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Virtual-Bound-State Formation in Pd-U Alloys*

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Measurements have been made from below 2°K to 300°K of the electrical resistivity and magnetic susceptibility of Pd-U alloys. Low-temperature specific heats have also been measured for Pd-U and Pd-Th alloys. At low actinide compositions, the electronic specific-heat coefficient and the magnetic susceptibility decrease with concentration and are the same as those of Pd-Ag versus electron/atom ratio, provided valences of 1, 4, and 6 are used for silver, thorium, and uranium, respectively. However, at compositions above 7.5 at. % uranium, the electronic specific-heat coefficient and susceptibility rise quickly with composition, and minima are found in the resistivity-temperature curves. The susceptibility indicates no localized-moment behavior. The results are interpreted by the formation at the Fermi level of uranium-5f virtual bound-state levels, whose width decreases with uranium concentration. The specific-heat data give evidence for a term due to interactions between conduction electrons and spin fluctuations. The negative resistivity-temperature derivatives are attributed to localized spin fluctuations.

I. INTRODUCTION

The actinide metals neptunium[1] and plutonium[2] have recently been observed to behave as magnetic impurities when dissolved in palladium. Alloys containing 250–1000 at. ppm neptunium in palladium have resistivity minima in the temperature range 7–12°K. In addition, $\Delta\rho/c$ is composition independent, and, below 4°K, $\Delta\rho$ for these alloys can be fitted to the expression $\Delta\rho=\Delta\rho(0)(1-T^2/T_K{}^2)$, where $\Delta\rho$ is the excess resistivity of the alloy over the pure palladium, c is the neptunium concentration, $\Delta\rho(0)$ is $\Delta\rho$ at $T=0$°K, and T_K is taken as a measure of the Kondo temperature.[3,4] The results are $T_K=26$°K and $\Delta\rho(0)/c=7.5$ $\mu\Omega$ cm/% Np. The electrical resistivities of Pd-Pu alloys decrease monotonically with decreasing temperature down to 1.3°K. The magnetic susceptibilities of alloys containing 0.5–2.0% neptunium or plutonium can be fitted to the expression $\chi=\chi_0+C/(T-\theta)$ at low temperatures. The θ values obtained are near -3 and -13°K for the plutonium and neptunium alloys, respectively. The value of $-\theta$ is often taken as another measure of the Kondo temperature.[5] Thus, the estimate for Pd-Np is $13<T_K<26$°K. The positive resistivity-temperature slope and negative θ value of Pd-Pu are properties analogous to those of Rh-Fe[6,7] and Ir-Fe.[7,8]

Bates and Leach previously measured the magnetic susceptibilities of alloys and intermetallic compounds containing up to 25 at. % uranium in palladium over the temperature range 77–293°K.[9] The room-temperature susceptibilities of Pd-U are plotted versus electron/atom ratio in Fig. 1. For purposes of comparison the Pd-Ag data of Hoare *et al.* are also shown.[10] The electron/atom ratio is Zx, where Z is the effective valence of the solute atom and x is the fractional composition of the solute. Z has been taken to be 6 and 1 for uranium and silver, respectively. Bates and Leach found a minimum in the room-temperature susceptibility versus electron/atom ratio at a concentration corresponding to about 8% uranium. In addition, the susceptibilities of samples containing greater than 12% uranium obeyed a Curie-Weiss law $\chi=C/(T-\theta)$ with $\theta\ll 0$. To explain these results they proposed the filling of a palladiumlike d band for low uranium concentrations and that at higher uranium concentrations, the valency of the uranium changes from $+6$ to $+4$, leaving two unpaired electrons to form localized magnetic moments on the uranium atoms. Bates and Unstead measured the susceptibilities of Pd-Th alloys and found that the Pd-Th data lie on the same curve as the Pd-Ag results if an effective valence of 4 is taken for thorium.[11] The magnetic proper-

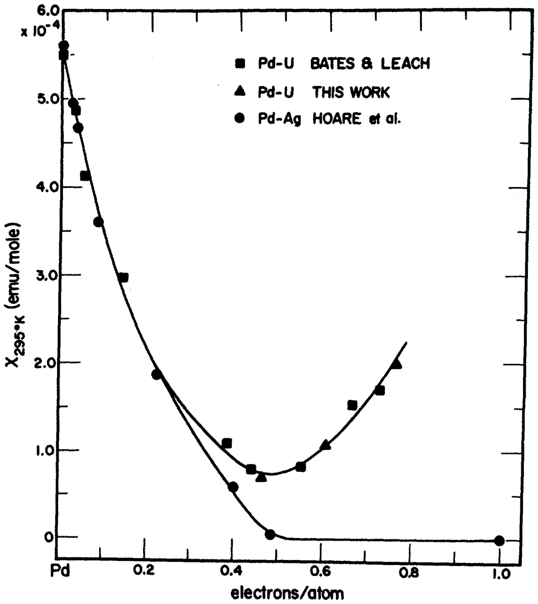

FIG. 1. Magnetic susceptibilities at room temperature of Pd-U and Pd-Ag alloys plotted versus the electron/atom ratio, defined as the effective valence of the solute times the fractional solute composition. Effective valences of 6 and 1 were taken for uranium and silver, respectively.

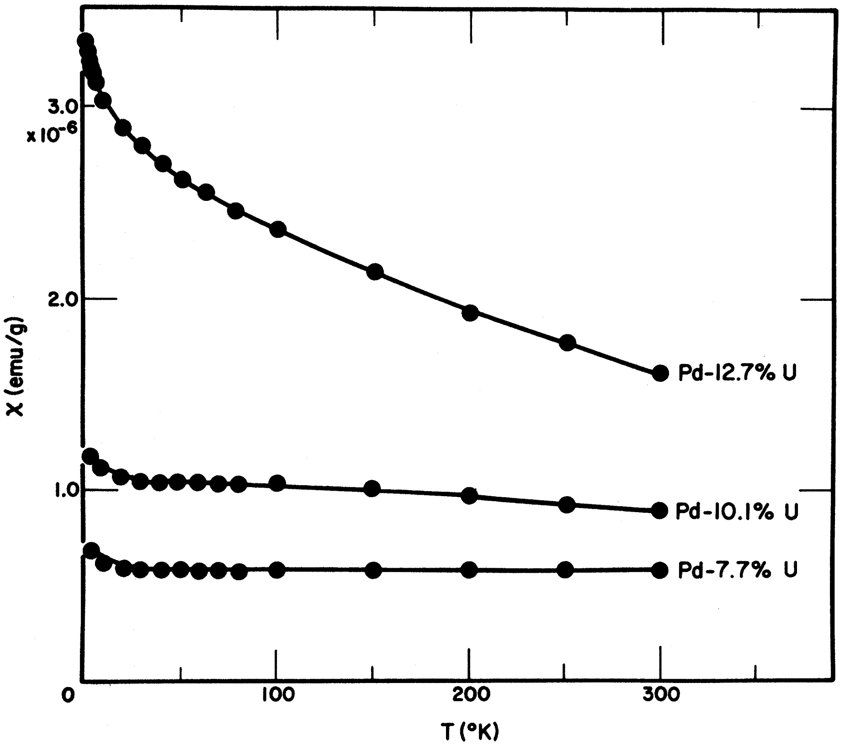

FIG. 2. Magnetic susceptibility of Pd-U alloys in the temperature range 1.8–300°K.

ties of the continuous series of solid solutions $U_xTh_{1-x}Pd_3$ ($0\leq x\leq 1$) were studied by Wernick *et al.* in the range 1.4–300°K.[12] They found Curie-Weiss behavior in the high-temperature region with 3.35 μ_B/U atom at low uranium concentrations and 2.6 μ_B/U atom for UPd_3. The results were interpreted in terms of the uranium-5*f* electrons forming a localized magnetic moment for low uranium concentrations, and the formation of a very narrow 5*f* band as the uranium concentration is increased. More recently Davidov *et al.* performed EPR measurements of Gd in $U_xTh_{1-x}Pd_3$.[13] They interpreted their results as an indication of the existence of a partially compensated spin state on the uranium in $U_xTh_{1-x}Pd_3$ and suggested the existence of the Kondo effect in concentrated spin systems.

Because of the interesting magnetic properties of the actinide metals neptunium and plutonium dissolved in palladium and because of evidence for magnetism in Pd-U systems, a more extensive study of Pd-U alloys was undertaken. We wish to report measurements of low-temperature specific heat, magnetic susceptibility, and electrical resistivity of Pd-U solid solutions containing up to 13 at. % uranium. Results are also presented on a number of Pd-Th alloys which in no case have shown magnetic behavior.

II. EXPERIMENTAL PROCEDURE

The specific-heat samples were prepared at Atomic Energy Research Establishment Harwell, and they were in the form of arc-melted buttons weighing about 20 g. The details of their preparation have been discussed fully elsewhere.[14] After the specific heats had been determined, a 4-mm cube was cut from three of the samples in order to make susceptibility measurements. The electrical resistivity samples were prepared at Argonne National Laboratory, the arc-melted buttons being rolled into sheet and machined to a final sample size of about 10×1.5×0.1 mm. This treatment was followed by an anneal at 950°C for 18 h and an oil quench prior to measurement. All the specimens discussed in this paper were in the fcc δ phase, and for alloys containing more than 8% uranium this phase had to be retained by quenching. According to Pells,[14] the δ phase can be retained in samples containing up to 15 at. % uranium, but we have found subsequently that this is reliably true only up to 13% uranium. Metallographic examination showed that specific-heat samples containing up to 12.7% uranium were free from inclusions of the second-phase U_2Pd_{17}. Similarly, the resistivity samples gave room-temperature resistivities which were in good agreement with previous values for single-phase alloys, and as Pells shows this is a sensitive test for the presence of a second phase.[14] Bates and Unstead[11] claim that 20% of thorium can be dissolved in fcc Pd, and our Pd-Th samples were well inside this limit.

The specific heat[15] and magnetic susceptibility[16] apparatus are described elsewhere. The electrical resistivity was measured in two manners. Data runs were made on all samples with an automated system described previously.[17] With this method, the temperature is increased

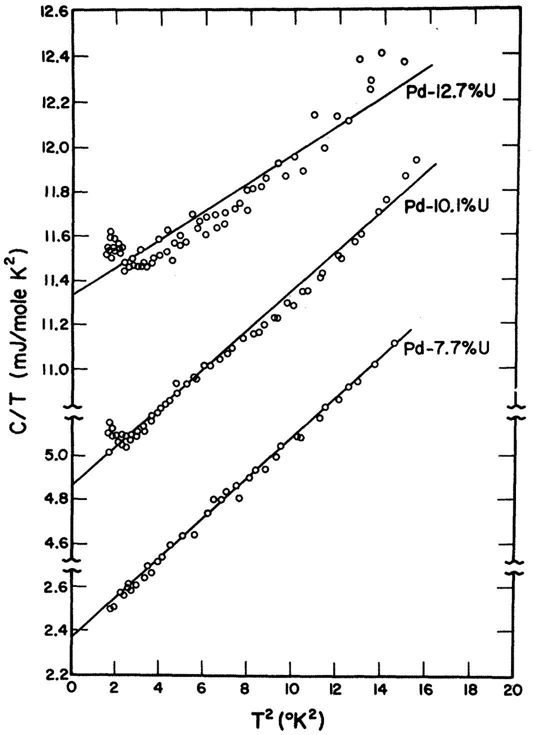

FIG. 3. Low-temperature specific heats of alloys containing 7.7, 10.1, and 12.7 at. % uranium.

continuously at a rate of $\frac{1}{3}$°K/min, and data are automatically recorded every 20 or 48 sec, depending on the temperature range. The low-temperature resistivities of alloys containing 7, 8, 9, and 11% uranium were also measured at constant temperature using a Honeywell six-dial potentiometer. Both types of runs gave the same results; e.g., the temperature and depth of the minimum of the Pd–8%-U resistivity is the same using either method. Absolute error in the resistivity is estimated to be 3%.

TABLE I. Specific-heat data on Pd-U and Pd-Th alloys.

Specimen (at. %)	γ (mJ mole^{-1} °K^{-2})	θ (°K)
Pd	9.43±0.02	272±2
Pd–1.0%-U	8.22	277
Pd–3.0%-U	6.03	272
Pd–5.0%-U	4.28	265
Pd–7.7%-U	2.36	277
Pd–10.1%-U	4.86	280
Pd–12.7%-U	11.3	313
Pd–3.9%-Th	6.71	258
Pd–7.8%-Th	3.88	246
Pd–11.8%-Th	1.36	313
Pd–14.7%-Th	0.99	302

III. RESULTS

A. Magnetic Susceptibility

The magnetic susceptibilities of alloys containing 7.7, 10.1, and 12.7% uranium in palladium were measured over the temperature range 1.8–300°K and are shown in Fig. 2. These results are in good agreement with those obtained by Bates and Leach in the range 77–293°K,[9] and the extension of susceptibility measurements to low temperatures for these alloy compositions shows no pronounced changes in the magnetic properties below 77°K. The susceptibilities of the 7.7 and 10.1% uranium alloys are almost independent of temperature, which suggests that no localized moments are forming on the solute atoms. Pd–12.7%-U has a considerably higher susceptibility with a strong negative temperature derivative, and this increases continuously down to the lowest temperature of measurement.

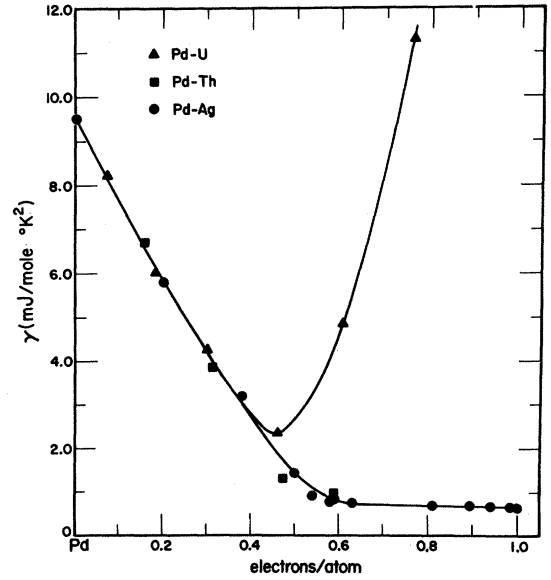

FIG. 4. Specific-heat γ values of Pd-U, Pd-Th, and Pd-Ag alloys plotted versus the electron/atom ratio, defined as the effective valence of the solute times the fractional solute composition. Effective valences of 6, 4, and 1 were taken for uranium, thorium, and silver, respectively.

B. Specific Heat

The specific heats of six alloys containing 1.0–12.7% uranium in palladium and of four alloys containing 3.9–14.7% thorium in palladium were measured from 1.3 to 4°K. Figure 3 shows the results for the three most concentrated uranium alloys, plotted in the usual way as C/T versus T^2. Pd–7.7%-U gives a good straight line, but Pd–10.1%-U and Pd–12.7%-U show a small but distinct upward curvature at lowest temperatures. All the other samples were accurately linear like Pd–7.7%-U. The derived values of γ and θ are listed in Table I. Clearly the values given for Pd–10.1%-U and Pd–12.7%-U are to be treated with some reservation

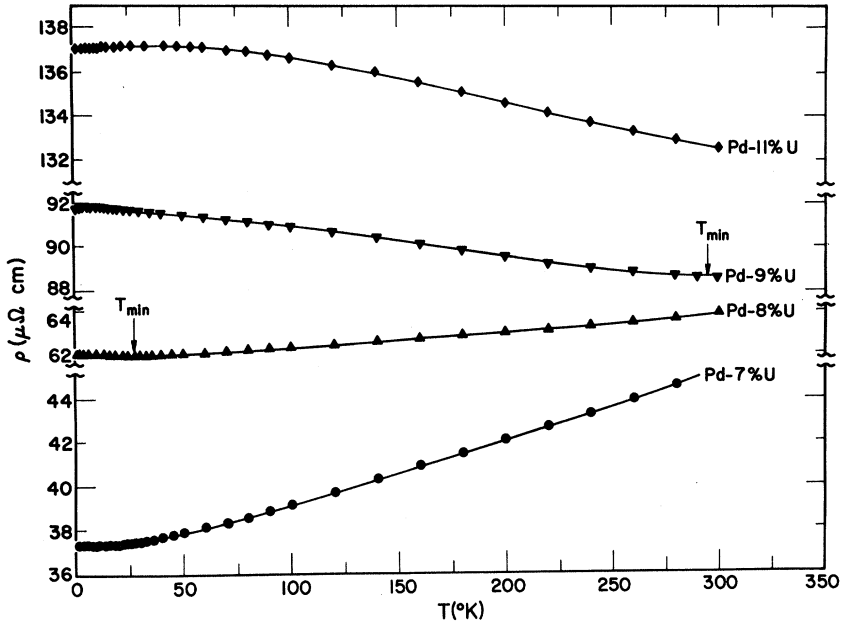

FIG. 5. Electrical resistivity of Pd-U alloys in the temperature range 1.3–300°K.

because of the curvature in the data. The data at lowest temperatures have too much scatter to determine the exact temperature dependence.

In Fig. 4 the electronic coefficient γ is plotted for all the alloys versus the electron/atom ratio Zx, the previous data for Pd–Ag being included for comparison.[15] Z has been taken to be 6, 4, and 1, for uranium, thorium, and silver, respectively. When presented in this way the Pd-Th curve agrees excellently with that for Pd-Ag, and the same is true for the Pd-U system in the concentration range below 7% uranium. At higher uranium concentrations, γ rises steeply. The composition for the minimum in γ is nearly the same as that at which the room-temperature susceptibilities of the Pd-U alloys have a minimum. Thus, for both Pd-Th and Pd-U the behavior of the electronic specific heat is qualitatively like that of the susceptibility.[9,11]

C. Electrical Resistivity

The electrical resistivities of palladium alloys containing up to 13 at. % uranium were measured over the range 1.3–300°K. The data for alloys containing 7, 8, 9, and 11% uranium are shown in Fig. 5. Alloys containing 7% uranium or less show no anomalies in resistivity versus temperature. However, between 7 and 8% uranium there is a marked change in the behavior of this alloy system. The Pd–8%-U alloy has a resistivity minimum 0.07 μΩ cm in depth at 25°K. The addition of only 1% more uranium increases the temperature of the minimum to room temperature (for the Pd–9%-U alloy). Further additions of uranium produce resistivity maxima near 50°K and negative resistivity-temperature slopes at 300°K for both the 11 and 13% uranium alloys.

The values of the resistivity at 4.2 and 300°K are plotted versus uranium concentration in Fig. 6. Figure 6 indicates an abrupt change in the composition dependence of resistivity at about 7.5% uranium. This abrupt change in the slope of resistivity versus composition correlates well with the temperature dependences of alloys in this composition region; i.e., no resistivity minimum is observed for the Pd–7%-U alloy, while the Pd–8%-U alloy has a minimum at 25°K. The composition at this slope change also correlates well with the minima in susceptibility and specific heat γ near 7.5% uranium.

IV. DISCUSSION

These results suggest the following model for the Pd-U system. For low uranium concentrations the decrease in magnetic susceptibility and specific heat with uranium composition is due to the filling of a palladium-like *d* band. Also, as uranium is added, broad uranium-5*f* Friedel-Anderson virtual-bound-state levels of spin up and spin down are formed at the Fermi level. These levels narrow with increasing uranium concentration and cause the increase in magnetic susceptibility and specific heat at higher uranium concentrations. The spin-up and spin-down virtual-bound-state levels are populated equally, and thus no static magnetic moment is formed. However, spin fluctuations can occur between these two virtual levels and form apparent moments with lifetimes sufficiently long to spin-flip scatter the conduction electrons and cause resistivity minima. This model agrees with the proposal of Bates and Leach[9] that band filling determines the properties of this system at low uranium concentrations, and the model extrapolates well to the proposal of Wernick *et al.*[12] that a very narrow uranium-5*f* band exists in UPd_3. However, it is in disagreement with the suggestion of Bates

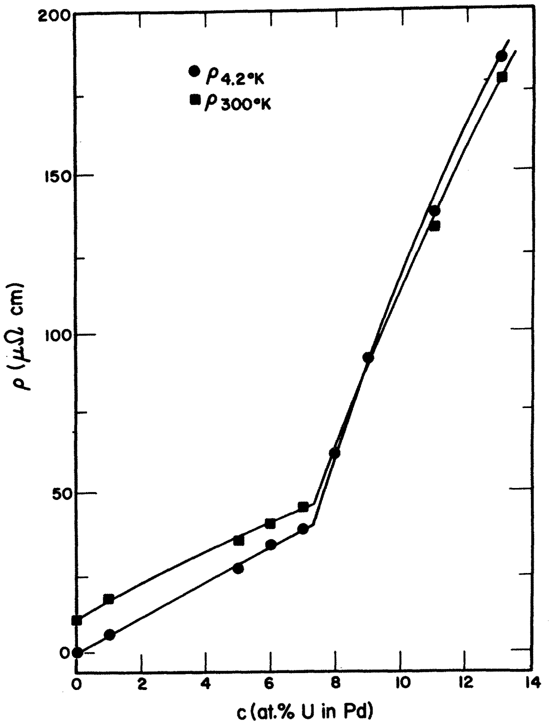

FIG. 6. Electrical resistivity at 4.2 and 300°K of Pd-U alloys plotted versus uranium composition.

and Leach that localized moments are formed on the uranium atoms for concentrations above 12% uranium.

A. Magnetic Susceptibility and Specific Heats

On the basis of the model proposed here, the magnetic susceptibility and specific heat are dependent on two competing processes. There is the band-filling process which decreases the values of these properties with increasing uranium composition, and there is the formation of uranium-5*f* virtual bound states which cause these properties to increase with uranium content. The fact that the magnetic susceptibility and specific heat decrease as uranium, thorium, and silver are added to palladium has in all cases, including this one, been attributed to the filling of a palladiumlike density of states. However, two qualifications of this idea must be made. First, Montgomery *et al.* have discussed Pd-Ag in detail and have pointed out that it is extremely unlikely that it is a rigid palladium density of states that is filled.[15] Second, the spatial extent of the band-filling process must also be considered. Although the scaling with the electron/atom ratio of the Pd-U and Pd-Ag susceptibility and specific heat for low uranium concentrations seems to imply that one uranium atom acts as six silver atoms by contributing six *s* electrons to the *d*-like average density of states of the alloy, such an idea is not plausible because of the complicated *s*-*d*-*f* electronic structure of uranium. A more realistic approach is a localized density-of-states model. Recent theories of Soven[18] on disordered substitutional alloys have indicated that the density of states localized about a particular type of atom can be very different from the average density of states of the material. High-valence impurities like uranium and thorium dissolved in palladium, which has a substantial number of *d*-band holes, can be expected to produce extreme variations in the localized density of states. However, the explanation of why Pd-U scales so well with Pd-Ag when an effective valence of 6 is taken for uranium has yet to be found, and it cannot be found by the measurement of macroscopic properties.

For uranium concentrations above 8%, the narrowing of the virtual-bound-state levels is the dominant effect in Pd-U. Wang *et al.* have recently developed a theory of itinerant ferromagnets above the Curie point.[19] They concentrated on the paramagnetic phase and neglected cooperative effects. This theory can qualitatively explain some features of the Pd-U data. They assumed an Anderson model[20] of an impurity in an effective band. The following expression was then developed for the static magnetic susceptibility:

$$\chi = (\mu_B^2/U)[2\pi\langle\xi_0^2\rangle - 1] + \chi_{\text{band}}, \qquad (1)$$

where μ_B is the Bohr magneton, U is the intra-atomic Coulomb interaction, and $\langle\xi_0^2\rangle$ is a function dependent on temperature, on U, and on the virtual-level width Γ. This theory gives a smooth interpolation between the correct, weakly exchange-enhanced Pauli susceptibility, $2\mu_B^2/\pi\Gamma$, for small $U/\pi\Gamma$, and the correct Curie law for a free spin $\frac{1}{2}$ when $U/\pi\Gamma$ is large. The region $U/\pi\Gamma \sim 1$ is characterized by spin fluctuations. Assuming that interactions between uranium atoms will not significantly alter the qualitative features of the model and assuming the Coulomb interaction U is relatively independent of composition, this model predicts an increase in both χ and in $-d\chi/dT$ as Γ decreases with increasing uranium content. Also, under the assumption that the weak temperature dependences of the susceptibilities of the 7.7 and 10.1% uranium alloys imply that $U/\pi\Gamma$ is small, Γ can be roughly estimated from the room-temperature susceptibilities and the relation $\chi = 2\mu_B^2/\pi\Gamma$. The results are $\Gamma = 0.26$ and 0.21 eV, respectively. The relation $U/\pi\Gamma < 1$ gives an upper bound on U of 0.8 eV. Taking into account possible large exchange enhancements and the fact that U is really not zero would raise these estimates of U and Γ. However, this approach does give an order-of-magnitude estimate of U and it is quite small. It suggests that the 5*f* electrons of a uranium atom can approach sufficiently close to one another so that the Pauli exclusion principle orients their spins in opposite directions. The result is that the spin-up and spin-down virtual levels are essentially equally populated for this range of uranium composition.

Bates and Leach fitted their results for alloys containing greater than 12% uranium to a Curie-Weiss law. The data for the 12.7% uranium alloy presented here can be fitted to within 1% by $\chi = \chi_0 + C/(T-\theta)$ in the range 30–300°K, with $\chi_0 = 0.14 \times 10^{-6}$ emu/g,

$\theta = -329°K$, and C corresponding to an effective moment of 2.7 μ_B/U atom. Below 30°K the fit underestimates the data (by 6% at 4°K). However, the validity of fitting the data to a Curie-Weiss law is very questionable. Consider the susceptibility of the alloys containing greater than 8% uranium. A reasonable estimate for the band contribution for one of these alloys is the susceptibility of a Pd-Ag alloy with a corresponding electron/atom ratio. Figures 1 and 4 show that the band contribution is small in this concentration range. Thus, essentially all of the temperature-independent part of the susceptibility is due to the width of the virtual level. As the virtual level narrows the temperature-independent contribution should increase. Thus, χ_0 for the 12.7% uranium alloy should be larger than the susceptibility of the 10.1% uranium alloy which is essentially temperature independent. But $\chi_0 = 0.14 \times 10^{-6}$ emu/g for the Pd–12.7%-U alloy is much smaller than the total susceptibility of the 10.1% uranium sample (0.9×10^{-6} emu/g). In addition, the usual method used to obtain the effective magnetic moment per impurity atom for systems having resistivity minima is to extract this parameter from Curie-Weiss fits to data taken well above $|\theta|$,[5] but all of the Pd-U data thus far are for temperatures below $|\theta|$. Thus, high-temperature data are needed before any significance can be attached to parameters obtained from Curie-Weiss fits.

The upturn at lowest temperature in C/T versus T^2 as seen in Fig. 3 is attributed to the conduction-electron–spin-fluctuation interaction. Doniach and Engelsberg[21] and Bennemann[22] proposed that the contribution to the specific heat due to such an interaction is $C/T \sim T^2 \ln T$ in the paramagnetic phase. Because the upturn is small, and because of the scatter in the specific-heat data for the 10.1 and 12.7% uranium alloys, no attempt was made to fit the data. (This aspect of the specific heat will be discussed again in Sec. IV C.)

B. Electrical Resistivity

The alloys containing 8 and 9% uranium have resistivity minima. The susceptibilities of these alloys are essentially temperature independent over the whole temperature range and thus no localized moments are formed. This fact rules out the Kondo effect[23] or Nagaoka-type spin compensation[3] as an explanation for the resistivity minima. These theories are based on *s-d* exchange scattering of conduction electrons by localized magnetic moments. The resistivity of Pd-U is better understood in terms of localized spin fluctuations between the spin-up and spin-down uranium-5*f* virtual levels. The concept of localized spin fluctuations has been discussed by a number of authors.[4,19,24] Caplin and Rizzuto used the concept to explain resistivity minima in Al(Mn) and Al(Cr).[24] They suggested that if the lifetimes of localized spin fluctuations at an impurity atom are long enough, a temporary magnetic moment may have sufficient time to equilibrate. This temporary moment could flip the spin of a conduction electron and contribute a logarithmic term to the resistivity. With the evidence against localized magnetic moments and in view of the susceptibility and specific-heat evidence for the formation of virtual levels, the localized spin-fluctuation model is the most plausible explanation for the resistivity minima.

One other aspect of the resistivity can be discussed. Above the composition of 8% uranium the resistivity at a given temperature increases much more rapidly with composition than it does for low uranium concentrations. This is also consistent with the formation of virtual bound states at the Fermi level. The virtual level increases the density of states at the Fermi level. Mott and Jones pointed out that the transition probability from one state to another is proportional to the density of states in the final state.[25] The increase in the density of states at the Fermi level means that there are more states into which conduction electrons can be scattered and thus the resistivity increases.

The question of why the electrical resistivity shows dramatic variation with uranium concentration near 7.5% uranium is an intriguing one. A possible explanation is that near this composition there are a sufficient number of uranium atoms so that some of these atoms have one or two uranium nearest neighbors.[26] In these "uranium-rich" regions there is a large localized density of states of uranium-5*f* and possibly uranium-6*d* electrons giving rise to localized exchange enhancements. Such effects are most probably occurring, but the narrow composition range where susceptibility and specific heat start to increase and at which resistivity minima occur suggests a more likely explanation. At the electron/atom ratio corresponding to 7.5% uranium the susceptibility of Pd-Ag alloys is fairly small and changes little for increasing silver content. The same general features are true of the specific heat of Pd-Ag. These results have been interpreted to mean that the number of holes in the palladiumlike density of states of the alloy is small and that this density of states is essentially filled.[15] This band filling is localized in the region of the solute atoms. Beyond 7.5% uranium the palladiumlike density of states is filled and the uranium-5*f* electrons no longer hybridize with the palladium-*d* electrons. The uranium-5*f* electrons are more localized and the virtual levels narrow appreciably.

In this model, uranium atoms in regions where the localized palladiumlike density of states is filled make the largest contributions to properties depending on virtual levels. At a composition near 7.5% uranium, most uranium electrons are filling the palladiumlike density of states and all the uranium atoms are not acting as "magnetic" impurities. However, as slightly more uranium is added, the band-filling process is completed throughout the alloy, and the number of magnetic uranium impurities approaches the actual chemical composition. An alloy containing 8% magnetic impurities would ordinarily be considered nondilute and could have resistivity maxima as well as minima, due to cooperative effects. (For example, Pd–8%-Np has both a resistivity maximum and a minimum.) But the resistivity of the Pd–8%-U alloy resembles that of a dilute alloy. The rapid variation of the depth and the temperature of the resistivity minimum between 8 and 9% uranium (the depth of the resistivity minimum is only 0.07 $\mu\Omega$ cm in Pd–8%-U but is 3.3 $\mu\Omega$ cm in Pd–9%-U) is also more easily understood if the effective concentration of magnetic uranium impurities has increased by much more than the one percent change out of eight in uranium content.

C. Related Systems

The properties of the Pd-U system are very similar to Cu-Ni and V-Fe. In the former system, the addition of nickel to copper causes the magnitude and negative temperature slope of the susceptibility to increase.[27] The specific heat γ also increases with nickel content,[28,29] and in the paramagnetic alloys there are minima in the resistivity-temperature curves.[30] V-Fe is even more similar to Pd-U, as small additions of iron cause an initial decrease in susceptibility[31] and γ [32] owing to *d*-band filling. Alloys containing more than 20% iron begin to show the negative slope in the susceptibility-temperature curves, and susceptibility and γ increase with solute contents as in Pd-U and Cu-Ni (Figure 2 of Ref. 31 is extremely similar to Fig. 2 here.) An additional term contributes to the specific heat in both of those systems, also, in paramagnetic alloys.[28,32]

Despite the similarities there is an important difference between Pd-U and the other two systems. The magnetism in Cu-Ni and V-Fe ranges from paramagnetism to superparamagnetism to ferromagnetism as the magnetic solute is added. No ferromagnetism has been observed in Pd-U and this is reasonable since pure uranium has no measurable magnetic moment,[16] unlike iron or nickel. The absence of ferromagnetism has an important bearing on the interpretation of the specific-heat data. Two models attempt to explain upturns at low temperatures in C/T versus T^2 for magnetic systems. One model is based on ferromagnetic clusters and predicts $C/T \sim T^{-1}$. The specific heat of both Cu-Ni and V-Fe have been interpreted on this basis.[28,29] The other model considers the interaction between conduction electrons and spin fluctuations and predicts $C/T \sim T^2 \ln T$. Rh-Ni [33] and Cu-Ni [22] have been explained from this viewpoint. Because of the absence of ferromagnetism, the spin-fluctuation model is the one more appropriate to the specific heat of Pd-U.

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[1] W. J. Nellis and M. B. Brodsky, J. Appl. Phys. **41**, 1007 (1970).

[2] W. J. Nellis and M. B. Brodsky, Phys. Letters **32A**, 267 (1970).

[3] Y. Nagaoka, Phys. Rev. **138**, A1112 (1965).

[4] N. Rivier and M. J. Zuckermann, Phys. Rev. Letters **21**, 904 (1968).

[5] M. D. Daybell and W. A. Steyert, Rev. Mod. Phys. **40**, 380 (1968).

[6] B. R. Coles, J. H. Waszink, and J. Loram, in *Proceedings of the International Conference on Magnetism, Notingham, 1964* (The Institute of Physics and The Physical Society, London, 1964), p. 165ff.

[7] G. S. Knapp, J. Appl. Phys. **38**, 1267 (1967).

[8] M. P. Sarachik, Phys. Rev. **170**, 679 (1968).

[9] L. F. Bates and S. J. Leach, Proc. Phys. Soc. (London) **B69**, 997 (1956).

[10] F. E. Hoare, J. C. Matthews, and J. C. Walling, Proc. Roy. Soc. (London) **A216**, 502 (1953).

[11] L. F. Bates and P. B. Unstead, in *Proceedings of the International Conference on Magnetism, Nottingham, 1964* (The Institute of Physics and the Physical Society, London, 1964), p. 188ff.

[12] J. H. Wernick, H. J. Williams, D. Shaltiel, and R. C. Sherwood, J. Appl. Phys. **36**, 982 (1965).

[13] D. Davidov, H. Lotem, D. Shaltiel, M. Weger, and J. H. Wernick, Phys. Rev. **184**, 481 (1969).

[14] G. P. Pells, J. Inst. Metals **92**, 416 (1963-64).

[15] H. Montgomery, G. P. Pells, and E. M. Wary, Proc. Roy. Soc. (London) **A301**, 261 (1967).

[16] J. W. Ross and D. J. Lam, Phys. Rev. **165**, 617 (1968).

[17] M. B. Brodsky, N. J. Griffin, and M. D. Odie, J. Appl. Phys. **40**, 895 (1969).

[18] P. Soven, Phys. Rev. **178**, 1136 (1969).

[19] S. Q. Wang, W. E. Evenson, and J. R. Schrieffer, Phys. Rev. Letters **23**, 92 (1969); W. E. Evenson, J. R. Schrieffer, and S. Q. Wang, J. Appl. Phys. **41**, 1199 (1970).

[20] P. W. Anderson, Phys. Rev. **124**, 41 (1961).

[21] S. Doniach and S. Engelsberg, Phys. Rev. Letters **17**, 750 (1966).

[22] K. H. Bennemann, Phys. Rev. **167**, 564 (1968).

[23] J. Kondo, Progr. Theoret. Phys. (Kyoto) **32**, 37 (1964).

[24] A. D. Caplin and C. Rizzuto, Phys. Rev. Letters **21**, 746 (1968); P. Lederer and D. L. Mills, *ibid.* **20**, 1036 (1968); M. Levine and H. Suhl, Phys. Rev. **171**, 567 (1968); D. R. Hamann, Phys. Rev. Letters **23**, 95 (1969).

[25] N. F. Mott and H. Jones, *The Theory of the Properties of Metals and Alloys* (Dover, New York, 1958), p. 267.

[26] In a Pd–7%-U alloy the probabilities are 2.7, 1.1, and 0.3% that a uranium atom will have only one, only two, or three or more uranium nearest neighbors, respectively.

[27] F. M. Ryan, E. W. Pugh, and R. Smoluchowski, Phys. Rev. **116**, 1106 (1959).

[28] K. Schröder, J. Appl. Phys. **32**, 880 (1961).

[29] C. G. Robbins, H. Claus, and P. A. Beck, J. Appl. Phys. **40**, 2269 (1969).

[30] R. W. Houghton, M. P. Sarachik, and J. S. Kouvel, Solid State Commun. **8**, 943 (1970).

[31] D. J. Lam, D. O. Van Ostenburg, M. V. Nevitt, H. D. Trapp, and D. W. Pracht, Phys. Rev. **131**, 1428 (1963).

[32] C. H. Cheng, C. T. Wei, and P. A. Beck, Phys. Rev. **120**, 426 (1960).

[33] E. Bucher, W. F. Brinkman, J. P. Maita, and H. J. Williams, Phys. Rev. Letters **18**, 1125 (1967); W. F. Brinkman, E. Bucher, H. J. Williams, and J. P. Maita, J. Appl. Phys. **39**, 547 (1968).