Observation of palladium clusters in the alloy $Au_{0.6}Pd_{0.4}$

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It is known that Ag-Pd, Au-Pd and Cu-Ni of nearly equiatomic composition have a minimum in the temperature dependence of the electric resistance at low temperatures.

For alloys of the Cu-Ni system, the observed anomalies are directly attributed to the presence of nickel clusters.[1] Analogous anomalies of the function $R=f(T)$ in the Au-Pd system are attributed by the authors of[2] to "spin packets" due to a special state of palladium. On the other hand, the method of diffuse x-ray scattering has revealed a short-range order in the $Au_{0.6}Pd_{0.4}$ alloy.[3] It can be assumed that palladium clusters are produced in the Au-Pd systems, in analogy with the nickel clusters in the Cu-Ni system.

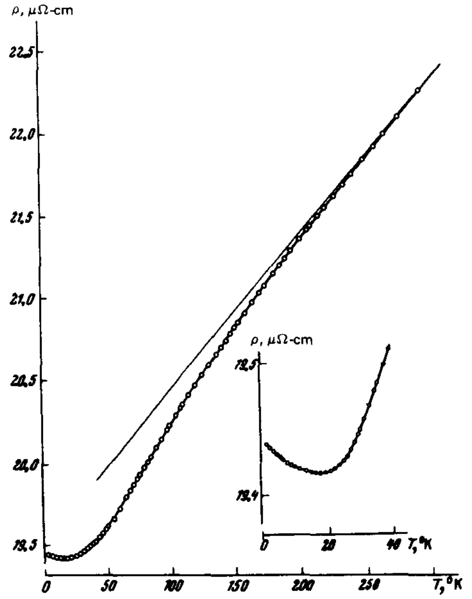

FIG. 1. Temperature dependence of the resistivity of the alloy $Au_{0.6}Pd_{0.4}$.

In view of their small dimensions and lack of distinct boundaries, the observation of clusters calls for special procedures, such as NMR, NGR, diffuse x-ray scattering and neutron scattering, or investigations with an electron microscope. In individual cases, however, it is apparently possible to observe the clusters by placing in them characteristic impurity atoms, the monitoring of which by simple physical methods makes it possible to investigate at least their immediate environment.

The present study was undertaken to check the aforementioned assumption concerning the presence of palladium clusters in the alloy $Au_{0.6}Pd_{0.4}$. We used for this purpose the ability of iron and cobalt atoms to polarize the immediate environment in Pd-Fe and Pd-Co alloys. The "giant" magnetic moments (10—12 μ_B) observed in these alloys are uniquely related to iron-palladium and cobalt-palladium spin clusters having, according to,[4] a diameter 10 Å.

We have assumed (from a comparison of the solubility of iron and cobalt in palladium and in a matrix enriched with gold[5]) that when iron or cobalt is diluted in the alloy $Au_{0.6}Pd_{0.4}$ the probability of the impurity falling into the palladium clusters is large. Owing to the polarization of the immediate environment, "giant" magnetic moments will be connected with the impurity atoms, and their presence can be easily revealed by their magnetic and electric properties.

We have investigated the temperature dependences of the magnetic susceptibility and of the resistivity of the alloys $Au_{0.6}Pd_{0.4}$, $(Au_{0.6}Pd_{0.4})Fe_{0.01}$, $(Au_{0.6}Pd_{0.4})Fe_{0.005}$, and $(Au_{0.6}Pd_{0.4})Co_{0.005}$. Metallographic and x-ray structure investigations have shown the samples annealed from 800 °C to be single-phase. The samples were wires of 0.1 mm diameter. The specific values of the magnetic suseptibility and of the resistivity of the $Au_{0.6}Pd_{0.4}$ alloy at room temperature did not differ from the published values.[6]

A minimum was observed in the temperature dependence of the resistivity of the $Au_{0.6}Pd_{0.4}$ alloy at $T = 20$ °K. This dependence has an anomalous form also at higher temperatures (Fig. 1). The peculiarities of the temperature dependence of the resistivity are due, according to,[2] to "spin packets" bound to the palladium atoms, and in accordance with the terminology of[1] to clusters of palladium atoms.

As seen from Fig. 2, addition of 1 at.% iron to the alloy increases strongly the growth of the paramagnetic susceptibility with increasing temperature, and also alters greatly the temperature coefficient of the resistivity (by a factor 18 at $T = 80$ °K).

The strong growth of the magnetic susceptibility with decreasing temperature, the satisfaction of the Curie-Weiss law for the reciprocal susceptibility starting with $\Theta_p \approx 40$ °K, and the abrupt change in the temperature coefficient of resistivity at $\Theta_c = 35$ °K are observed also for the alloys $(Au_{0.6}Pd_{0.4})Fe_{0.005}$ and $(Au_{0.6}Pd_{0.4})Co_{0.005}$. In these alloys, the average magnetic moment per impurity atom, calculated from the magnetic susceptibility, is 10 μ_B.

On the basis of the results and of a comparison of the magnetic moments of the impurity atoms and of the ordering temperatures of the considered alloys with the corresponding values for the Pd-Fe and Pd-Co solutions tions[7] we can draw the following conclusions: 1) The $Au_{0.6}Pd_{0.4}$ alloy contains small regions (clusters) that are highly enriched with palladium. 2) Iron and cobalt impurities are apparently predominantly distributed in these regions. 3) It can be assumed that the dimensions of the observed clusters are close to the dimension of the spin clusters in the Pd-Fe and Pd-Co cluster, and amount, according to[4] to 10 Å.

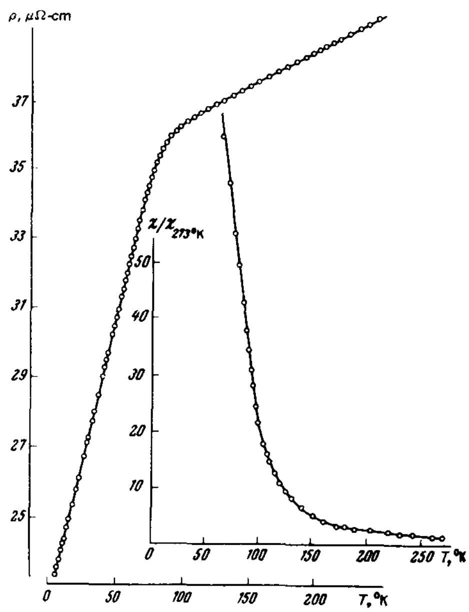

FIG. 2. Temperature dependence of the resistivity and of the relative magentic susceptibility of the alloy $(Au_{0.6}Pd_{0.4})Fe_{0.01}$.

[1]R.W. Hought, M.P. Sarachik, and I.S. Kouvel, Solid State Comm. 8, 1943 (1970).

[2]L.R. Edwards, C.W. Chen, and S. Levgold, ibid. 8, 1403 (1970).

[3]W. Copeland and M.E. Nicholson, Acta Metallurgica 12, 321 (1964).

[4]G. Low, Proc. Internat. Conf. on Magnetism. Nottingham, 1964, p. 133.

[5]E.M. Savitskii, V.P. Polyakova, and M.A. Tylkina, Splavy palladia (Palladium Alloys), 1967.

[6]H.L. Logie, I. Jackson, I.C. Anderson, and F.R.N. Nabarro, Acta Metallurgica 9, 707 (1961).

[7]R.M. Bosorth, P A. Wolf, *et al.*, Phys. Rev. 122, 1157 (1961); B. Matthias, M. Peter, H. Williams, A. Clogston, E. Corenzwit, and R. Sherwood, Phys. Rev. Lett. 5, 542 (1960); Phys. Rev. 125, 541 (1962).