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Spin-Glass-Like Behavior in Very Dilute *Pd*Fe at Very Low Temperatures

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Measurements of the electrical resistivity of palladium, residual resistivity ratio ~22 500, containing 1.7 ppm of Fe have been made from 5.5 K to 1.8 mK. 15 Hz ac susceptibility measurements from 1 K to 1.8 mK together with the resistivity data suggest that a transition into a spin-glass state occurs near 7 mK.

The low-temperature properties of pure palladium as well as its alloys have been the subject of numerous theoretical and experimental investigations in recent years.[1] Pure Pd exhibits a large susceptibility enhancement over the normal Pauli susceptibility and is nearly ferromagnetic. Scattering of electrons from spin density fluctuations (paramagnons) are thought to play an important role in the electrical resistivity at temperatures below about 6 K. Furthermore, just as in the case of superfluid ^{3}He, it is believed that for a very pure sample at sufficiently low temperatures these paramagnon effects could result in an attractive interaction strong enough to allow pairing of the electrons in a triplet $S = 1$ spin state leading to P-wave superconductivity.[2,3] The addition of as little as 0.1% Fe or Co to Pd results in ferromagnetism with the effective moment of the magnetic impurity enhanced as much as a factor of 2 or more. Recently it has been predicted that at very low Fe concentrations, less than 600 ppm, *Pd*Fe will exhibit a spin-glass transition.[4] In this Letter, we present both electrical resistivity and susceptibility data that suggests Pd with only 1.7 ppm of Fe exhibits such a transition at $\cong 7$ mK.

The adiabatic demagnetization cell used for these experiments has been described elsewhere.[5] The Pd sample was contained in a magnetically shielded tower located above the refrigerant and thermal contact was provided by liquid ^{3}He. Resistivity measurements were made using a superconducting quantum interference device (SQUID) in an ac bridge configuration[6] in the frequency range 3 Hz to 160 Hz. Susceptibility measurements were performed at 15 Hz using a second SQUID. Temperatures below 0.3 K were determined from 15-Hz mutual-inductance measurements on 10 mg of cerium magnesium nitrate located in a second magnetically shielded tower using a third SQUID. Above 0.3 K, temperatures were determined with use of a calibrated germanium resistance thermomenter. The 14-mm-long and 0.95-mm-average-diameter sample of Pd was prepared with use of a zone-refining technique in air.[7] Current leads were soft soldered to the ends of the sample and the superconducting voltage leads were connected with use of a minimum amount of soft solder 3.5 mm from each end. The room temperature to 4.2 K resistance ratio was measured using a standard four-wire dc technique and found to be 15 300.

We employed two techniques for determining the low-temperature resistivity. The first of which was to measure the frequency dependence of the resistivity at low frequencies and extrapolate to zero frequency. The disadvantage of this approach is that the precision of the measurement quickly decreases with decreasing frequency because most of the Johnson-noise power coupled to the SQUID, arising from the random thermal fluctuations of the conduction electrons in our sample, is confined to be within the ≈ 0.3 rad/sec characteristic frequency of the measuring circuit.[8] The advantage of this technique is that skin depth considerations become less important as the frequency is lowered. The second approach was to make use of the fact that at high frequencies the resistivity is proportional to the square root of the measuring frequency and the slope,

$\Delta\rho(\omega)/\Delta\omega^{1/2}$, is equal to $br(\mu\rho)^{1/2}$, where r is the radius of the sample, μ is the magnetic permeability, b is a constant that depends only on the geometry of the sample, and ρ is the zero-frequency resistivity. Thus a measurement of this slope should be linearly proportional to the square root of the zero-frequency resistivity, so long as the magnetic permeability of the sample is constant. The advantage here is that the precision associated with the measurement of $\rho(\omega)$ at high frequencies, can be several orders of magnitude greater than a similar measurement[6] near $\omega \sim 0$. Also, the time required to obtain an accurate measurement of the slope was only a few hours, while for the extrapolation technique more than six hours of measurements were frequently required. From 15 mK to 5.5 K the value of the zero-frequency electrical resistivity determined with use of both techniques agreed to better than 0.5%.

The electrical resistivity from 40 mK to 5.5 K measured in a 0.001-Oe magnetic field was found to be equal to

$$\rho(T) = 4.28\times10^{-10} + 1.59\times10^{-11}T^2(\mathrm{K})\ \Omega\ \mathrm{cm}, \quad (1)$$

leading to a $T=0$ residual resistance ratio of 22 500. Both electron-paramagnon and electron-electron scattering can contribute to the T^2 term, but the relative importance of these contributions cannot be experimentally determined. The coefficient of the T^2 term in Eq. 1 is nearly a factor of 2 smaller than that reported in earlier work[9,10,11] using, for the most part, polycrystalline samples. In contrast, our sample was a single crystal, carefully handled to avoid any strains during mounting or cooling, with a purity nearly 15 times higher than in the earlier work. This suggests that there may be a significant difference in the T^2 behavior of the electrical resistivity of Pd in the clean and dirty limits.

Figure 1 displays the low-temperature resistivity data obtained in 0.001-Oe static field using both the slope and $\omega=0$ extrapolation techniques. The solid curve is the fit to the data using Eq. (1). Below 40 mK the resistivity decreases somewhat faster than expected from Eq. (1) and below 15 mK the resistivity determined using the high-frequency slope decreases faster than that determined from the $\omega=0$ extrapolation. This suggests that either the permeability of the sample is decreasing below 15 mK or that something more complicated in the dynamical response of this system is occurring at these low temperatures.

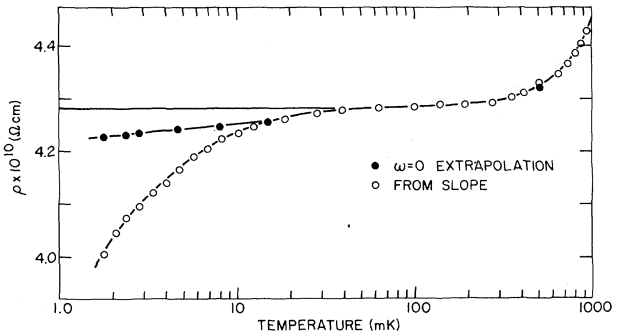

FIG. 1. Electrical resistivity of Pd as a function of temperature in a 0.001-Oe field. Solid curve is a fit to the data from 5.5 K to 60 mK assuming $\rho(T)=\rho_0+bT^2$. Open circles are data obtained from the slope $\Delta\rho(\omega)/\Delta\omega^{1/2}$. Solid circles are data obtained from an $\omega=0$ extrapolation.

Evidence for the latter is shown in Fig. 2 where the change in the 15-Hz susceptibility of this sample relative to the susceptibility at 1 K is displayed as a function of temperature in a 20-Oe static field. The peak-to-peak ac measuring field was 6×10^{-4} Oe. Because of the presence of the solder used to connect the leads to the sample, an absolute value of the susceptibility could not be determined. From 1 K to $\cong$20 mK the susceptibility is Curie-like with a Curie constant of $C=7.1\times10^{-5}$ K. Below 20 mK the susceptibility increases much more slowly than expected from a Curie behavior, reaches a maximum value near 7 mK and decreases fairly rapidly with decreasing temperature below 7 mK. The maximum in

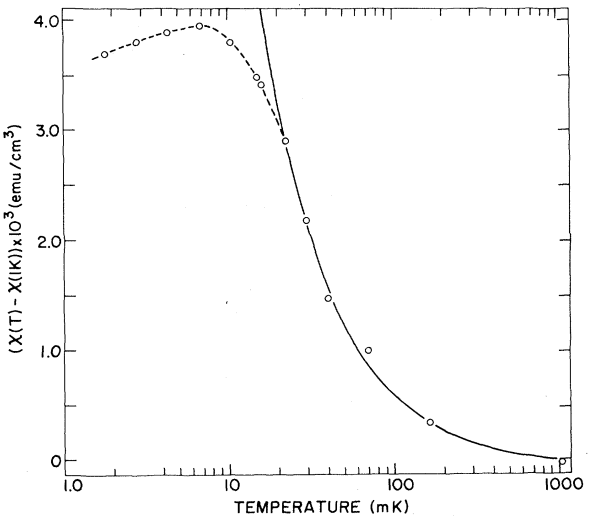

FIG. 2. 15-Hz susceptibility of Pd as a function of temperature in a 20-Oe static field. The peak-to-peak measuring field was 0.6×10^{-3} Oe. The solid curve is a fit assuming $X(T)-X(1\ \mathrm{K})=C/T$.

the ac susceptibility indicates that a magnetic ordering has occurred in this sample and that this ordering is not ferromagnetic in character.

After this experiment was completed, the sample was analyzed using a very sensitive atomic absorption technique and was found to contain 1.7 ± 0.3 ppm of Fe by weight and from a batch analysis of the starting material, before zone refining, the level of all other magnetic impurities is known to be well below 1 ppm. Using the magnitude of the measured Curie constant together with the Fe concentration, we find that the effective magnetic moment of Fe in this sample is $(7.6 \pm 0.7)\mu_B$ which is in reasonable agreement with the concentration dependence of the effective Fe moment in Pd reported by Crangle and Scott.[12]

The observed maximum in the ac susceptibility is a feature exhibited by most metallic spin glasses[13] but generally the zero-field susceptibility peak is much sharper than that displayed in Fig. 2. However, in the presence of a small static field (compared with the field which saturates the remnant magnetization of a spin glass), it is known that the susceptibility peak becomes broadened and the temperature at which the maximum occurs, the spin-glass ordering temperature T_G, is decreased slightly.[14] Furthermore, there is evidence that in some spin glasses the peak in ac susceptibility becomes less well defined as the magnetic impurity concentration is reduced.[13]

Another interesting feature of our data is shown in Fig. 3 where the low-temperature field dependence of the electrical resistivity is displayed. The data displayed were obtained using the slope technique but for the 200-Oe, 70-Oe, and 20-Oe data there was better than 1% agreement with the $\omega = 0$ extrapolation procedure. The 20-Oe, 70-Oe, and 200-Oe data were obtained after first warming the experimental cell up to 9 K and trapping the field inside the superconducting shield. The electrical resistivity measured in these fields indicates that the sample exhibits a positive magnetoresistance. However, the 29.6-Oe data displayed in Fig. 3 were obtained from two separate demagnetizations by first cooling the sample to 1.8 mK in 0.001 Oe and then applying the field via a long superconducting coil that completely surrounded the sample. The resulting resistivity below 7 mK (the temperature of the ac susceptibility maximum) was always *smaller* than the zero-field value. Above 7 mK, the 29.6-Oe resistivity was always larger than the zero-field data and above 10 mK the resistivity was approximately equal to the value one would expect from the magnetoresistance behavior of the other data.

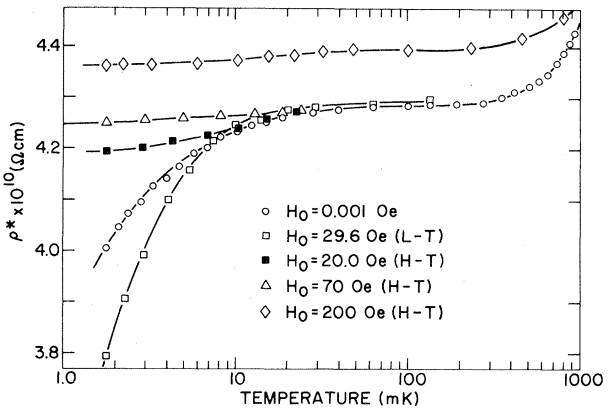

FIG. 3. Field and temperature dependence of the resistivity of Pd. All fields except the 29.6-Oe field were trapped inside a superconducting shield at high temperatures.

To our knowledge this type of field dependence of the resistivity has never been observed in a system that orders antiferromagnetically but is similar to the kind of hysteretic behavior observed for the magnetization in spin-glasses below the ordering temperature.[14] For example, if a field is applied to CuMn, a well known spin-glass system, after first cooling through T_G in zero field, the resulting magnetization is much smaller, initially than the magnetization one would measure if the sample were cooled through T_G in the same field, and increases logarithmically in time with a time constant that can be on the order of days or longer.[15]

The results presented here indicate that *Pd*Fe with only 1.7 ppm of Fe undergoes a transition from a paramagnetic state into some new magnetic state which is neither ferromagnetic nor antiferromagnetic. Furthermore, the absence of any logarithmic temperature dependence of the electrical resistivity would seem to rule out any possible Kondo type of phenomena. However, this ordering is very similar to the type of order present in most metallic spin-glasses and supports the recent theoretical predictions that *Pd*Fe should exhibit spin-glass behavior at low Fe concentrations. Further measurements of the field dependence of both the ac and dc susceptibilities for several concentrations of Fe, as well as studies of the spin dynamics, will be necessary before the exact nature of this very low-temperature transition can be determined.

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[1]G. J. Nieuwenhuys, Adv. Phys. 24, 515 (1975).
[2]D. Fay and J. Appel, Phys. Rev. B 16, 2325 (1977).
[3]K. Levin and O. Valls, Phys. Rev. B 17, 191 (1978).
[4]G. J. Nieuwenhuys, Phys. Lett. 67A, 237 (1978).
[5]R. A. Webb, J. B. Ketterson, W. P. Halperin, J. J. Vuillemin, and N. B. Sandesara, J. Low Temp. Phys. 32, 659 (1978).
[6]R. P. Giffard, R. A. Webb, and J. C. Wheatley, J. Low Temp. Phys. 6, 533 (1972).
[7]N. B. Sandesara and J. J. Vuillemin, Metall. Trans. 8B, 693 (1977).
[8]R. A. Webb, R. P. Giffard, and J. C. Wheatley, J. Low Temp. Phys. 13, 383 (1973).
[9]C. Uher and P. A. Schroeder, J. Phys. F 8, 1 (1978).
[10]G. K. White and S. B. Woods, Philos. Trans. Roy. Soc. London 251A, 273 (1959).
[11]P. A. Schroeder and C. Uher, Phys. Rev. B 18, 3884 (1978).
[12]J. Crangle and W. R. Scott, J. Appl. Phys. 36, 921 (1965).
[13]J. L. Tholence and R. Tournier, J. Phys. (Paris), Colloq. 35, C4-229 (1978).
[14]V. Cannella and J. A. Mydosh, Phys. Rev. B 6, 4220 (1972).
[15]C. N. Guy, J. Phys. F 8, 1309 (1978).

First-Order Phase Transitions and the Three-State Potts Model

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We have used the Monte Carlo renormalization-group method to study the three-state Potts model in three and four dimensions. In both cases, we find a first-order transition without an associated discontinuity fixed point. The transition in three dimensions is "almost second order" in the sense that some evidence was found for the existence of second-order fixed points associated with singularities in the metastable region just beyond the first-order transition.

The three-dimensional, three-state, ferromagnetic Potts model[1] is described by

$$H = K_{\mathrm{nn}} \sum_{\langle ij \rangle} \delta_{\sigma_i \sigma_j}, \tag{1}$$

where $\sigma_i = 1$, 2, or 3, the sum is over the nearest-neighbor pairs, and K_{nn} includes a factor of $-1/k_B T$. It is expected to describe the critical properties of a number of physical systems such as ferromagnets with cubic anisotropy in a magnetic field.[2] Although Landau theory predicts a first-order phase transition (which is certainly correct for sufficiently high dimensionality), the transition is known to be second order in two dimensions.[3] Several attempts have been made to determine the nature of the transition in three dimensions, but it proved to be difficult to find, theoretically or experimentally, an unambigous answer to the problem,[4-20] although the experimental results point to a first-order transition.[21,22]

We have studied the three-state Potts model in both three and four dimensions using the Monte Carlo renormalization-group (MCRG) method.[23-26] We have been able to determine that the transition is first order in both cases. In addition, the rather novel position-space renormalization-group flows obtained from the MCRG analysis have clarified the nature of these first-order transitions (including the existence of metastable branches) and explained the difficulties encountered by other methods. We believe that these results should have broad application to the general theory of first-order transitions.

The usual description of first-order transitions in the context of position-space renormalization-group theory (which includes the MCRG) was first given by Nienhuis and Nauenberg.[27] They showed that a *sufficient* condition for a first-order transi-

