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TITLE: MAGNETIC PROPERTIES AND ELECTRONIC STRUCTURE OF THE  
ACTINIDE SYSTEMS  $Uf_2$ ,  $NpA_2$ , and  $PuA_2$

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**Magnetic Properties and Electronic  
Structure and the Actinide Systems  
 $UA\ell_2$ ,  $NpA\ell_2$ , and  $PuA\ell_2$**

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**Abstract**

The electronic ground state of  $UA\ell_2$ ,  $NpA\ell_2$  and  $PuA\ell_2$  have been determined by band theory techniques. These calculations indicate that both actinide d-f hybridization and spin-orbit coupling are important in these systems. Using a new scheme for obtaining the paramagnon parameters for  $UA\ell_2$  it is shown that calculated and experimentally derived  $5f$  band width are consistent. The fact that orbital angular momentum may not be quenched in these systems is suggested as a possible mechanism for damping spin-fluctuations in  $NpA\ell_2$  and  $PuA\ell_2$ .

### Introduction

The anomalous properties of the lighter actinides (Th-Pu) compounds are due to the 5f electron states in these systems. In many actinide compounds the 5f electrons exhibit localized behavior while in others they tend to behave as band states. A rationale for this variety of behavior was given by Hill [1] in terms of a correlation between magnetic properties and actinide-actinide internuclear separation. Although Hill obtained a good correlation his criteria does not explain why  $UA\varrho_2$ ,  $NpA\varrho_2$  and  $PuA\varrho_2$ , with almost equal actinide-actinide separations, have such different magnetic properties at low temperatures.

$UA\varrho_2$  is a spin-fluctuating material with a  $T^3 \ln T$  term in the low temperature specific heat data [2], while both  $NpA\varrho_2$  [3] and  $PuA\varrho_2$  [4] order magnetically. However it is unclear whether or not the low temperature behavior of  $NpA\varrho_2$  and  $PuA\varrho_2$  is band or local moment magnetism. The spin-fluctuation properties of  $UA\varrho_2$  have been interpreted [5] in terms of non-local (band) states and the paramagnon theory leads to narrower 5f bands (except for the newly discovered heavy-fermion systems [6]) than have ever been observed or calculated in metallic systems. Another argument for band states for the 5f electrons in  $UA\varrho_2$ ,  $NpA\varrho_2$ , and  $PuA\varrho_2$  is that they

participate in bonding and that this increased bonding yields the observed smaller lattice constants [7]. In order to obtain more information on these 5f states we have performed a systematic study of the electronic structure of these material using band theory [8]. From these results as well as from a new analysis of the paramagnon model and a recent calculation by Brooks and Kelly [9] on the orbital contribution from itinerant states, a new interpretation of the magnetic properties of these systems is given.

Table 1

| Compound               | UAl <sub>2</sub> | NpAl <sub>2</sub> | PuAl <sub>2</sub> |
|------------------------|------------------|-------------------|-------------------|
| 5f-band width          | 2.0              | 1.8               | 1.3               |
| occupied 5f band width | 0.14             | 0.39              | 0.29              |
| 5f occupation          | 2.25             | 3.81              | 5.17              |
| 6d occupation          | 1.95             | 1.61              | 1.41              |
| DOS ( $E_F$ )          | 7.1              | 7.5               | 9.4               |

Table 1. All bandwidths in eV. Density of states in units of states/eV cell.

#### Band Calculations

The band calculations were performed using a scalar relativistic linear augmented-plane-wave method with a warped muffin-tin potential to obtain the self-consistent electronic structure. The spin-orbit coupling was determined non-self-consistently using the scalar relativistic self-

consistent potential. The details of these calculations have been given elsewhere [8]. Here we give the results along with a brief discussion of these results. In Table 1 the total 5f-band width, the occupied 5f-band width, the density of states (DOS) at the Fermi energy ( $E_F$ ), and the occupation numbers for the 5f and 6d electrons are given for each system. As can be seen, the 5f band narrows from 2.0 eV in  $UA\varrho_2$  to 1.3 eV in  $PuA\varrho_2$ . Also the occupation numbers have a smooth variation; the number of 6d electrons decrease from U to Pu while the number of 5f electrons increase upon substituting Np and Pu for U. However, notice that the occupied 5f-band width is largest for  $NpA\varrho_2$ . In Fig. 1 the density of states (DOS) for  $UA\varrho_2$  is plotted. The calculated DOS functions for  $NpA\varrho_2$  and  $PuA\varrho_2$  are similar. The details of the differences between the DOS functions of these systems have been given in ref. 8. The sharp features at and above  $E_F$  are due to the spin-orbit split 5f bands. Although the detailed structure is different and there is some d-f hybridization in these systems, a crude picture of these systems is that of filling the spin-orbit split 5f band in going from  $UA\varrho_2$  to  $PuA\varrho_2$ . Therefore the different magnetic properties are evolving from a similar electronic structure.

### Paramagnon model

Having established that the electronic structure of all three systems is similar we now turn to the behavior of  $UA\varrho_2$ . Because of the  $T^3 \ln T$  behavior of the low temperature specific heat this system was fit to a paramagnon model [5]. This analysis showed that the occupied 5f-band had a width of around  $\sim 5 \times 10^{-3}$  eV. This differs by an order of magnitude from the band results given above. However this analysis was incomplete in that it contained one undetermined parameter. It was shown in ref. 8 that all the paramagnon parameters could be determined from experimental data. While most of the parameters in the new analysis are similar to the old values, i.e., a spin-fluctuation temperature ( $T_{SF}$ ) of around 40°K and a Stoner enhancement factor (S) of around 10, the Fermi temperature is different and yields an occupied 5f-band width of  $\sim 5 \times 10^{-2}$  eV. This means that the calculated electronic structure and that inferred from experiment are now consistent. Therefore the narrow 5f band in  $UA\varrho_2$  leads to spin-fluctuation, and now consideration must be given to why the similar electronic structure in  $NpA\varrho_2$  and  $PuA\varrho_2$  does not lead to the same phenomena.

### Orbital content of band states

One underlying assumption in the electronic structure picture of the actinide compounds is that the angular momentum is quenched just as it is in the transition metal compounds. Recently Brooks and Kelly [9] have given the argument that due to the spin-orbit interaction the spin drags an orbital contribution along and that angular momentum is not quenched in the actinide systems. He was also able to calculate the orbital contribution and showed that the results were more consistent with experimental data than those which assumed no orbital contribution. The implications this has for  $UA\ell_2$ ,  $NpA\ell_2$ , and  $PuA\ell_2$  is the following: Going across this series the spin-orbit interaction as well as the number of f electrons increases by a factor of 2. Therefore the orbital contribution to the angular momentum can change significantly from  $UA\ell_2$  to  $PuA\ell_2$  and raises the possibility that the orbital contribution can damp the spin-fluctuation excitations.

### Conclusion

Although there are differences in the details of the one-electron electronic structure of these three systems, their overall structure is similar and in fact they can be viewed as the same system,

with just an increased filling the  $5f_{5/2}$  band in going from U to Pu. It was also shown that the calculated electronic structure of  $UA\ell_2$  is consistent with its spin-fluctuation behavior. Finally it was argued that the orbital angular momentum may not be quenched in these systems and that this may be the mechanism that kills spin-fluctuations in  $NpA\ell_2$  and  $PuA\ell_2$ . While this gives a reasonable rationale for the behavior of these systems, we have not proved that it is the correct explanation. The fine details of the differences in the electronic structure in these systems for, example, may be equally important.



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Figure Caption

Figure 1 Total densities of state function for  $UAl_2$  in states/eV-cell.

