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BAND MAGNETISM DUE TO f-ELECTRONS

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ABSTRACT

Specific heat data illustrate several types of itinerant or band magnetism in actinide intermetallic compounds. The results show ferromagnetic spin fluctuations in UAl_2 with $T_{sf} = 25K$, itinerant antiferromagnetism in $NpSn_3$ with $T_N = 9.5K$, and itinerant ferromagnetism in $NpOs_2$ with $T_C = 7.9K$. Specific heat studies of dilute $U_{1-x}Th_xAl_2$ show the theoretically predicted modifications due to impurity scattering in a spin fluctuation system. For $NpSn_3$ it is possible to show the BCS nature of the transition due to the gap formation.

INTRODUCTION

Contrary to early beliefs nurtured by an emphasis on chemistry research, the light actinides Th, Pa, U, Np, and Pu are more closely related to 3d electron systems in the solid state than they are to 4f electron systems [1]. This happens because of two facts: (1) the stabilization of the 5f electronic states occurs much more slowly than in the corresponding 4f series, so that even by Pu, the occupied 5f electronic levels are not very far below the Fermi surface; and (2) the radial extent of the 5f wave functions is considerably larger than for 4f electrons, and even approaches that of 3d electrons. As a consequence of these two facts, the 5f electrons readily form bands due to direct overlap and/or hybridization with s-p-d electrons, and these bands are likely to overlap the Fermi level. Clear evidence of 5f electron band formation has been seen in optical spectroscopy [2] and deHaas-vanAlphen measurements [3].

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The similarity of the 3d and 5f series to each other is also seen in attempts to characterize the magnetism of actinide intermetallic compounds as being localized or itinerant. This paper discusses the properties of three well-characterized examples of itinerant 5f-electron magnetism, with strong emphasis on specific heat data. These are UAl_2 , a nearly ferromagnetic spin fluctuation compound [4]; $NpSn_3$, an itinerant antiferromagnet (IAF) [5]; and $NpOs_2$, a weak itinerant ferromagnet (WIF) [6,7]. As will be pointed out these are far from being isolated cases of 5f-band magnetism.

UAl_2 , A SPIN FLUCTUATION COMPOUND

UAl_2 is a cubic Laves phase compound ($MgCu_2$ -type) with an actinide-actinide separation of $3.38 \text{ \AA}$. This separation is below the $3.4\text{--}3.6 \text{ \AA}$ proposed by Hill [8] as the cut-off for local moment formation in uranium compounds as a consequence of 5f-5f wave function overlap. The electrical resistivity at low temperatures has been shown to vary as T^2 . The resistivity and the magnetic susceptibility data, shown in Fig. 1 and discussed below, has lead to the proposal of ferromagnetic spin fluctuations [9]. Nuclear magnetic resonance and nuclear spin-lattice relaxation times, T_1 , measured on the ^{27}Al nucleus have shown that UAl_2 does not order magnetically, but the T_1 measurements could not confirm the existence of spin fluctuations [10].

From the theory of Doniach and Engelsberg [11], the leading terms in the spin fluctuation contribution to the electronic specific heat for $T \ll T_{sf}$ are

$$C/T = d + f T^2 \log(T/T_{sf}) + \dots \quad (1)$$

where $d = (\pi^2/3) \gamma$ is the spin fluctuation enhanced electronic coefficient. The term f is proportional to S/T_{sf}^2 , where S is the Stoner exchange enhancement. The second term in Eq. (1) is negative and is responsible for a drop in C/T as $T \rightarrow 0$ as is seen in Figs. 2 and 3. Figure 2 shows the low

temperature specific heat data for UAl_2 from Ref. 4 as a function of applied magnetic field. The zero field data are well described by Eq. (1) with $S = 4$, $T_{\text{sf}} = 25\text{K}$, and $\gamma = 142 \text{ mJ}/(\text{mole-K}^2)$ and are given as the solid line [4]. This fit is also shown in Fig. 3 for the open circles.

Most importantly, C/T is seen to be diminished by less than 2% for an applied field of 43 kOe, in excellent agreement with the calculation of Béal-Monod, Ma, and Fredkin [12]. They showed that the shift $\delta C/T$ caused by an applied field H should be

$$\delta C/T \approx 0.1 \left[\frac{\mu H}{k_B T_{\text{sf}}} \right]^2 \frac{S}{\log S} \frac{C}{T} \quad (2)$$

and for $T_{\text{sf}} = 25\text{K}$ and $S = 4$ (as determined in Ref. 4), a field of 43 kOe yields $\delta C/T \approx 1\%$, in good agreement with the data. This result strongly confirms the existence of ferromagnetic spin fluctuations in UAl_2 . It should be noted that the invalidity of the spin fluctuation interpretation of the specific heats of nearly ferromagnetic alloys such as Rh-Ni was shown by such in-field measurements [13]. In that case, fields of 40 kOe totally suppressed the contribution thought to be due to spin fluctuations, and which are now known to be associated with Ni clusters.

Fulde and Luther [14] have shown that the effect of nonmagnetic scattering centers is to modify the $T^2 \log T$ term in Eq. (1) while leaving the constant term essentially unchanged. Introducing a characteristic temperature $T_i \propto 1/\ell$ where ℓ is the mean free path associated with the impurities, one finds $T^2 \log T \rightarrow T^2 \log(T + T_i)$. We now have for Eq. (1)

$$C/T = d + f T^2 \log[(T + T_i)/T_{\text{sf}}]. \quad (3)$$

Figure 3 shows the specific heats of $\text{U}_{1-x}\text{Th}_x\text{Al}_2$ for small x . Increasing the Th content causes an increase in γ (the curves are all parallel above 9K) and a smaller minimum in C/T . Eq. (3) predicts that as T_i grows due

to a decrease in ℓ , the magnitude of the minimum in C/T will be reduced. The magnitude of C/T at $T = 0$ will be unchanged except as the reduced 5f-5f overlap may increase the tendency towards magnetism. It is possible to estimate T_1 independently from the fits of the data to Eq. (3) or by using the residual resistivities [15]. The calculated values are given in Table I and agree quite well with each other. These results provide strong support for the previously untested Fulde-Luther theory, as well as additional evidence for the validity of the spin fluctuation model for UAl_2 .

Returning to Fig. 1 we see from the inset that at low temperatures $\chi \propto 1 - aT^2$ in keeping with the calculation of Ref. 12. The coefficient a corresponds to $T_{sf} = 30K$ which is very consistent with the specific heat and resistivity results. Above 50K there seems to be another region where $\chi \propto 1 - bT^2$, which is probably associated with the narrow band structure. A modified Curie-Weiss behavior is observed for $T > 100K$. This is consistent with the values of S and T_{sf} observed at low temperatures, since for the narrow 5f band a degeneracy temperature $T_F = T_{sf}$ $S \approx 100K$ is expected.

$NpSn_3$, AN ITINERANT ANTIFERROMAGNET

$NpSn_3$ crystallizes, probably peritectically, in the ordered cubic $AuCu_3$ structure (type $L1_2$). Mössbauer measurements have shown that there is a magnetic transition at 9K with a zero degree hyperfine field H_{hf} at the Np site of 0.55 MOe [16]. Dunlap and Lander have shown that for a large number of Np intermetallic compounds there is a linear correspondence between H_{hf} and the ordered moment μ found by neutron diffraction measurements [17]. This relationship, $\mu = (0.522 \mu_B/MOe)H_{hf}$, yields a very small $\mu = 0.29 \mu_B/Np$ for $NpSn_3$. Since the linear relationship between H_{hf} and μ may only be valid for local moment systems, the quantitative value of μ obtained for $NpSn_3$ may not be exact, but nonetheless gives a strong indication of its nonlocal character.

The antiferromagnetic nature of the magnetism in NpSn_3 is shown by the two site character of the Sn Mössbauer spectrum below 9K, and the weak temperature dependence of the susceptibility χ [5,17]. Our susceptibility data are shown in Fig. 4 for samples prepared by two different methods. Since neither technique yielded a sample free of a magnetic second phase material, the data are not to be considered as final. At T_N the theory of Fedders and Martin for IAF metals [19] predicts a rapid rise in χ , which is seen in the data. For $T > T_N$, χ is dominated by the Pauli contribution and its temperature dependence is determined by structure in the density of states. Just as $d\chi/dT$ is at a maximum value at T_N , so is the slope of the resistivity curve [5].

Figure 5 shows the electronic specific heat of NpSn_3 , C_E . Data for the nonmagnetic isostructural compound USn_3 were used to determine the phonon contribution [20]. These data show a very large electronic term $\gamma_p = 242 \text{ mJ}/(\text{mole-K}^2)$ for $T > T_N$ and a reduced, but still large, γ_o for $T < T_N$ of $88 \text{ mJ}/(\text{mole-K}^2)$ and $T_N = 9.5 \pm 0.1\text{K}$. The value of γ_p is the largest electronic term found for any pure actinide compound thus far. These results are in keeping with the IAF model, i.e., a large $N(\epsilon_F)$ in the paramagnetic region with $\gamma_o < \gamma_p$ due to the formation of a gap in $N(\epsilon_F)$. The mean-field treatment of Fedders and Martin [19] for IAF with perfect nesting of electron and hole sheets of Fermi surface leads to a gap equation and energy spectrum quite similar to those for a superconductor. In Fig. 5 we have shown the results of a BCS model for C_E . This calculation requires T_N and $(\gamma_F - \gamma_o)$ but has no adjustable parameters. The agreement between theory and experiment is remarkable and is strong evidence for a second order itinerant transition. At the lowest temperatures, where collective modes are important, and at $T \geq T_N$, where C_E has a tail, we

expect the observed deviation from the mean-field model. We observe for NpSn_3 a jump in C_E , $\Delta C_E(T_N) = 1.63(\gamma_p - \gamma_o)T_N$, which is quite close to the BCS prediction.

The excess magnetic entropy ΔS_m due to the antiferromagnetic transition is given by $\Delta S_m = \int (C_E - \gamma_p T)(dT/T)$. This has been determined from the data in Fig. 5 and is found to be $-0.049 \text{ J/(mole-K)} = -0.007 R \ln 2$. The negative value is not physical, but it is due to uncertainties in the experimental data, $\sim \pm 1.0\%$, and to uncertainties in the phonon contribution to C . Thus, within the accuracy of the measurement $\Delta S_m = 0$, and $\Delta S_m \ll R \ln 2$, which is the minimum value expected for magnetism due to local moments.

NpOs_2 , A WEAK ITINERANT FERROMAGNET (WIF)

Mössbauer, neutron diffraction, and magnetization, M , results have shown that NpOs_2 is ferromagnetic with $T_C = 7.5\text{K}$ and an ordered moment $\mu = (0.25-0.44)\mu_B$ [6]. Neutron diffraction results show that the moment is not localized at the Np site [6]. The slope of the resistivity-temperature curve is a maximum at 7.4K [21]. The specific heat data of NpOs_2 are plotted in Fig. 6. The solid line represents an estimate of the phonon contribution determined with the aid of data for NpRu_2 [7], which does not order magnetically, plus γ_p for NpOs_2 .

Again, $\gamma_p = 92 \text{ mJ/(mole-K}^2\text{)}$ is very large, but $\gamma_o = 205 \text{ mJ/(mole-K}^2\text{)}$ is unexpectedly larger by a factor of two. The peak in C occurs at $T_C = 7.9 \pm 0.1\text{K}$. A magnetic entropy $\Delta S_m = (0.2 \pm 0.05)R \ln 2$ is obtained from the data. Despite the uncertainty, ΔS_m is too small to be due to local moments.

Murata and Doniach have shown that C/T below T_C may be larger than C/T above T_C in an itinerant ferromagnet [22]. Alternatively, the relative values of γ_p and γ_o may be due to a pinning of the Fermi energy at a large $N(\epsilon_F)$ in one of the exchange split subbands such that $N(\epsilon_F)_p < N(\epsilon_F)_o$.

It should be possible to use these data and the magnetization results [6] to test the Stoner-Wohlfarth-Edwards [23] theory for weak itinerant ferromagnetism on NpOs_2 . At the outset, there are two difficulties: (1) the M^2 vs H/M curves are not linear below T_C making it difficult, for example, to determine $M(0,0)$, M at $H = 0$, and $T = 0$, with any accuracy, and (2) the value of $M(0,0) \approx 0.4 \mu_B$ may not fit the criterion for WIF as defined in Ref. 23, $M(0,0) \ll 1$.

From Refs. 6 and 23 there are several ways to determine $M(0,0)$. We may take the high field magnetization data alone and obtain $M(0,0)$ from the extrapolation to $H/M = 0$. This plot is linear above 23 kOe and yields $M(0,0) = 0.39 \mu_B$. In Ref. 6 the data are fit to a low field term due to the approach to saturation and the high field band term. This yields $M(0,0) = 0.44 \mu_B$. A plot of the intercept I of the M^2 vs H/M curves on the M^2 axis should vary as T^2 [23], and is given by

$$I(T) = [M(0,0)]^2 - [M(0,0)]^2 T^2/T_C^2. \quad (4)$$

The data for NpOs_2 , plotted in Fig. 7, are seen to fall into two sets, for $T > T_C$ and $T < T_C$. For $T > T_C$ we obtain $M(0,0) = 0.38 \mu_B$ and $T_C = 9.0\text{K}$; and for $T < T_C$, $M(0,0) = 0.33 \mu_B$ and $T_C = 8.6\text{K}$. Both T_C 's derived in this way are too high. In summation, therefore, $M(0,0) = 0.33$ – $0.44 \mu_B$; we will use the mean value $0.38 \mu_B = 2110 \text{ emu/mole}$.

If we define χ_0 as the slope of the M^2 vs H/M curve for $T > T_C$ (where the curves are linear) we obtain $\chi_0 = 0.123 \text{ emu/mole}$. Clinton and Viswanathan [24] have pointed out that the equations of Ref. 23 yield for the jump in C at T_C

$$\Delta C(T_C) = [M(0,0)]^2/\chi_0 T_C \quad (5)$$

from which we calculate $\Delta C/T_C = 460 \text{ mJ}/(\text{mole-K}^2)$ vs the experimental value of 190 -- about the same order of agreement found for ZrZn_2 [24].

Before proceeding further, it is necessary to determine the Stoner exchange enhancement, S , which may be found from

$$S = \frac{N(\epsilon_F) \chi}{N(\epsilon_F) \chi_\gamma} = \frac{\chi_p / 2\mu_B^2}{3\chi_p / [2\pi^2 k_B^2 (1 + \lambda)]} \quad (6)$$

where χ_p is the Pauli spin susceptibility and λ is a many-body enhancement, which includes electron-phonon and electron-spin fluctuation contributions.

In order to determine χ_p we need to know the orbital contribution and a lesser diamagnetic term χ_{dia} . If we assume $\chi_0 = \chi_p + \chi_{\text{orb}} + \chi_{\text{dia}} = 0.123 \text{ emu/mole}$ and if $\chi_{\text{orb}} + \chi_{\text{dia}}$ is of the order as that found for UAl_2 , $\sim 10^{-3} \text{ emu/mole}$, then $\chi_p = 0.122 \text{ emu/mole}$. For a reasonable $\lambda = 0.3-0.5$, thus from Eq. (6) $S = 136$, which is about twice as large as for ZrZn_2 [25]. We note that $1/S = 1 - N(\epsilon_F)I$ where I is the effective exchange parameter. Using Wohlfarth's expressions [23] for the first and second derivative of the density of states (for parabolic bands), and assuming that ϵ_F lies at a maximum in the density of states we find for the second derivative $v_2 = d^2N(\epsilon_F)/d\epsilon^2 = -958 \text{ ev}^{-2}$ and the half-width of a Lorentzian shaped band at half maximum is $\Gamma = \sqrt{2} (-v_2)^{-1/2} = 0.046 \text{ ev}$. Thus, there are about three electrons in the half-filled, high density of states band.

DISCUSSION

The materials discussed above have several similarities among them. Each has a very large electronic specific heat coefficient [ranging from γ 's of $88-242 \text{ mJ}/(\text{mole-K}^2)$] corresponding to a very high $N(\epsilon_F)$. Each one has its scaling or ordering temperature (T_{sf} , T_N , or T_C) small enough to permit easy observation of the phenomena via specific heat measurements.

Thus, our attempts to see the low temperature upturn in the proposed spin fluctuation compounds NpRh_3 ($T_{\text{sf}} \sim 100\text{K}$) [9] and NpRu_2 ($T_{\text{sf}} \sim 65\text{K}$) [7] have not been successful. These high T_{sf} 's merely cause the $T^2 \log T$ term in Eq. (1) to appear as a modified phonon contribution at low temperatures. Thus, for NpRu_2 , we cannot differentiate between no spin fluctuation contribution, a Debye temperature $\Theta_D = 174\text{K}$, and $\gamma = 118 \text{ mJ}/(\text{mole-K}^2)$ or a $T_{\text{sf}} = 65\text{K}$, $\Theta_D = 163\text{K}$, and $\gamma = 84 \text{ mJ}/(\text{mole-K}^2)$ [7]. Presumably, this could be clarified by an elastic constant determination of Θ_D and by more accurate specific heat data.

The agreement of the $\text{U}_{1-x}\text{Th}_x\text{Al}_2$ data with the model of Fulde and Luther is very interesting. It shows that in our pure UAl_2 $T_i \ll T_{\text{sf}}$, and the data are not strongly modified by the scattering centers. It is possible to calculate T_i from a free-electron model and expressions in Ref. 14. If we use a bare γ of $80 \text{ mJ}/(\text{mole-K}^2)$ and take $m^*/m = \gamma/\gamma_{\text{free}}$ as the free electron calculated value, we find $m^*/m \approx 100$ and $T_i \approx 1.0\text{K}$. The latter approach has yielded T_i 's for UAl_2 - ThAl_2 in good agreement with the values needed to fit the C data (see Table I).

Another example of the great usefulness of a low ordering temperature is for NpSn_3 with $T_N = 9.5\text{K}$. Unlike the case for Cr where $T_N = 312\text{K}$, we can easily separate the phonon contribution, which only makes up about 16% of the total specific heat at T_N . Thus, a comparison of BCS theory [19] to the data shown in Fig. 5 cannot be made as unambiguously on the few other IAF systems known to date. Better tests of the theory of IAF will require better samples than have been prepared thus far. To that end, attempts will be underway soon to grow single crystals of NpSn_3 . It should be noted that NpSn_3 is one of those actinide compounds whose magnetic properties could not be predicted from Hill's criterion [8]. The Np-Np spacing of $4.63 \text{ \AA}$ is much larger than would be expected for 5f broadening

due to direct f-f wave function overlap. The same is true of USn_3 and PuSn_3 which do not order magnetically. Thus, the Hill criterion are useful for predicting magnetism in those actinide systems where f-f overlap is the dominant mechanism, but cannot address itself to the more complex mechanism of f-(s,p,d) mixing.

Although NpOs_2 has too large a moment to allow the rigorous application of the Stoner-Wohlfarth-Edwards theory, the results do fit the theory reasonably well.

Finally, it should be noted that in addition to the three compounds discussed here many other compounds seem to be likely candidates for band magnetism. Reference 9 lists many likely spin fluctuation compounds. From our preliminary specific heat measurements and some other known properties [6] it seems that NpIr_2 may also be an IAF. PuPt_2 , NpMn_2 , and perhaps NpNi_2 will probably prove to have a major itinerant component to their ferromagnetism. Thus, effort is continuing on these actinide materials and hopefully future work will yield additional interesting results.

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Table I. Calculated Values of T_i for $U_{1-x}Th_xAl_2$

x	T_i (C) ^(a) (°K)	T_i (ρ) ^(b) (°K)
0.00	0.0 ^(c)	0.9
0.02	2.8	1.3
0.05	4.0	3.1
0.10	6.0	4.2

- (a) From a fit of the specific heat data.
- (b) From the mean free path via the resistivity data.
- (c) A value of $T_i=0$ was used in the fit of the data to Eq. (3).

FIGURE CAPTIONS

- Fig. 1. Magnetic susceptibility of UAl_2 . Inset shows the data below 10K as a function of T^2 .
- Fig. 2. Specific heat of UAl_2 below 4.5K as a function of applied magnetic field. The solid line represents the zero-field data.
- Fig. 3. C/T vs T^2 for $U_{1-x}Th_xAl_2$ below 9K.
- Fig. 4. Magnetic susceptibility of $NpSn_3$ between 3-60K. The open circles are for an arc-melted sample and the solid points are for a sample prepared by reaction at 870K.
- Fig. 5. Electronic specific heat of $NpSn_3$ plotted as C_E/T vs T . The solid curve is the mean-field, BCS prediction.
- Fig. 6. C/T vs T for $NpOs_2$ below 20K. The solid curve is the estimated phonon contribution plus γ_p .
- Fig. 7. M^2 at $H/M = 0$ for $NpOs_2$ vs T^2 .