (1965).

¹⁰R. P. Guertin, F. E. Crow, and R. D. Parks, *Phys. Rev. Letters* **16**, 1095 (1966).

¹¹A. M. Toxen, P. C. Kwok, and R. F. Gambino, *Phys. Rev. Letters* **21**, 792 (1968).

¹²F. Owen, M. E. Brown, V. Arp, and A. F. Kip, *Phys. Chem. Solids* **2**, 85 (1957).

¹³V. I. Chechernikov, I. Pop, O. P. Naumkin, and V. F. Terekhova, *J. Exptl. Theoret. Phys.* **44**, 387 (1963).

¹⁴W. E. Gardner and J. Penfold, *Phil. Mag.* **11**, 549 (1965).

¹⁵B. Giovannini, M. Peter, and J. R. Schrieffer, *Phys. Rev. Letters* **12**, 736 (1964).

¹⁶A. M. Clogston, *Phys. Rev. Letters* **19**, 583 (1967).

¹⁷A. Arrott, in *Magnetism*, edited by G. T. Rado and H. Suhl (Academic Press, Inc., New York, 1966), Vol. II(B), p. 328.

¹⁸G. S. Fleming and T. L. Loucks, to be published, and private communication. These authors point out a characteristic wave vector which is derivable from the band structure of Sc and seems to be related to a spiral magnetic structure in Sc:Tb alloys.

¹⁹M. A. Jensen, thesis, University of California, San Diego, 1965 (unpublished).

MAGNETIC ORDERING IN PALLADIUM-IRON ALLOYS

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We have studied the temperature dependence of the electrical resistivity of palladium-iron alloys in the composition range 0.5–12 at.% Fe with particular emphasis on the neighborhood of the magnetic-ordering temperature. Values of the critical temperature T_C determined from the temperature of the maximum of $d\rho/dT$ fall consistently below those obtained from saturation-magnetization measurements. The dependence of T_C on Fe concentration is discussed and correlated with other properties of this system.

The ferromagnetic-ordering temperature in palladium-iron alloys has been studied by a variety of methods including saturation magnetization,^{1,2} Mössbauer effect,^{2,3} heat capacity,⁴ and electrical resistivity,^{5,6} but there are significant discrepancies between the ordering temperatures deduced from the different measurements. We report here a systematic study of the electrical resistivity in a series of Pd:Fe alloys ranging in composition from 0.5 to 12 at.% Fe with particular emphasis on the temperature dependence in the neighborhood of the magnetic-ordering temperature where pronounced anomalies in the temperature dependence of the electrical resistivity are observed. As the criterion for determining the ordering temperature (T_C) we take the temperature at which there is a maximum in the tempera-

ture derivative of the resistivity, $d\rho/dT$. The recent measurements of Craig et al.⁷ on ferromagnetic nickel have demonstrated the relevance of such a criterion, but we might also mention that this choice gives values for T_C which are consistent with values obtained from the most reliable analysis of the data obtained by other methods.

The measurements were made using the standard four-point probe technique on samples which were usually about 3×10^{-3} cm thick, 2 mm wide, and about 2 cm long prepared from homogenized ingots. Data for all the samples were obtained between 4.2 and 300°K. For each sample $d\rho/dT$ was obtained from the $\rho(T)$ data by point-by-point differentiation using a computer. The temperature dependence of ρ and $d\rho/dT$ in the neighborhood of the ordering temperature for a sample of

$\text{Pd}_{94}\text{Fe}_6$ is shown in Fig. 1. The behavior shown in Fig. 1 is representative of the concentration range studied except below 2 at.%. In particular all the $\rho(T)$ curves show a pronounced break in the neighborhood of the ordering temperature, and the $d\rho/dT$ curves show a gentle rise towards the maximum and a sharp drop just above the maximum. For samples of 2 at.% and above, the magnetic transition occurs in a temperature range where the phonon scattering dominates and ρ (in the absence of spin-dependent effects) is essentially linear in T . However, in the more dilute alloys, the transition occurs in a region where ρ is a much more rapidly varying function of T , and therefore the details of the $\rho(T)$ and $d\rho/dT$ curves are somewhat altered from those shown in the figure. The break in $\rho(T)$ around 173°K, which we believe is due to the magnetic transition, is not extremely sharp. The ordering temperature obtained from the maximum of $d\rho/dT$ occurs at the lower temperature 161°K. We believe that the width of the transition region is a manifestation of short-range order immediately above T_C which in $\text{Pd}_{94}\text{Fe}_6$ extends to about 185°K.

The behavior of $d\rho/dT$ shown in Fig. 1 is very similar to the behavior in Ni reported in Ref. 7 in which $d\rho/dT$ was measured directly. It is interesting to do a critical-point analysis in the

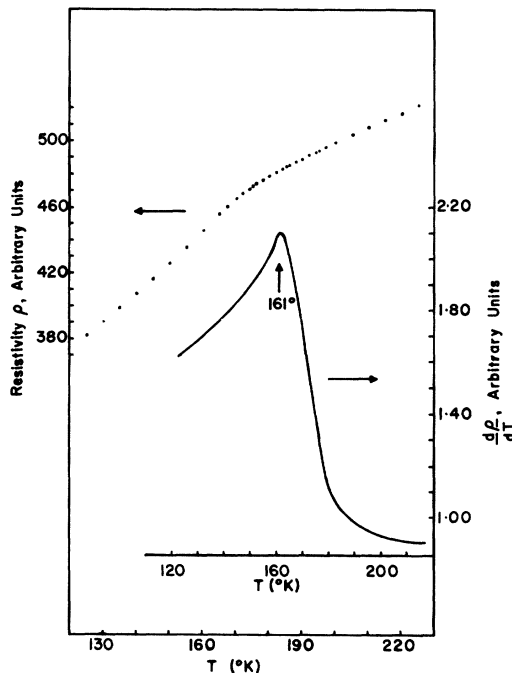


FIG. 1. $\rho(T)$ and $d\rho/dT$ for a sample $\text{Pd}_{94}\text{Fe}_6$ in the neighborhood of the magnetic-ordering temperature.

immediate neighborhood of the magnetic transition. Our data for $\rho(T)$ were used to generate $d^2\rho/dT^2$ via computer for samples with Fe concentrations of 3, 6, and 9 at.%. Although there is scatter in the resulting values for $d^2\rho/dT^2$, an analysis similar to that in Ref. 7 yielded no evidence of a logarithmic divergence above T_C up to temperatures 50°K higher than T_C . In fact $d\rho/dT$ varies as $(T-T_C)^{-\lambda}$, where $1.2 < \lambda < 1.9$ in rough agreement with the behavior in nickel metal.⁷ However, the temperature above T_C at which this behavior sets in is approximately proportional to the impurity concentration; for the $\text{Pd}_{94}\text{Fe}_6$ sample it occurs at about 16°K above T_C . Below this temperature $d^2\rho/dT^2$ decreases slowly with temperature. It is clear that the mean free path plays an important role in determining the range of the critical spin fluctuations above T_C .⁷ A more complete discussion of our results along with additional data will be presented in a future publication. Fisher and Langer⁸ have given a theoretical treatment of the problem of critical fluctuations in which the importance of short-range fluctuations above T_C is stressed. In particular they argue that above T_C , $d\rho/dT$ and the magnetic specific heat should behave similarly. It is very interesting to note the similarity between $d\rho/dT$ shown in Fig. 1 and the magnetic-specific-heat curves for a few lower concentration samples obtained by Veal and Rayne.⁴ In both cases one has a rise below a maximum, which is roughly linear, and then a sharp drop above the maximum.

The magnetic-ordering temperature for our series of alloys, defined to be the temperature of the maximum of $d\rho/dT$, is shown as a function of iron concentration in Fig. 2. A few of the results of other determinations are also shown for comparison purposes. The first thing to note is that our results fall consistently below the values of T_C obtained by saturation magnetization measurements, and also below the values of Coles, Wazsink, and Loran,⁵ who also measured the electrical resistivity. The latter discrepancy is due to the fact that in Ref. 5 the ordering temperature is taken to be the temperature of the kink in $\rho(T)$ rather than the maximum of $d\rho/dT$. It is clear from Fig. 1 that this kink will appear above the temperature of the maximum of $d\rho/dT$. The disagreement between the saturation magnetization determinations of T_C and our results is most likely due to the failure of the extrapolation technique used to obtain T_C from the magnetization data. These techniques usually involve a linear

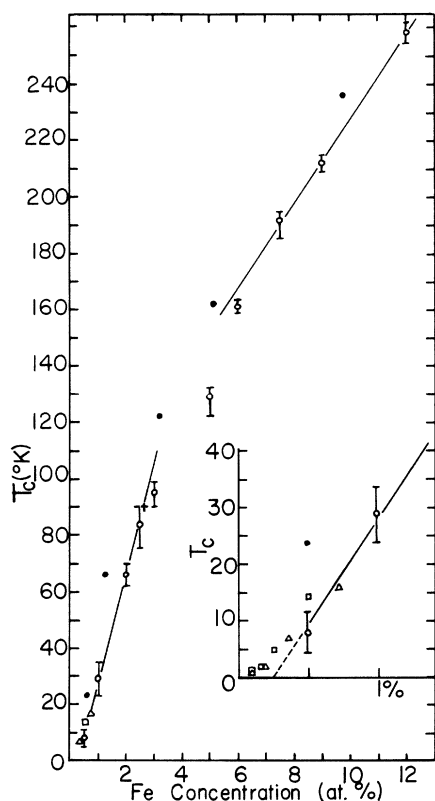


FIG. 2. Variation of the magnetic-ordering temperature T_C with Fe concentration in Pd:Fe. Open circles: maximum of $d\rho/dT$, this work; filled circles: spontaneous magnetization, Crangle and Crangle and Scott (Ref. 1); cross: Mössbauer effect (Ref. 2); square: anomaly in the electrical resistivity (Ref. 5); triangles: maximum of the magnetic specific heat, Ref. 4. For additional data, not shown above, see the references above and cited in the text. The inset shows the concentration range below 1 at.% Fe on a magnified scale. The data from Refs. 4 and 5 deviate from the linear behavior found for the maxima in $d\rho/dT$ between 0.5 and 3 at.% which extrapolates to $T_C = 0$ at a concentration of about 0.25 at.%.

extrapolation of the square of the saturation magnetization to zero applied field. Unfortunately, because of the nonlinearity in the limit of zero applied field in systems like Pd:Fe, the extrapolation is sensitive to the field dependence of the saturation magnetization at high fields. It is quite reasonable that in inhomogeneously polarized systems like Pd:Fe, the applied field plays a much more dominant role than it does in homogeneous systems. As has been shown for the Pd:Fe system by Kim and Schwartz,⁹ the range of the spin polarization of the Pd matrix is a strong function of the amount of polarization and therefore should be a sensitive function of any applied field. Craig *et al.*² have given an extensive dis-

ussion of the insufficiencies of extrapolation techniques used to obtain a T_C from saturation-magnetization data. In particular, they have shown, for a sample of $\text{Pd}_{97.35}\text{Fe}_{2.65}$, that the extrapolated T_C is considerably higher than T_C obtained from the sharp change in the Mössbauer linewidth (the appearance of a hyperfine splitting due to an internal field in the absence of an applied field). We can argue that the appearance of this internal field, if it were a local effect rather than a result of long-range magnetic order, and if we ignore possible relaxation effects, would come at a temperature higher than the temperature at which long-range order sets in rather than lower as is observed. Therefore, we would conclude that the Mössbauer determination of T_C is a better indicator of the long-range magnetic-ordering temperature. The value for T_C obtained from the Mössbauer measurements on $\text{Pd}_{97.35}\text{Fe}_{2.65}$ is in excellent agreement with our $d\rho/dT$ data and gives us confidence in the reliability of our criterion for choosing T_C .

A few points from Veal and Rayne⁴ giving the temperature of the maximum of the magnetic specific heat are also shown for comparison in Fig. 2. Again, since there is evidence of considerable short-range order in the Pd:Fe system, we argue that it is more appropriate to take the temperature of the maximum of the magnetic specific heat rather than the temperature at which it vanishes as the better indicator of the presence of long-range order. The tail in the specific heat (as in $d\rho/dT$) above T_C is then a measure of the short-range order present.

Now that we have discussed the differences between the values of T_C obtained from different measurements we would like to point out some interesting features of the data shown in Fig. 2. First of all, between 0.5 and 3 at.% Fe, T_C increases linearly with iron concentration. It is clear from other measurements,^{4,5,10} however, that this linear dependence does not hold for concentrations below 0.5%. This nonlinear behavior of T_C with Fe concentration we believe can be explained in terms of the model of clusters of polarization, surrounding each impurity atom, which have a large size (as observed by neutron scattering¹¹) and couple together to produce a long-range-ordered state. It is reasonable to expect that, in this region, T_C would probably be quadratic in the iron concentration, i.e., proportional to the square of the number density of clusters. Note that if one extrapolates to $T_C = 0$ the lower concentration linear portion of the data

shown in Fig. 2, the intercept is at a concentration of 0.25% where there is a maximum in the giant moment.¹ Furthermore, it has been proposed¹² that at about this concentration one would expect a peak in the high-field susceptibility resulting from the band splitting induced by the iron impurities. This splitting places the Fermi level at the peak in the density of states which gives rise to the maximum in the pure Pd susceptibility at about 80°K. It is also interesting that a deviation from the lower concentration linear region shown in Fig. 2 sets in at about 3% where the neutron scattering cross section becomes independent of the scattering vector and one might conclude that the Pd polarization becomes essentially uniform.¹¹

Furthermore, above about 6% Fe in Pd, T_C again becomes linear in concentration with a slope which is roughly a factor of 3 smaller than in the lower concentration linear region. If one extrapolates to zero concentration the higher concentration linear region shown in Fig. 2, one obtains a T_C intercept of 80°K. This coincides with the temperature of the peak in the pure Pd susceptibility. If pure Pd were a true ferromagnet rather than an incipient ferromagnet, its Curie temperature would correspond to the maximum of the observed susceptibility, namely 80°K. It appears therefore that the linear higher concentration dependence of T_C on iron concentration, where the Pd polarization is uniform and each Pd has on the average at least one iron near neighbor, is determined essentially by the Pd band splitting produced by the iron impurities acting simply as the source of a uniform molecular field.

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ter submission of this Letter a parallel and independent study by Longworth and Tsuei¹³ of the critical behavior of the resistivity in Pd:Fe alloys appeared.

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¹J. Crangle, *Phil. Mag.* **5**, 335 (1960); J. Crangle and W. R. Scott, *J. Appl. Phys.* **36**, 921 (1965); A. M. Clogston et al., *Phys. Rev.* **125**, 541 (1962); R. M. Bozorth, D. D. Davis, and J. H. Wernick, *J. Phys. Soc. Japan* **17**, Suppl. B1, 112 (1962); M. McDougald and A. J. Manuel, *J. Appl. Phys.* **39**, 961 (1968).

²P. P. Craig, R. C. Perisho, R. Segnan, and W. A. Steyert, *Phys. Rev.* **138**, A1460 (1965).

³W. L. Trousdale, G. Longworth, and T. A. Kitchens, *J. Appl. Phys.* **38**, 922 (1967).

⁴B. W. Veal and J. A. Rayne, *Phys. Rev.* **135**, A442 (1964).

⁵B. R. Coles, J. H. Waszink, and J. Loran, in Proceedings of the International Conference on Magnetism, Nottingham, England, 1965 (The Institute of Physics and The Physical Society, London, England, 1965), p. 165.

⁶R. Schwaller, thesis, University of Strasbourg, 1967 (unpublished).

⁷P. P. Craig, W. I. Goldberg, T. A. Kitchens, and J. I. Budnick, *Phys. Rev. Letters* **19**, 1334 (1967).

⁸M. E. Fisher and J. S. Langer, *Phys. Rev. Letters* **20**, 665 (1968).

⁹D. J. Kim and B. B. Schwartz, *Phys. Rev. Letters* **20**, 201 (1968).

¹⁰A systematic study of the concentration region below 0.5% is now being carried out by some of us.

¹¹G. G. Low and T. M. Holden, *Proc. Phys. Soc. (London)* **89**, 119 (1966).

¹²J. I. Budnick, J. Lechaton, J. H. Wernick, S. Foner, E. J. McNiff, D. J. Kim, and B. B. Schwartz, *J. Appl. Phys.* **39**, 960 (1968).

¹³G. Longworth and C. C. Tsuei, *Physics Letters* **27A**, 258 (1968).