# 5f Magnetism in Palladium-Actinide Solid Solutions\*

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The electrical resistivities of palladium alloys containing 2.0 at.% or less neptunium or plutonium have been measured in the range 1.3–300 °K. The magnetic susceptibilities of these alloys have also been measured in the range 2.5–300 °K. Resistivity minima are observed for the PdNp alloys, but the resistivities of the PdPu alloys have positive slopes at low temperatures. The magnetic susceptibilities of the alloys can be fitted to  $\chi = \chi_0 + C/(T-\Theta)$  at low temperatures. For PdNp,  $\Theta \simeq -13$  °K and  $\mu_{\rm eff} = 2.0 \mu_{\rm B}/{\rm Np}$  atom. For PdPu,  $\Theta \simeq -3$  °K and  $\mu_{\rm eff}$  increases from 0.55 to 1.3  $\mu_{\rm B}/{\rm Pu}$  atom with decreasing plutonium concentration. Apparently the 5f level is sufficiently localized to form a magnetic moment in PdNp, and is spin compensated at low temperatures. The Kondo temperature is estimated to be  $T_K \simeq 15$  °K. The PdPu system is discussed in terms of localized spin fluctuations of the 5f electrons, with a spin-fluctuation temperature  $T_S \simeq 1$  °K. The PdU system was discussed earlier. It is suggested that in these alloys, indirect interactions between uranium impurities via the palladium conduction electrons and the modification of the 4d subband in the vicinity of uranium atoms cause the 5f level to narrow and lower to the Fermi level.

#### I. INTRODUCTION

The actinide metals uranium, neptunium, and plutonium (atomic numbers 92-94) have recently been shown to have a wide variety of magnetic properties when dissolved in palladium. 1-4 Actinides have been found to act as magnetic impurities in other hosts as well. 5,6 Numerous actinide compounds order magnetically. 7 All these properties are due to the localization of actinide 5f electrons. The magnetic behavior of the actinides in nonactinide environments is in marked contrast to the properties of the pure metals. For although the paramagnetic susceptibilities of these elements are large, the susceptibilities are only weakly temperature dependent. 8-10 Most evidence indicates that none of these metals possess a magnetic moment in the pure state. 8,11,12

In this paper we report data obtained on alloys made very dilute to show the effects of noninteracting solutes. The results for PdNp are found to follow the Kondo theory. <sup>13</sup> The PdPu system, which is found to be similar to Rh Fe, <sup>14,15</sup> will be discussed in terms of a localized-spin-fluctuation model. The composition dependence of the 5f level in PdU, as observed in an earlier study, will be discussed further in terms of indirect interactions between uranium impurities via the conduction electrons.

# II. EXPERIMENTAL PROCEDURE

The alloys were prepared by arc melting suitable amounts of palladium and a host alloy containing 2.0-at.% neptunium or plutonium in an argonhelium atmosphere. A susceptibility sample of approximately 0.5 g was cut from each button, and

the remaining piece was rolled into sheet and machined to final sample size of about 10×1×0.1 mm. Resistivity samples were used in sheet form for two reasons. There was only 1 g of high-purity neptunium available for these experiments, and the rapid quenching required for the PdU alloys was easily achieved with the thin samples. The starting palladium was of 99.997% purity and contained a total of 10-ppm iron, nickel, and cobalt impurity. The neptunium and plutonium contained no detectable transition-metal impurities. All samples were annealed at 500 °C for 4 h prior to measurement. A pure palladium sample was melted, fabricated, and annealed in the same manner as the alloys and its resistivity ratio is 170. The room-temperature solubility of plutonium in palladium is about 14 at. %. 16 Metallography and bulk density measurements on samples containing up to 9% plutonium or 13% neptunium indicated that solid solutions were obtained throughout this concentration range.

The preparation of the PdNp samples on which earlier measurements were made<sup>1</sup> differed in two respects. First, there was considerable uncertainty in the geometrical factor, and, therefore, in the absolute value of the resistivity. Secondly, the samples were annealed at 900 °C for 24 h, which yielded a resistivity ratio of only 15 for a pure palladium sample. Both of these objections have been remedied in the samples discussed in this paper.

The magnetic susceptibility was measured by the Faraday method. The apparatus is similar to that described previously. The samples were encapsulated in aluminum for radiation safety purposes. At a given temperature, M/H is calculated

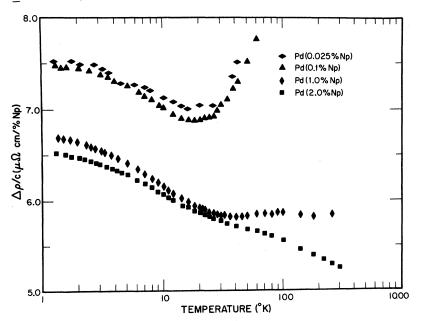


FIG. 1. Temperature dependences of  $\Delta \rho/c$  for dilute  $Pd\mathrm{Np}$  alloys.

at six magnetic fields in the range 3-14.5 kOe. The results are computer fitted by a least-squares procedure to a two-term expression linear in 1/H. The fits are quite good, with the maximum deviation of any one point being typically less than 1%.  $\chi$  is taken as M/H extrapolated to 1/H=0. The absolute accuracy of the susceptibility is about  $\pm 3\%$ .

The electrical resistivity was measured with the standard four-probe technique using an automated system described previously.<sup>3,17</sup> The extreme sensitivity of this system is indicated by the fact that dynamic resistivity runs on uranium single crystals clearly resolved two small slope changes at  $^{17}$  36 ± 1 and 42 ± 1  $^{\circ}$ K not observed previously. Absolute accuracy of the resistivity is about ± 3%.

# III. RESULTS

# A. Palladium-Neptunium

The electrical resistivities of Pd Np alloys containing 0.025-2.0 -at.% neptunium were measured in the range 1.3-300 °K. Minima in the resistivity-temperature curves are found in all cases.

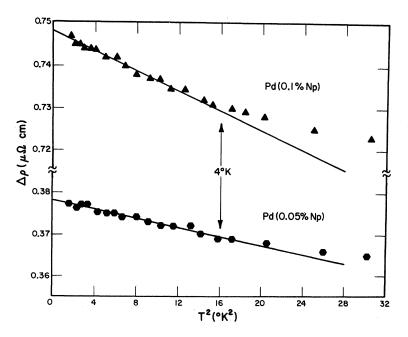


FIG. 2.  $(-T^2)$  dependences between 1.3 and  $4 \, ^{\circ}$ K of  $\Delta \rho$  for dilute PdNp alloys.

Figure 1 is a plot of  $\Delta \rho/c$  vs  $\log_{10} T$ , where  $\Delta \rho$  is the excess resistivity of an alloy over that of pure palladium and c is the atomic concentration of neptunium. For clarity the data for the 0.05% neptunium allov are not shown, but are the same as that of the 0.025% neptunium alloy below 20 °K and about 4% larger above 20 °K. The three most dilute alloys have the largest values of  $\Delta \rho/c$ , and  $\Delta \rho/c$  is the same for these compositions. Below 4 °K  $\Delta \rho$  of the 0.05 and 0.1% neptunium alloys can be fitted to  $\Delta \rho = \Delta \rho(0)(1 - T^2/T_K^2)$ , <sup>18</sup> where  $\Delta \rho(0)$ is  $\Delta \rho$  at T=0 °K and  $T_K$  is taken as a measure of the Kondo temperature. <sup>13</sup> Figure 2 is the plot of  $\Delta \rho$  vs  $T^2$ . The results are  $\Delta \rho(0)/c = 7.5 \,\mu\Omega$  cm/%Np and  $T_{\kappa} = 26$  °K for both the 0.05 and 0.1% neptunium alloys. The true Kondo temperature, however, is probably smaller than the value obtained in this fashion. The  $\Delta \rho$  used here includes not only the magnetic resistivity but also the normal impurity resistivity as well. On the basis of resistivity measurements on nonmagnetic Pd(1%Th) and Pd(1%U) alloys about half the 7.5  $\mu\Omega$ cm/%Np is due to normal impurity scattering. Thus, the above estimate of the Kondo temperature should be reduced by about  $\sqrt{\frac{1}{2}}$ , or a more realistic estimate is  $T_K \simeq 18$  °K. Assuming that  $c \simeq 50 T_K$  (in ppm) is the cutoff concentration for the single impurity region, 19 these results should yield a good estimate of  $T_K$  for single neptunium impurities in palladium.

As is evident in Fig. 1,  $\Delta \rho/c$  is smaller for the more concentrated alloys. However, all the curves lie within overlapping error bars due to an estimated 10% uncertainty in  $\Delta \rho/c$ . Thus, concentration effects are much weaker than might be expected in a palladium host.

At higher temperatures the dilute alloys have minima in  $\Delta \rho$  as well as in  $\rho$ , and the temperature derivative of  $\Delta \rho/c$  changes sign as the neptunium content is increased. Ordinarily, only a step in  $\Delta \rho \, \text{vs} \, \log_{10} T \, \text{is expected.}^{20}$  The reason for this behavior is a breakdown in Matthiessen's rule, and there are several factors responsible. First, the large mass difference between palladium and neptunium suggests that there are local distortions of the phonon spectrum near the massive neptunium atoms. Second, the large number of neptunium valence electrons and the large number of holes in the palladium d band will result in local distortion of the electronic structure of the palladium host, 21 i.e., the host d band will partially fill in the vicinity of neptunium atoms. Local distortions of these types should be largest in the most dilute alloys ( $c \lesssim 0.1\%$ ) because of the extreme departures from periodicity they produce. In the more concentrated alloys ( $c \gtrsim 0.1\%$ ) phonons generated near different neptunium atoms can interact and the band filling becomes more uniformly distributed throughout the alloy. Such effects and spin-spin interactions would serve to decrease the resistivity per impurity of the concentrated alloys relative to the dilute alloys. A third important cause of deviations from Matthiessen's rule is the energy-dependent scattering of conduction electrons from magnetic impurities. Recent theoretical work has shown that inelastic scattering can have a very significant effect on the electron-phonon, electron-electron, and electron-magnon interactions. <sup>22</sup>

Figure 3 is a plot of  $\log_{10} T_{\min} vs \log_{10} c$ , where  $T_{\min}$  is the temperature of the resistivity minimum in an alloy containing c atomic percent neptunium. According to the Kondo theory  $T_{\min} {}^{\infty} c^{1/n}$ , where the resistivity of the host varies as  $T^n$  in the region of the minima. 13 The slope of the line in Fig. 3 gives a value of n = 3.1. The resistivity of the pure palladium increases as  $T^3$  in the range 20-50 °K, but only the 1 and 2% neptunium alloys have minima in this range. Below 20 °K the palladium resistivity obeys a  $T^2$  plus a  $T^5$  dependence. Thus, it is uncertain why the data of Fig. 3 should fit a straight line with a slope of  $\frac{1}{3}$  so well. Within the limits of uncertainty, shown in Fig. 3, the three most dilute alloys yield 2.1 < n < 4.6. The depth of the minimum is proportional to c for the 0.025 and 0.05% neptunium alloys (0,006 and 0,013  $\mu\Omega$ cm, respectively), and this is also in agreement with the Kondo theory. 13 It should be noted that if  $T_{\min}$  for the 0.5, 1.0, and 2.5% neptunium alloys reported earlier are plotted in Fig. 3, those values lie about 3 °K above the line. The reason for this is unknown.

The magnetic susceptibilities in the range 2.5– $300\,^\circ$ K of pure palladium and of alloys containing 0.1–2.0-at.% neptunium are shown in Fig. 4. At high temperatures the susceptibility decreases with concentration, while at low temperatures it increases. These features are indicative of two main effects. The filling of the host d-band in the vicinity of neptunium atoms causes the band con-

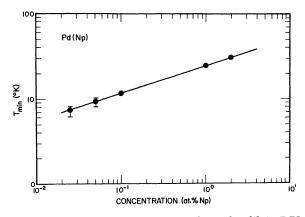


FIG. 3. Concentration dependence of  $T_{\min}$  for dilute PdNp alloys.

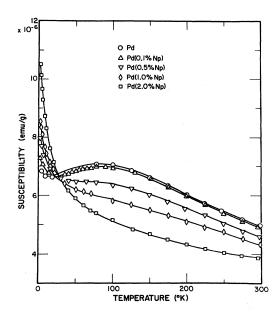


FIG. 4. Magnetic susceptibilities of dilute *PdN*p alloys in the temperature range 2.5-300 °K.

tribution to decrease with neptunium concentration, while the total localized-moment contribution increases with neptunium content. Throughout most of the temperature range ( $T \gtrsim 30\,^{\circ}\text{K}$ ) the alloy susceptibilities are less than that of the pure palladium. Thus, the palladium susceptibility cannot be subtracted from the susceptibilities of the alloys to obtain the impurity contributions. A fitting procedure was, therefore, used to analyze the data.

For the 0.5, 1.0, and 2% neptunium alloys the data in the range  $T \le 50$  °K were computer fit to a temperature-independent term plus a Curie-Weiss law:  $\chi = \chi_0 + C/(T-\theta)$ . Figure 5 is a plot of  $(\chi - \chi_0)^{-1}$  vs T. The fits are good for the 1.0 and 2.0% alloys. The data for the 0.5% alloy show departures at lowest temperatures from the fit obtained using the data taken between 12 and 40 °K. The temperature dependence of the Pd(0.1% Np) data at low temperatures is not sufficiently great to obtain a meaning-

TABLE I. Parameters obtained from computer fits of the susceptibility data to  $\chi = \chi_0 + C/(T-\Theta)$ . The Curie-Weiss constant has been converted to the effective moment per actinide atom.

Sample	(10 <sup>-6</sup> emu/g)	$\mu_{ t eff}$ $(\mu_B/{ m atom})$	⊚ (°K)
Pd(0.5%Np)	$6.1 \pm 0.2$	$2.0 \pm 0.2$	$-13$ $\pm 3.0$ $-12.3 \pm 1.0$ $-15.2 \pm 1.0$
Pd(1.0%Np)	$5.50 \pm 0.10$	$1.94 \pm 0.06$	
Pd(2.0%Np)	$4.37 \pm 0.08$	$2.06 \pm 0.06$	
Pd(0.5% Pu)	$5.51 \pm 0.10$ $4.97 \pm 0.10$ $4.06 \pm 0.08$	$1.15 \pm 0.06$	$-4.2 \pm 0.8$
Pd(1.1% Pu)		$0.79 \pm 0.05$	$-3.0 \pm 0.8$
Pd(2.0% Pu)		$0.55 \pm 0.05$	$-3.2 \pm 0.8$

TABLE II. Magnetic susceptibilities at 300 °K of PdNp and PdPu solid solutions. Two batches of palladium were used and are indicated as PdI and PdII. PdII is the more pure and was used to make both the magnetic susceptibility and electrical resistivity samples for dilute alloys ( $\leq 2\%$ ).

Sample	χ (10 <sup>-6</sup> emu/g)	Sample	χ (10 <sup>-6</sup> emu/g)
Pd I	4.77	PdII	4.98
Pd II (0.1%Np)	4.90	Pd II (0.1% Pu)	4.86
Pd II (0.5%Np)	4.61	Pd II (0.5% Pu)	4.54
Pd II (1.0%Np)	4.34	Pd II (1.1% Pu)	4.03
Pd II (2.0%Np)	3.86	Pd II (2.0% Pu)	3.39
Pd I (2.5%Np)	3.55	Pd I (2.5% Pu)	2.67
Pd I (5.0%Np)	3.02	Pd II (5.0% Pu)	2.07
Pd I (7.5%Np)	2.83	Pd I (6.8%Pu)	1.45
Pd I (9.8%Np)	2.89	Pd I (8.7%Pu)	0.98
Pd I (13%Np)	3.49		

ful fit. The parameters corresponding to the solid lines in Fig. 5 are listed in Table I. The values of  $\mu_{\rm eff}$  and  $\theta$  for dilute  $Pd{\rm Np}$  are rather insensitive to neptunium concentration. Assuming that  $T_{\rm K} \simeq -\theta \simeq 13~{\rm ^{\circ}K}$  and that  $\theta$  is relatively independent of neptunium concentration, even in more dilute alloys, the estimates of  $T_{\rm K}$  from the susceptibility and resistivity are in close agreement, i.e., 13 and 18  ${\rm ^{\circ}K}$ , respectively. The difference between  $\mu_{\rm eff}$  for the dilute alloys given in Table I and a value of  $\sim 2.5 \mu_B/$  Np atom found for the concentrated alloys  $^1$  ( $c \ge 5\%$ ) is not necessarily significant, as the validity of assuming  $\chi_0$  to be temperature independent is ques-

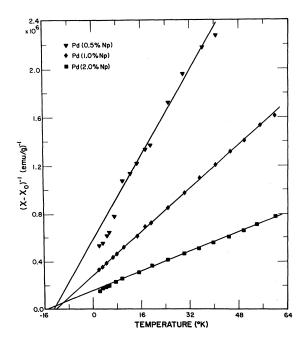


FIG. 5. Plots of  $(\chi - \chi_0)^{-1}$  vs temperature for dilute PdNp alloys.  $\chi_0$  is obtained by least-squares fitting.

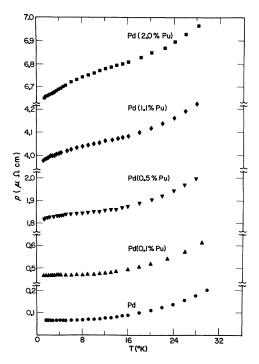


FIG. 6. Electrical resistivities of dilute PdPu alloys in the temperature range 1.3-30 °K.

tionable. The room-temperature magnetic susceptibilities of the PdNp alloys and the palladium from which they were made are shown in Table II. Two batches of palladium were used and are designated as Pd I and Pd II. The values for more concentrated alloys are also included.  $^1$ 

#### B. Palladium-Plutonium

The electrical resistivities of the PdPu alloys were also measured in the range 1.3–300 °K. The low-temperature data are shown in Fig. 6 along with the results for the pure Pd II. These alloys have positive resistivity-temperature slopes at the lowest temperatures. The slope is extremely small for the Pd(0.1% Pu) alloy, where the resistivity decreases only 0.003  $\mu\Omega$ cm between 4.2 and 1.3 °K. It was shown earlier that  $\Delta\rho/c$  for these alloys could be described by a  $\ln T$  dependence at lowest temperatures. In addition,  $\Delta\rho/c$  for these alloys lies within overlapping errors bars ( $\pm$ 10%).

The magnetic susceptibilities between 2.5 and  $300\,^\circ\mathrm{K}$  of the palladium and of solid solutions containing  $0.1-5.0\,$  -at.% plutonium are shown in Fig. 7. Two main effects are observed. Throughout this temperature range the susceptibility decreases with increasing plutonium concentration. This effect can be attributed to local filling of the host d band. At low temperatures the susceptibilities have large negative temperature derivatives. This effect can be attributed to the localization of plutonium 5f

electrons. Because of the high purity of the plutonium, this effect is not due to magnetic impurities in the plutonium.

Proceeding as for PdNp, the susceptibilities of the 0.5, 1.1, and 2.0% plutonium alloys can be fitted quite well in the range 2.5-25 °K by  $\chi = \chi_0 + C/(T-\theta)$ . Figure 8 is a plot of  $c(\chi-\chi_0)^{-1}$  vs T. The parameters corresponding to the solid lines in Fig. 8 are shown in Table I. The Curie-Weiss temperature is essentially independent of plutonium concentration. However, the effective moment is strongly composition dependent. The room-temperature susceptibilities of the PdPu alloys and the palladium from which they were made are given in Table II. The values for more concentrated alloys are also included.  $^4$ 

### IV. DISCUSSION

### A. Palladium-Neptunium

The electrical resistivity and magnetic susceptibility of PdNp alloys have been measured throughout the region of solid solubility.<sup>1,4</sup> These properties together with a negative magnetoresistance for alloys containing greater than 2% neptunium<sup>4</sup> indicate that localized moments are formed on the neptunium atoms throughout the range of solid solubility. The effective moment is about  $2-2.5\mu_B/\text{Np}$  atom and the Curie-Weiss temperature is in the range -12 to  $-25\,^{\circ}\text{K}$ . The resistivity minima in dilute PdNp are indicative of the Kondo effect.

Since neptunium has localized moments in pal-

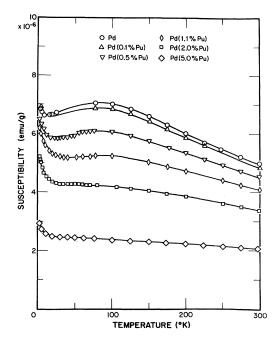


FIG. 7. Magnetic susceptibilities of *PdPu* alloys in the temperature range 2.5-300 °K.

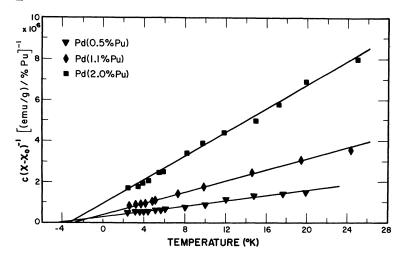


FIG. 8. Plots of  $c(\chi-\chi_0)^{-1}$  vs temperature for dilute PdPu alloys.  $\chi_0$  is obtained by least-squares fitting.

ladium, the question arises as to why such effects as giant moments and magnetic ordering are not observed even with 13% neptunium. These are the dramatic properties of dilute PdFe and PdCo.24 The answer very probably lies in the large number of neptunium valence electrons. Neptunium has seven electrons beyond a filled radon core. The electrical resistivity and magnetic susceptibility indicate ~2 electrons localized at each neptunium atom. Mössbauer data for a Pd(2.0%Np) alloy provide supporting evidence for this configuration tion. 25 The remaining 5 or 6 neptunium electrons can then be considered as valence electrons. Palladium has a large density of states which decreases rapidly above the Fermi level, 26 and a large exchange enhancement factor of about 10.26,27 The numerous neptunium valence electrons can fill the palladium d band in a region about each neptunium impurity. The uniform exchange enhancement factor, the local palladium magnetic susceptibility, and the indirect exchange interaction in the palladium host can then be sufficiently reduced to prevent phenomena like giant moments and magnetic ordering. That is to say, the modification of the palladium d band in the vicinity of neptunium atoms greatly reduces the local polarizibility of the palladium matrix. Such a model could apply to dilute<sup>28</sup> PdCr as well since chromium has six electrons beyond a filled 3p shell. It is interesting to note that neptunium and chromium, which do not have localized moments in the pure metal, are the only solutes which induce the Kondo effect in palladium.

## B. Palladium-Plutonium

In contrast to local-moment Kondo systems like PdNp, alloys like PdPu are not well understood. RhFe is the best known of these systems. It is unlikely that plutonium has a localized magnetic moment in palladium. The effective paramagnetic moment obtained from the susceptibility data for

the dilute alloys is extremely composition dependent. Thus, the Curie-Weiss fits may be fortuitous. For plutonium concentrations in the range  $2.5 \le c \le 8.7\%$ ,  $\chi_{4} \circ_{\rm K} - \chi_{300} \circ_{\rm K}$  is small and roughly constant. However, for concentrations up to 8.7% plutonium, the resistivities have positive slopes at low temperatures. The magnetoresistance of a 6.8% plutonium alloy is essentially zero, i.e., at  $4.2\,^{\circ}{\rm K}$  and  $77\,{\rm kOe}$ ,  $\Delta\rho/\rho_0 = +0.00012.^4$  Thus, data for the concentrated alloys are not characteristic of local-moment systems, and explanations of Pd Pu data which are based on localized magnetic moments<sup>29, 30</sup> do not seem appropriate.<sup>2</sup>

Recently, theories of localized spin fluctuations have been used to describe systems whose low-temperature impurity resistivity decreases with decreasing temperature. Lederer and Mills<sup>31</sup> showed that the scattering of conduction electrons by localized paramagnons causes a low-temperature resistivity varying as  $T^2$ , as is observed in dilute PdNi alloys.<sup>32</sup> Kaiser and Doniach have extended this calculation to higher temperatures.<sup>33</sup> As discussed below, the spin-fluctuation approach seems the most appropriate for PdPu.

In the theory of Kaiser and Doniach localized moments are not formed on the impurity atoms. Instead the impurities are nearly magnetic with large local-exchange enhancements. The susceptibility is assumed to be Pauli type with the temperature dependence due to the temperature variation of the exchange enhancement. The resistivity changes gradually from a  $T^2$  to a T dependence at a temperature  $T_C \simeq 0.25 T_S$ , where  $T_S$  is the spin-fluctuation temperature and  $k_B T_S$  is the energy of the peak in the localized-spin-fluctuation excitation spectrum.

For PdPu, no  $T^2$  dependence of the resistivity is observed. Only a resistivity linear in T over a small range  $(1.3 \le T \le 4 \, ^{\circ}\text{K})$  is indicated. The absence of a  $T^2$  dependence can possibly be due to the fact that  $T_C$  is below the temperature range of the

measurements. Hence,  $T_S$  would be quite low and a rough estimate is  $T_S \stackrel{<}{\sim} 1$  °K. A similar approach has been used for  $Rh {\rm Fe.}^{33}$  The model also says that  $T_s \propto \alpha^{-1}$ , where  $\alpha$  is the enhancement factor for localized spin fluctuations. Thus, a low value of  $T_S$  indicates large local enhancements at the plutonium atoms and these enhancements can explain the low-temperature susceptibilities of the  $Pd {\rm Pu}$  alloys. The one experimental fact inconsistent with the localized-spin-fluctuation model is the neglect of potential scattering in the calculations. The large low-temperature resistivities of these alloys (~3.5  $\mu {\rm \Omega cm}/$ % Pu) indicate that this scattering is not small.

In PdNp cooperative effects become important for  $c \ge 0.1\%$ . In PdPu the effects of the plutonium impurities are not observed unless the plutonium concentration is about 0.1% or more. Hence, it is possible that the plutonium 5f band does not become sufficiently narrow to manifest magnetic effects unless the plutonium impurities can interact in some manner. One possible mechanism is the indirect interaction between plutonium atoms via the conduction electrons  $^{34}$  (see Sec. IVC).

#### C. Palladium-Uranium

Localized spin fluctuations at the actinide atoms also occur in the PdU system but not for dilute uranium concentrations. In PdU the 5f level is above the Fermi level until the 4d subband of the alloy is filled near 8% uranium. For higher concentrations the 5f level narrows with increasing uranium content. In addition, localized spin fluctuations are associated with the occupation of the 5f subband. A similar approach has been taken for ThU.

A possible mechanism for the band narrowing is the indirect interaction of uranium atoms via the conduction electrons. Kim has recently developed a model to describe systems whose magnetic properties depend on the environment of the magnetic atoms.<sup>34</sup> The approach takes into account that the width in energy of an impurity state depends on the density of states of the host metal at the impurity site. This consideration is important for palladium alloys for concentrations at which the 4d subband is nearly filled. The theory demonstrates that the width of the impurity virtual bound state (vbs) is always increased by the direct transfer interaction between impurities. However, indirect impurity interactions through the host-metal conduction electrons can either broaden or narrow the width of the vbs. Considering only nearest-neighbor interactions and a simple-cubic lattice. Kim shows that

$$\delta \Delta_i / \Delta_i^0 = 4z_i \cos(2k_F d) / (2k_F d)^2 \quad , \tag{1}$$

where  $\delta\Delta_i/\Delta_i^0$  is the fractional change in the width of the impurity vbs due to indirect interactions,  $z_i$  is the number of other impurity nearest neighbors, d is the interatomic distance, and  $k_F$  is the effective Fermi wave number of the conduction-electron band. It is this interaction which is most probably responsible for the narrowing of the uranium 5f vbs in PdU.

### V. CONCLUSION

The presence of 5f electrons at the Fermi level yields a wealth of magnetic phenomena in palladium-actinide solid solutions. In neptunium alloys the 5f level is sufficiently localized to form a magnetic moment, which is spin compensated at low temperatures. In the case of plutonium, local moments are nearly formed and localized-spin-fluctuation enhancements are very large. In concentrated PdU alloys indirect interactions between uranium impurities and the modification of the palladium d band in the vicinity of uranium atoms cause the 5f level to narrow at the Fermi level.

The existence of localized moments or narrow 5f bands in these solid solutions suggests that chemically ordered intermetallic compounds might possess magnetic order. Several of these compounds are indeed magnetic. NpPd3 and PuPd3 are antiferromagnetic with  $T_N = 53$  and  $25\,^{\circ}\mathrm{K}$ , respectively. NpPd3 also has a small net magnetization below  $21\,^{\circ}\mathrm{K}$ , the Curie point. Preliminary data show that  $\mathrm{Pu_3Pd_4}$  and  $\mathrm{Pu_5Pd_4}$  also order magnetically at low temperatures.

It is noteworthy that the strong 5f character of pure neptunium and plutonium and the weaker 5f character of pure uranium as seen, for example, in the electrical resistivity is retained in palladium alloys. That is, the room-temperature resistivities of neptunium and plutonium are very large ( $\rho > 100~\mu\Omega$ cm), whereas that of uranium is much smaller ( $\rho \simeq 30~\mu\Omega$ cm). <sup>17, 36</sup> This difference in magnitude can be attributed to relatively narrow 5f bands with large density of states at the Fermi level in pure neptunium and plutonium.

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