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Spin fluctuations in actinide intermetallic compounds*

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Electrical-resistivity and magnetic-susceptibility data for sixteen actinide systems are considered in terms of localized spin fluctuations (lsf). Each of the systems had been proposed earlier as an lsf system on the basis of a T^2 resistivity dependence at low temperatures. The data are examined at higher termperatures, and it is shown that the various temperature dependences predicted by Rivier and Zlatic for lsf dilute alloys are found in these ordered-lattice systems. Various spin-fluctuation temperatures (T_{SF}) are determined for each compound. By use of the Friedel-Anderson virtual-bound-state model, values of T_{SF} are calculated, and are shown to scale simply with T_{SF}'s estimated from the susceptibility data. The results are used to explain the lack of temperature-dependent susceptibilities for the pure lsf metals α-Np and α-Pu.

I. INTRODUCTION

A number of authors have proposed the existence of localized spin fluctuations (lsf) in various pure actinides, their alloys, and intermetallic compounds.[1-5] Although strong support for this proposal is found in specific-heat results on *Pd*-U[6] alloys and pulsed-NMR results[7] on UAl_2 and $PuAl_2$, the main criterion for the proposal has been an electrical resistivity ρ which varies as T^2 at low temperatures. This criterion had been extrapolated from the dilute-alloy picture of Kaiser and Doniach[8] with the proposal that the characteristic spin-fluctuation temperature, T_{SF}, is defined as four times the temperature where the resistivity-temperature dependence changes from T^2 to T^1.

Doniach has shown[3] that in narrow-band intermetallic compounds, the low-temperature resistivity also increases initially as T^2 due to the low-frequency spin-fluctuation spectral density. However, at intermediate temperatures $0\,^\circ\mathrm{K} < T < T_{SF}$, the strong scattering due to one nearly magnetic atom per unit cell can cause the resistivity to rise faster than T^2 below the T^1 regime. This temperature region is also related to the temperature at which the coherent nature of the electron wave functions is strongly felt. Thus, as in solid-solution alloy cases, some of the intermetallic compounds, e.g., $PuAl_2$, exhibit resonant scattering, i.e., $\rho = 1 - \alpha T^2$ (normalized to unity). However, at low temperatures the fairly narrow 5*f* band hybridizes with the conduction electrons into a well-defined Fermi surface. At low temperatures, the strong resonant scattering of the conduction electrons does not contribute to the resistivity, so $\rho \rightarrow 0$ and $T \rightarrow 0\,^\circ\mathrm{K}$. This effect is certainly not present in the dilute-alloy case but seems to be operative in well-annealed $PuAl_2$ and α-Pu.[1,3,7]

Additional evidence in support of the spin-fluctuation model had been seen in several of the cases to be discussed where the magnetic susceptibility was exchange enhanced and followed a Curie-Weiss law at temperatures above T_{SF}, but tended towards a zero temperature dependence below T_{SF}. A demonstration of the gradual development of true local-moment behavior (leading to low-temperature long-range magnetic order) in an actinide series, via a state in which the 5*f* levels are still too broad to permit magnetic order, is found in the series, $ThRh_3$, URh_3, $NpRh_3$, and $PuRh_3$.[9] In this series, $ThRh_3$ and probably URh_3 show no evidence for magnetic excitations, while $PuRh_3$ order antiferromagnetically at 6.6 °K. The intermediate compound $NpRh_3$ shows $\rho \propto T^2$, changing to T^1 at 15 °K (implying $T_{SF} \sim 60\,^\circ\mathrm{K}$). The magnetic susceptibility follows a Curie-Weiss law above 125 °K and is proportional to $1 - T^2$ below that temperature.

A weakness of the earlier proposals of actinide lsf's is that they rely most heavily on the T^2 dependence of the resistivity as evidence for lsf's. This temperature dependence may be accidental in some cases, or due to other scattering processes, e.g., Baber scattering,[10] spin-disorder scattering in systems with weak antiferromagnetic ordering,[11] etc. Rivier and Zlatic (RZ)[12] have shown that in addition to the T^2 and T^1 regimes described above, in the dilute-alloy lsf systems there are $\ln T$ and $1/T$ dependences also. Furthermore, the value of T_{SF} determines the strengths of the scattering in the various regimes and/or the temperature range of their validity. Thus, T_{SF} nearly defines the full ρ-T curve due to lsf conduction-electron scattering. Although RZ point out that the same curves could be obtained for any quantum scattering containing internal structure, evidence for the excitations being magnetic in a particular case may be obtained from the expectation of a Curie-Weiss susceptibility above T_{SF}.

This paper examines the resistivity and susceptibility data for sixteen actinide systems which

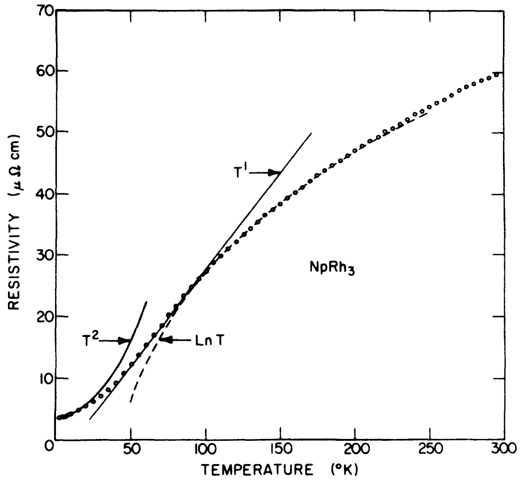

FIG. 1. Resistivity vs temperature curve for $NpRh_3$. Various temperature dependences are indicated.

have been proposed as lsf materials. It is concluded that the dilute alloy model of RZ is appropriate to these systems. The formation of virtual bound states (vbs) and lsf scattering in dilute transition-metal intermetallic compounds has also been proposed, recently.[13–15]

II. RESULTS

Figures 1 and 2 show resistivity-temperature data for $NpRh_3$ and $PuZn_2$, which have not been previously published. These compounds typify the entire set in that $NpRh_3$ has a positive $d\rho/dT$ at all temperatures, while the ρ-T curve for $PuZn_2$ has a maximum and a negative $d\rho/dT$ at temperatures above the maximum. The latter type of behavior is found in $PuAl_2$ and α-Pu also, which are referred to in Sec. I. Figure 3 shows the high- and low-temperature T^2-dependent resistivity of $PuZn_2$. The magnetic susceptibility data for $PuZn_2$ are plotted in Fig. 4. It is seen to follow a modified Curie-Weiss law above 15 °K with $\chi = \chi_0 + C/(T+\theta)$, with $\chi_0 = 4.1\times10^{-4}$ emu/mole, $\theta = -30$ °K, and the Curie constant C equivalent to an effective moment of $0.71\mu_B$. Figures 5–7 show the ln$T$ dependences for all of the compounds (and pure α-Np and α-Pu) considered here. Figures 8–10 show the T^{-1} approach to the saturation values. In Fig. 1, these regimes are labeled on the full curve for $NpRh_3$.

It is seen that for all of the materials considered here, there are ln$T$ and T^{-1} dependences. In all of these cases, except for $PuIr_2$ and $PuRu_2$, there are also regions of T^2 behavior which have been reported previously.[1,4,7] As pointed out by RZ the T^1 regime is an artifact of the curve, and it is seen in all of these cases, also. From the work of RZ there are several ways in calculating T_{SF}

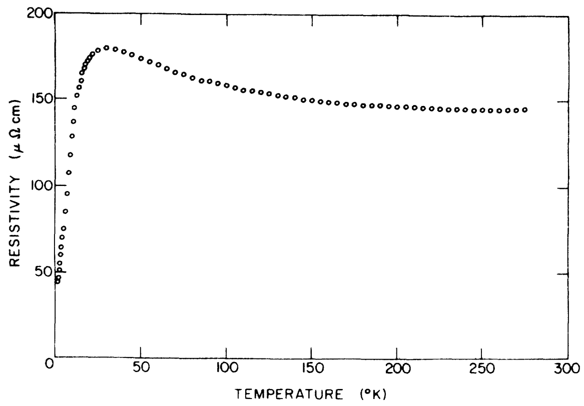

FIG. 2. Resistivity vs temperature curve for $PuZn_2$.

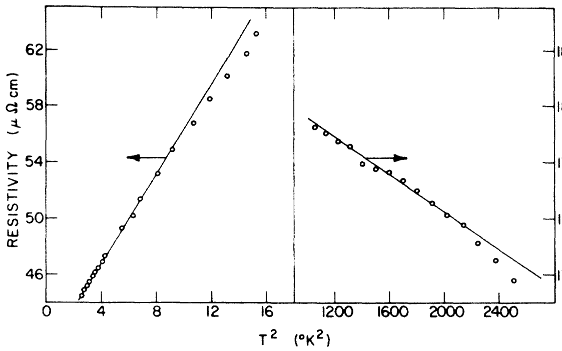

FIG. 3. Resistivity of $PuZn_2$ plotted as a function of temperature squared. (a) below the resistivity maximum; (b) above the resistivity maximum.

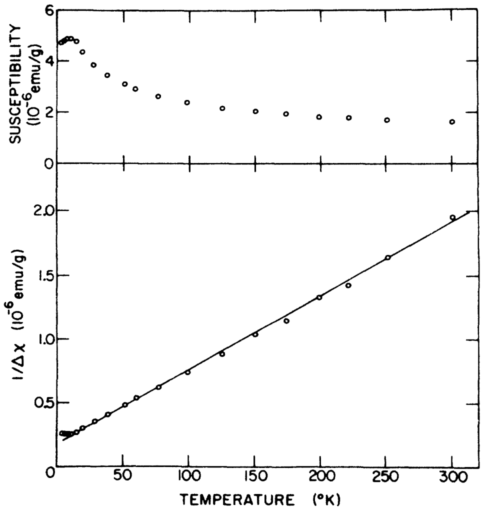

FIG. 4. Magnetic susceptibility vs temperature of $PuZn_2$.

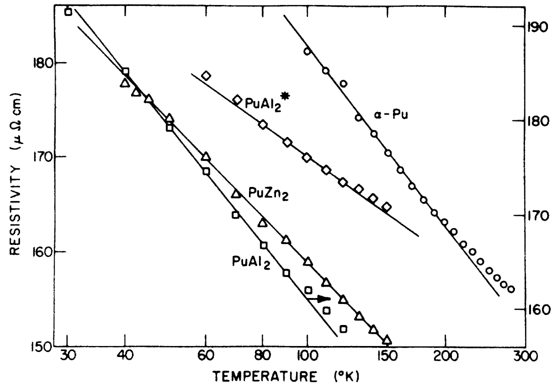

FIG. 5. Dependence of the resistivity of various actinides on the logarithm of temperature.

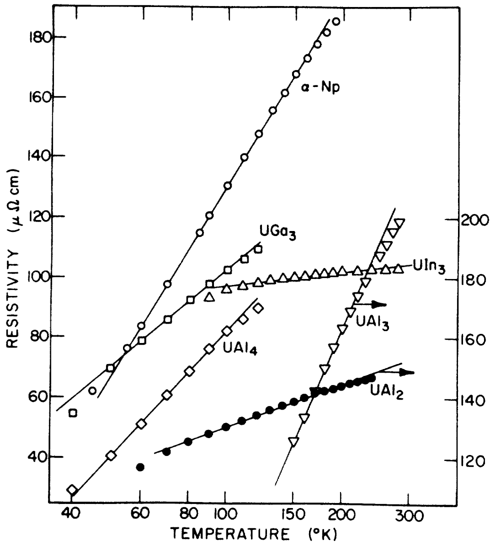

FIG. 7. Dependence of the resistivity of uranium intermetallic compounds on the logarithm of temperature. Data for UGa_3, UIn_3, UAl_3, and UAl_4 from Ref. 4.

from the ρ-T curves, where the high-temperature limit has been normalized to unity: (a) The low-temperature end of the $\ln T$ region is identified with T_{SF}. (b) The high-temperature end of the T^2 region is equal to $T_{SF}/2\pi$ (leading to higher T_{SF}'s than the method of Kaiser and Doniach).[16] (c) The slope of the T^2 region is given by $d\rho/d(T^2) = 3\pi^2/4T_{SF}^2$. (d) The slope of the T^1 region is $d\rho/dT = 1.12/T_{SF}$. (e) The approach to the high-temperature limit is $-d\rho/d(1/T) = T_{SF}$. (f) At some temperature above T_{SF}, the susceptibility will begin to follow a Curie-Weiss law, and that temperature may be taken as a measure of T_{SF}.

Thus, it is seen that if one adopts the results of the dilute-alloy case, there can be up to six calculations of T_{SF} for each material. Table I lists the values so calculated using the data of Refs. 1, 4, and 9. In addition to the theoretical uncertainty in using the dilute-alloy calculations of RZ, there is an experimental uncertainty because the calculations of T_{SF} have used the total resistivity. Unlike the dilute-alloy case, where it can be argued that the temperature-dependent resistivity of a nonmagnetic alloy may be used to approximate the electron-phonon scattering contribution, that is not possible in the present case. Since the electron-phonon contribution must be averaged over the Fermi surface, it will not be the same, for example, in $NpRh_3$ as in $ThRh_3$ because the $5f$ electrons will cause a major change in the Fermi surface. However, in *Al*-Mn, for example, it is easily argued that in dilute alloys the Fermi surface is essentially the same (for gross scattering

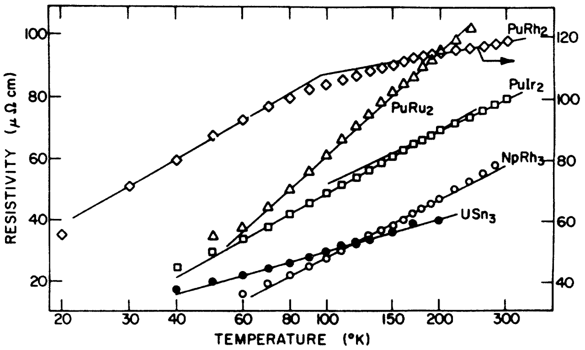

FIG. 6. Dependence of the resistivity of actinide intermetallic compounds on the logarithm of temperature. USn_3 data from Ref. 4.

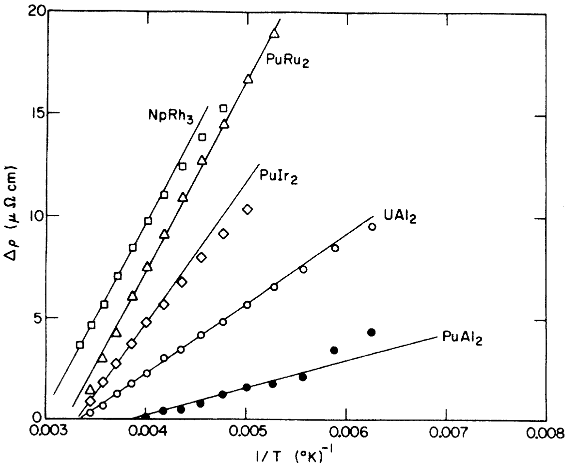

FIG. 8. Approach of the resistivity of various actinides to their limiting values as the reciprocal of temperature.

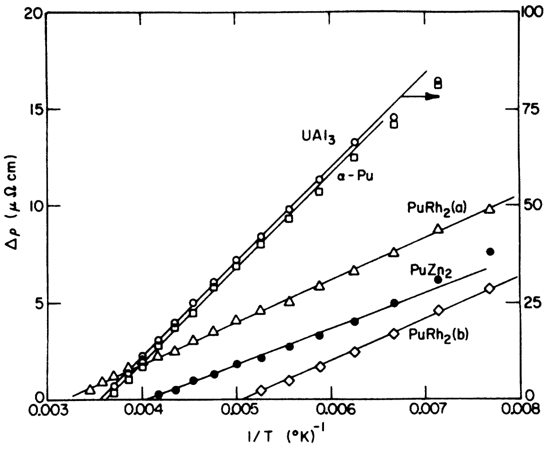

FIG. 9. Approach of the resistivity of various actinides to their limiting values as the reciprocal of temperature. UAl_3 data from Ref. 4.

purposes) as in *Al*-Cu or *Al*-Zn.[17] Thus it is not surprising that the various T_{SF}'s are not the same. There is further error in method *E* due to uncertainty in the lsf scattering limit. Obviously it is expected that in a given material one or more of the T_{SF}'s could be in large error whenever the lsf scattering is not the dominant temperature-dependent contribution. Occasionally it is found that the data for a given material follow a particular temperature dependence in more than one temperature range. Thus, it is seen in Fig. 6 that the data for $PuRh_2$ follows a ln*T* law between 30–60 °K and 180–300 °K. However, there is some general agreement among the values.

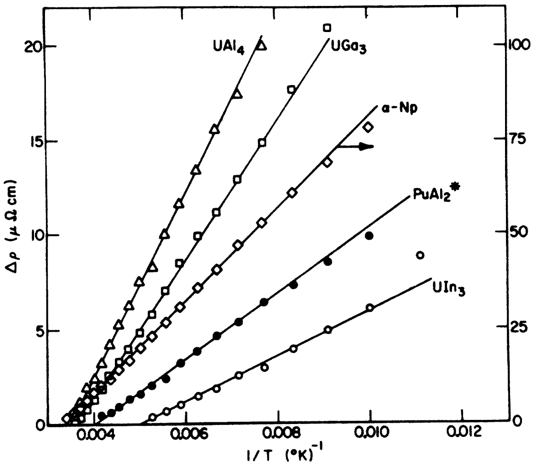

FIG. 10. Approach of the resistivity of various actinides to their limiting values as the reciprocal of temperature. Data for UAl_4, UGa_3, and UIn_3 from Ref. 4.

TABLE I. Calculated spin-fluctuation temperatures for various actinides.

	Spin-fluctuation temperature (°K), calculated by method					
System	*A*[a]	*B*[b]	*C*[c]	*D*[d]	*E*[e]	*F*[f]
USn_3[g]	70	69	30	17	...	50
UGa_3	50	310	192	94	29	200
UAl_3	190	750	480	246	123	600
UIn_3	40	250	138	86	12	150
USi_3	...	440	930	1400	...	>1000
UGe_3	...	440	910	1100	...	600
UAl_2	85	16	57	712	24	100
α-Np	60	63/250	600/225[h]	210	59	>300
α-Pu	100	63	170	30	32	>300
UAl_4	45	220	550	130	43	150
$PuRh_2$	30	82	67	56	18	30
$PuAl_2$	40	9	27	3	10	30
$PuAl_2$[i]	90	310	466	548	35	30
$NpRh_3$	100	94	257	248	142	125
$PuZn_2$	45	19/220	21/472	15/380	13	20
$PuIr_2$	70/180	250	...	234	90	>300
$PuRu_2$	60	...	...	203	82	>300

[a] Ln *T* region.

[b] T^2 region.

[c] Slope in T^2 region.

[d] Slope in T^1 region.

[e] Slope in T^{-1} region.

[f] Susceptibility.

[g] The data of Refs. 1, 4, 7, and 9 were used in this analysis.

[h] Where two values are given, the first one is for low temperature.

[i] Radiation-damaged $PuAl_2$.

III. DISCUSSION

It would be useful to review the electronic-structure model being used to discuss these materials. It is assumed that in each case we have a nearly magnetic system due to the presence of 5*f* electrons at or near the Fermi surface. Unlike the case in 4*f* electron systems the 5*f* states are broadened due to direct overlap of 5*f* wave functions or to 5*f*-6*d* hybridization (either direct or indirect) or to a 5*f*–conduction-electron interaction.[18] This situation is similar to the case of dilute transition-metal alloys which have a broadened *d*-electron virtual bound state. Thus the compounds may be discussed in terms of the Friedel-Anderson model.[19] Experimental support for this approach is found in susceptibility data, where the Curie-Weiss constants usually yield effective moments close to 4, 2.5, and 1 for U, Np, and Pu, respectively. In this model, the Hamiltonian is given by

$$H = H_s + H_f + H_{sf} \qquad (1)$$

with H_s the unperturbed free electron energy, H_f is the unperturbed energy of a single *f* level, and H_{sf} is the *s*-*f* mixing term due to the interaction V_{kf}. In the following qualitative discussion, *s*-*f* and *d*-*f* mixing will be combined into a single V_{kf}:

TABLE II. Results from the Friedel-Anderson model.

System	χ_0[a] (10^{-4} emu/mole)	$n_f(E)$ (eV^{-1})	α	U (eV)	$T_{\mathrm{SF}}^{\mathrm{CALC}}$ (°K)	Δ (eV)	$U/\pi\Delta$	V_{kf} (eV)
USn_3	99.5	72[b]	4.3	0.0107	11.9	0.0043	1.2	0.037
UGa_3	18.6	22.2[b]	2.6	0.0277	63.9	0.017	0.51	0.074
UAl_3	15.2	17.8[b]	2.6	0.0346	79.7	0.052	0.21	0.128
UIn_3	32.3	21.3[b]	4.7	0.0370	36.9	0.013	0.91	0.064
USi_3	5.3	6.0[b]	2.7	0.1049	228	$\geqslant$0.086	0.39	$\geqslant$0.165
UGe_3	10.9	8.7[b]	3.9	0.0856	109	0.052	0.53	0.128
UAl_2	3.8	35.6[c]	3.5	0.020	29.7	0.0086	0.74	0.165
α-Np	5.5	6.0[d]	3.0	0.110	207	(0.071)[f]	(0.49)	(0.150)
α-Pu	5.5	10.6[e]	1.6	0.0363	215	(0.074)	(0.16)	(0.154)
UAl_4	16.6	15.47	(3.5)	(0.0462)	68	0.013	(1.1)	...
$PuRh_2$	3.7	3.45	(3.5)	(0.207)	307	0.0028	(23)	...
$PuAl_2$	13.2	12.29	(3.5)	(0.0581)	86	0.0028	(6)	...
$NpRh_3$	19.1	17.8	(3.5)	(0.0401)	59	0.011	(1.2)	...
$PuZn_2$	4.07	3.79	(3.5)	(0.1883)	279	0.0017	(28)	...
$PuIr_2$	5.75	5.36	(3.5)	(0.1332)	197	(0.07)	(0.6)	...
$PuRu_2$	9.3	8.67	(3.5)	(0.0823)	317	(0.07)	(0.4)	...

[a] Taken from Refs. 1, 4, 7, and 9.

[b] H. J. van Daal *et. al.* (Ref. 20).

[c] A. C. Gossard, V. Jaccarino, and J. H. Wernick, Phys. Rev. 128, 1038 (1962).

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[f] All parenthetic values are estimated without experimental data.

$$H_f=\epsilon_f(n_{f\uparrow}+n_{f\downarrow})+Un_{f\uparrow}n_{f\downarrow} \qquad (2)$$

where ϵ_f is the energy of the unperturbed f level with just one electron and U is the f-f Coulomb repulsion energy for f electrons of opposite spin on the actinide site:

$$U=\int\int|\phi_f(\vec{r}_1)|^2\frac{e^2}{|\vec{r}_1-\vec{r}_2|}|\phi_f(\vec{r}_2)|^2 d\vec{r}_1 d\vec{r}_2 \quad , \qquad (3)$$

where $\phi_f(\vec{r})$ is the f-electron wave function and e is the electronic charge. The interaction U tends to separate electrons of opposite spin, leading to magnetism, while V_{kf} broadens the f state to a half-width of Δ, increases the vbs overlap and can prevent magnetism. The condition for magnetism is

$$U/\pi\Delta>1 \quad , \qquad (4)$$

where

$$\Delta=\pi\,\langle V_{kf}^2\rangle\, n_c(E) \qquad (5)$$

with $n_c(E)$ being the density of conduction-electron states at the Fermi level. The condition for spin-fluctuation behavior is

$$U/\pi\Delta\lesssim 1 \quad . \qquad (6)$$

From measurements of the specific heats and Pauli-band susceptibility, χ_0, we can calculate the Stoner enhancement,

$$\alpha=\chi_0/\chi_c \quad , \qquad (7)$$

where χ_c is the calculated band susceptibility assuming that the measured electronic specific-heat coefficient γ is due to the bare density of states. These data are available for the uranium compounds[20] and are listed in Table II. The enhancement factor is also given by

$$\alpha=[1-Un_f(E)]^{-1} \quad , \qquad (8)$$

where $n_f(E)$ is the f density of states at the Fermi level. The lifetime τ_{SF} of the fluctuating spin populations is given by

$$\tau_{\mathrm{SF}}=\alpha\pi n_f(E) \quad . \qquad (9)$$

Calculated values of α, U, and $T_{\mathrm{SF}}^{\mathrm{CALC}}=(h/k_B\tau_{\mathrm{SF}})$ are also given in Table II, where the total density of states has been substituted for $n_f(E)$.

In view of the uncertainties in determining T_{SF} from the *total* resistivity, as discussed in Sec. II, the magnetic susceptibility was used to estimate T_{SF}. It is assumed that the temperature at which a temperature-dependent susceptibility begins, T_{SF}^F in Table I, is a meaningful measure of where the lsf's appear to be magnetic. Figure 11 shows a plot of $T_{\mathrm{SF}}^{\mathrm{CALC}}$ vs T_{SF}^F—the apparent simple relationship is quite good. Note that $T_{\mathrm{SF}}^F/T_{\mathrm{SF}}^{\mathrm{CALC}}\approx 4$–5, which is similar to the situation found in Kondo alloys.[21] Some measure of the errors introduced into the calculation of $T_{\mathrm{SF}}^{\mathrm{CALC}}$ via estimates of $n_f(E)$, χ_0, and χ_c may be seen, for example, in the case of UAl_2. If it is assumed that the measured γ is enhanced by a factor of 2 by the electron-phonon interaction, and that $n_f(E)$ should be corrected by the density of states of $\frac{1}{3}$ g atom of Al (replacing UAl_2 by "Al Al_2"), the calculation yields $\alpha=7$

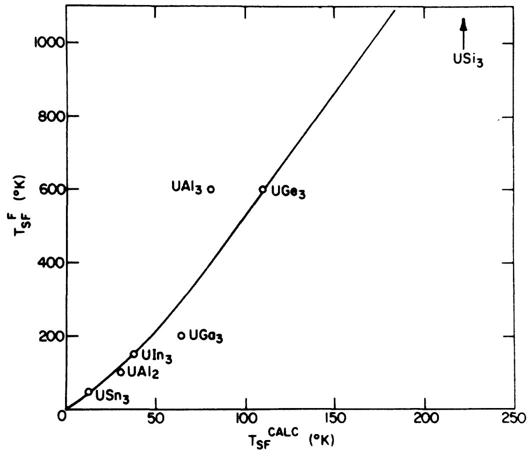

FIG. 11. Plot of T_{SF}^{CALC} for uranium intermetallic compounds vs T_{SF}^{F} from Table I.

$U = 0.051$ eV, and $T_{SF}^{CALC} = 31.4$ °K vs $\alpha = 3.5$, $U = 0.020$ eV, and $T_{SF}^{CALC} = 29.7$ °K. These changes do not become substantial until α is changed by factors larger than 3. Also given in Table II are the values of Δ (assuming $\Delta = k_B T_{SF}^{F}$), $U/\pi\Delta$, and V_{kf}, assuming $n_c(E) = 1$ state/eV. It is seen that $U/\pi\Delta$ is close to unity for all of the uranium compounds, in agreement with the proposal of lsf's. Even within the uncertainties of the calculation, $U/\pi\Delta > 1$ only for USn_3, which is nearest to being magnetic, i.e., τ_{SF} is large.

Also listed in Table II are calculations for α-Pu and α-Np; these yield values for T_{SF}^{CALC} of 215 and 270 °K, respectively. If we assume $T_{SF}^{F}/T_{SF}^{CALC} = 4$, as for Fig. 11, then we should not expect temperature-dependent susceptibilities in either material below 800 °K, well above their transformation temperatures. Table II also lists expectations for the other compounds (for which no specific-heat data exist) based on the assumption of $\alpha = 3.5$. Moderate agreement with the curve of Fig. 11 is obtained for UAl_4, $NpRh_3$, $PuAl_2$, and perhaps for $PuRu_2$ and $PuIr_2$. The results are not in agreement for $PuZn_2$ and $PuRh_2$. However, exchange enhancements in excess of 15 for both compounds would find them in agreement with the curve of Fig. 11. A good test of the present calculations will be provided by specific-heat measurements on the latter two compounds.

Although the calculated U, Δ, and V_{kf} values are in good internal agreement, they are substantially different from the set of internally consistent values used by Jullien *et al*,[22] to explain the retardation of magnetic order in pure actinides until curium (element 96) due to V_{kf}. Their values for U, Np, and Pu are $\Delta = 0.1$ eV, $V_{kf} = U = 20\Delta = 2$ eV. Their set gives $U/\pi\Delta = 6.3 > 1$ and therefore would predict magnetism if not for the large V_{kf}.

IV. CONCLUSIONS

It is shown that the resistivity-temperature dependences expected for dilute-alloy lsf systems are found for pure actinides and actinide intermetallic compounds. Although it is difficult to subtract away the electron-phonon scattering contributions in these systems, the various T_{SF}'s calculated from the resistivity and susceptibility-temperature curves are in moderate agreement. Applications of the Friedel-Anderson model to the systems for which low-temperature specific-heat data exist yields calculated T_{SF}'s which scale nicely with experimental values. The calculation explains in a simple way why the lsf systems α-Pu and α-Np do not show a temperature-dependent susceptibility.[23] The calculated band parameters are in agreement with the requirements of lsf's.

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[23] A recent discussion of this problem from another point of view may be found in R. Jullien, M. T. Beal-Monod, and B. Coqblin, Phys. Rev. Lett. 30, 1057 (1973). In that work, however, no explanation is given for the lack of a temperature-dependent susceptibility, and $\alpha = 0$ is assumed for α-Pu. See also Ref. 3.