

uable theoretical advice, and R. A. Shanny for his support as well.

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High-Temperature Resistance Minima in Concentrated Pd-Ag and Ni-Cu Alloys

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The temperature dependence of the electrical resistivity of Pd₆₀Ag₄₀ between 4.2 and 900 K is shown to contain the same anomalies as previously found in Ni-Cu alloys in the midrange of composition. For Pd-Ag these effects cannot be due to scattering at giant polarization clouds, and are attributed to a temperature-dependent decrease of the impurity resistivity due to a reduction in *s-d* scattering with increasing temperature.

Three years ago Houghton, Sarachik, and Kouvel¹ reported that, at room temperature and above, the electrical resistivity ρ of concentrated paramagnetic Ni-Cu alloys shows a temperature dependence that is most unusual. Above ~ 200 K, in all alloys containing between about 50 and 62% copper, ρ falls with increasing T with the formation of a set of resistance minima at ~ 600 K in each case. This anomalous decrease in ρ they associated with the presence of giant polarization clouds that had previously been shown to exist in such alloys by neutron-diffraction measurements.² The suggestion was that a substantial fraction of ρ at low temperature can be attributed to electron scattering from these giant moments, but that as T rose and the clouds "dispersed" this form of scattering fell away. At the same time this reduction in ρ was overtaken by an increasing phonon resistivity, and this, qualitatively, was the origin of the resistance minima. Recently, Levin and Mills³ have reported a detailed calculation of ρ based on a model

of giant spin clusters, and discuss the internal dynamics of the clusters necessary to lead to the experimental results.

However, some further recent measurements by Houghton, Sarachik, and Kouvel⁴ have cast doubts on this interpretation. They find that in Ni-Rh alloys close to the critical composition—a series in which there is again ample evidence of giant polarization clouds—there is no suggestion of a decrease in ρ at any temperature between 2 and 700 K.

We should now like to report the existence of a resistance minimum at ~ 700 K in the alloy Pd₆₀Ag₄₀—that is, in an alloy that, while chemically equivalent to Ni-Cu, has magnetic properties that are quite different. Giant polarization clouds can only form in a metal that is *almost ferromagnetic*, and this is certainly not the case in Pd diluted by 40% Ag. Our data are shown in Fig. 1 together with measurements on a Ni₃₅Cu₆₅ sample (i.e., an alloy of similar resistivity that is paramagnetic at all temperatures) for pur-

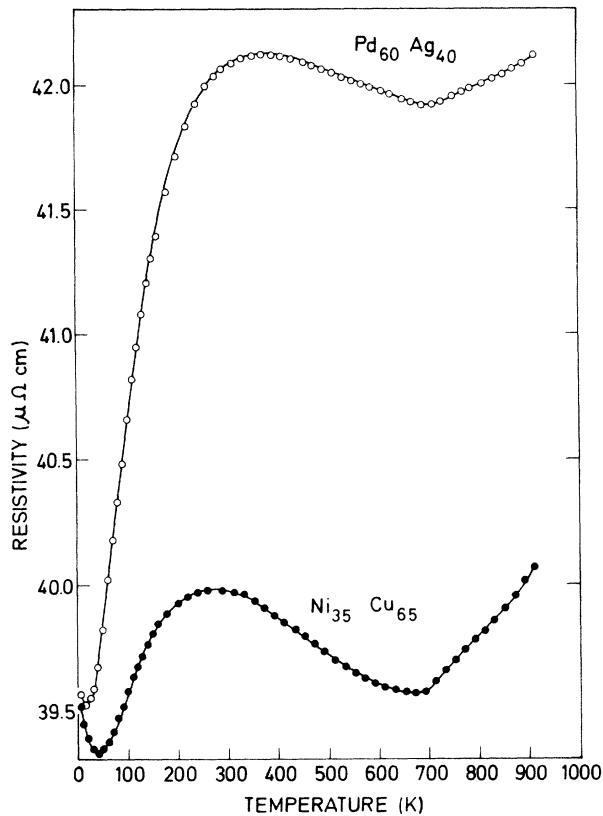


FIG. 1. Temperature variation of the electrical resistivity of the two alloys.

poses of comparison. Although the fractional change in ρ between the maximum and the high-temperature minimum is rather less in $\text{Pd}_{60}\text{Ag}_{40}$ than in $\text{Ni}_{35}\text{Cu}_{65}$, the general behavior of the two curves above 200 K is remarkably similar.

If we now consider the behavior of ρ at low temperatures, we see that there is a further similarity between the two alloys. Edwards, Chan, and Legvold³ showed the existence of resistance minima at ~ 20 K in alloys containing between 25 and 55% Ag, and in Fig. 1 we see that this behavior is reproduced in our specimen. Furthermore, as previously found by Houghton, Sarachik, and Kouvel,¹ resistance minima also occur at low temperatures in Ni-Cu in the midrange of composition, and this effect can also be seen in Fig. 1. There is therefore a striking experimental correlation between the existence of resistance minima at both high and low temperatures in concentrated Pd-Ag and Ni-Cu alloys, and a complete absence of minima at any temperature in Ni-Rh.

As an explanation of the results in Pd-Ag we propose that the decrease in ρ can be attributed

to a temperature-dependent decrease in the impurity (residual) resistivity ρ_0 brought about by a decrease in the interband s - d scattering with increasing temperature. Such an effect occurs if there is a reduction with increasing T of the mean density of available d states, $N_d(\mathcal{E})$, within a range kT of the Fermi energy \mathcal{E}_F . It can be shown^{6,7} that any resistive process, electron-phonon or electron-impurity, that ends with scattering into a d state will then be modified with increasing T by a factor which, to a first approximation, is given by $1 - AT^2$. In particular, if A is positive, the impurity resistivity at low temperatures decreases from the value at the absolute zero according to $\rho_0(T) = \rho_0(0)(1 - AT^2)$. The ideal resistivity $\rho_i(T)$, on the other hand, while also subject to this modification, always increases monotonically with T (although at different rates at different temperatures), so that the sum of $\rho_0(T)$ and $\rho_i(T)$ can lead to a resistance minimum.

In many transition metals and alloys the ideal resistivity at low temperatures is given by $\rho_i(T) \approx BT^2 + CT^4$, so that a low-temperature minimum will occur whenever $\rho_0(0)A > B$. We have recently used this mechanism to explain the existence of minima in dilute PdRh and PdRu alloys, and have argued the case in detail for favoring this rather than a "magnetic" explanation of the results.^{8,9} In these dilute alloys the inequality $\rho_0(0)A > B$ is satisfied because of the large value of A brought about by the presence of a peak at \mathcal{E}_F in the $N_d(\mathcal{E})$ curve. We now suggest that in $\text{Pd}_{60}\text{Ag}_{40}$ the same inequality can hold at low temperatures, partly because there is again a sharp change in $N_d(\mathcal{E})$ close to \mathcal{E}_F ¹⁰ leading to a fairly large A , and partly because of the unusually large measured value of $\rho_0(0)$.⁷

Around room temperature $\rho_i(T)$ is roughly proportional to T so that an impurity resistivity falling as $-AT^2$ can, in favorable circumstances, again lead to a reduction in the overall ρ with increasing T . At still higher temperatures, however, the $1 - AT^2$ factor must be modified. Since $\rho_0(T)$ must always be positive—and certainly cannot change sign at $T = A^{-1/2}$ —this modification must lead to a decrease or even a change of sign in $d\rho_0(T)/dT$. Consequently, at sufficiently high temperatures the total resistivity will always rise with T . It is therefore clear that, as at very low temperatures, conditions can again favor the formation of a minimum. For dilute palladium alloys above room temperature, $\rho_0(0)$ is always too small in comparison to $\rho_i(T)$ to give rise to

an actual minimum, but the effect is clearly seen in $PdRh$ and $PdRu$ as large negative deviations from Matthiessen's rule.⁹ In $Pd_{60}Ag_{40}$, however, $\rho_0(0)$ is sufficiently dominant to give rise to the second minimum at 700 K.

Although the detailed calculation of this effect is not easy, we should draw attention to two earlier investigations. (i) At low temperatures it is possible to obtain⁶ an analytical expression for A in terms of the first and second energy derivatives of $N_d(\mathcal{E})$ at \mathcal{E}_F , and we have used this, together with a rather crude rigid-band model, to obtain a value for $Pd + 4\%Rh$ that agrees remarkably well with the experimental result.⁸ (ii) Also Hindley,¹¹ some ten years ago, on the basis of the best model for the band structure of $Pd-Ag$ then available, calculated both $\rho_0(T)$ and $\rho_i(T)$ above room temperature, and actually obtained a shallow minimum in the total resistance of $Pd_{60}Ag_{40}$ at ~ 900 K.

Before concluding we should again point out that an exact calculation of the coefficients A and B is extremely difficult, and the simple models outlined merely serve as a guide to what can happen. Any detailed treatment must take account of alloying effects beyond the rigid-band model, and these could change A and B significantly, both from values in pure palladium and its dilute alloys, and from values estimated in concentrat-

ed alloys from the rigid-band approximation. Finally, we should add that, considering the chemical similarity of $Pd-Ag$ and $Ni-Cu$, the mechanism discussed above may also contribute to the resistance minima in concentrated $Ni-Cu$ alloys.

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