CONF

MASTER

DISCLAIMER

CONF-810809--18

MAGNETIC SUSCEPTIBILITY OF CURIUM PNICTIDES*

S. E. Nave, P. G. Huray, J. R. Peterson, D. A. Damien,** and R. G. Haire

Transuranium Research Lab., (Chemistry Div.) Oak Ridge National Laboratory
Oak Ridge, Tennessee 37830 and The University of Tennessee,
Knoxville, Tennessee 37916

The magnetic susceptibility of microgram quantities of ^{248}CmP and ^{248}CmSb has been determined with the use of a SQUID micromagnetic susceptometer over the temperature range 4.2-340 K and in the applied magnetic field range of 0.45-1600 G. The fcc (NaCl-type) samples yield magnetic transitions at 73 K and 162 K for the phosphide and antimonide, respectively. Together, with published magnetic data for CmN and CmAs, these results indicate spatially extended exchange interactions between the relatively localized 5f electrons of the metallic actinide atoms.

* Research sponsored by the Div. of Chemical Sciences, Office of Basic Energy Sciences, U.S.D.O.E. under contracts DE-AS05-79ER10348 and DE-AS05-76ER04447 with the University of Tennessee (Knoxville) and W-7405-eng-26 with Union Carbide Corporation.

**Present Address: CEN-FAR, France.

Localization and interatomic Coulomb correlations are of considerable consequence in determining the magnetic properties of the 5f electrons [1]. In the lighter actinides (Th through Pu) the overlap of the 5f electrons produces a significant bandwidth and hybridization with the 6d-7s-7p bands requiring an itinerant or delocalized description. In the transplutonium actinides increasing spatial localization produces smaller overlap.

The degree of the 5f overlap is assumed to depend upon the An-An internuclear separation which may be altered through the use of a series of binary compounds. One such series is provided by the actinide-monopnictides (N, P, As, Sb and Bi) which exhibit [1, 2] fcc (NaCl-type) structures in which there is an increasing An-An distance with the heavier pnictogen. Uranium and neptunium monopnictides have been found to exhibit magnetic transitions with transition temperatures which increase monotonically with An-An distance [1]. This tendency is contrary to an expected behavior of a direct dipole-dipole or overlap exchange mechanism and suggests an RKKY or superexchange interaction via the pnictogen. The plutonium monopnictides exhibit magnetic transitions, the temperatures of which increase at first with the An-An separation, reach a maximum near 4A, and then decrease. Americium nominally has a 5f electronic structure which is non-magnetic based on the L-S coupling and Hund's rule. Measurements of the magnetic susceptibility of the metal, AmN and AmAs [3] have shown little temperature dependence and are consistent with an electronic arrangement exhibiting only a small Curie-Weiss magnetic moment.

Kanellakopulas has measured the magnetic properties of ^{244}CmN and ^{244}CmAs [3] and has found a T_c=109 K with μ_{eff}=7.02 μ_B for the nitride and a T_c=88 K with μ_{eff}=6.58 μ_B for the arsenide. A study of dhcp curium metal using the longer-lived ^{248}Cm [4] has shown that it exhibits an antiferromagnetic transition with T_N=65 K. The fcc metallic phase of ^{248}Cm exhibited a ferrimagnetic transition at T_N=205 K with two apparent moment sites having μ_{eff}=5.73 μ_B and μ_{eff}=6.53 μ_B.

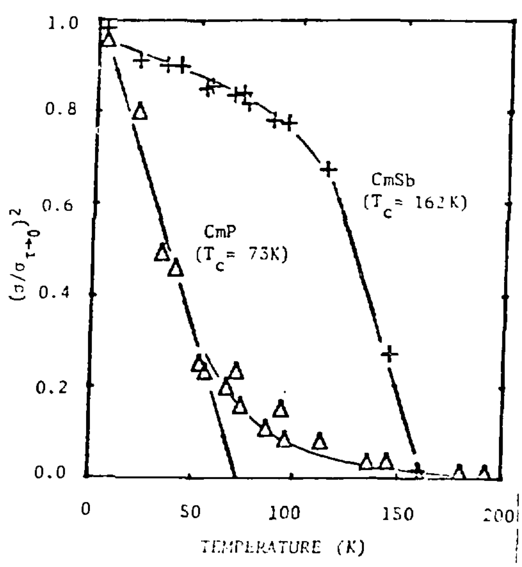

Fig. 1 : Square of the magnetic moment density of the curium pnictides (normalized to 0 K) as a function of temperature.

We have more recently used our SQUID micro-magnetic susceptometer [5] to determine the temperature dependence of ^{248}CmP and ^{248}CmSb. Both are found to exhibit a high temperature Curie-Weiss behavior with a magnetic transition at low temperature. The results are shown in Fig. 1 for an applied field of 1601 G. Here the square of the magnetic moment (normalized to the extrapolated 0K value) is plotted as a function of temperature to identify more clearly the Curie temperature T_c. It may be seen from this figure that for CmP, T_c=73 K and that for CmSb, T_c=162 K.

These results combined with values for CmN and CmAs are plotted as a function of An-An separation in Fig. 2. Closed symbols are used for ferromagnetic transition temperatures, T_c and open symbols are used for antiferromagnetic transition temperatures, T_N.

It may be seen from this figure that in contrast to the lighter actinides, the Cm transition temperatures yield a minimum within the range of An-An spacings provided by the monopnictide compounds. The more tightly bound, localized, high magnetic moment, 5f wave function of the heavier Cm will thus likely require extended exchange mechanisms like the RKKY type or superexchange with the pnictogen to explain this observation. Even though the RKKY interactions must take into account modified Fermi surfaces for the specific compounds, it is difficult to see how such a picture can be consistent with this experimental trend without a change of magnetic type. It is interesting to note, for example, that the lanthanide 4f homologue of Cm, which is Gd, forms a series of monopnictides which have transition temperature of 72K, 15K, 25K, and 28K for the compounds N, P, As and Sb, respectively. However, in the Gd series the first compound is ferromagnetic and the others are antiferromagnetic.

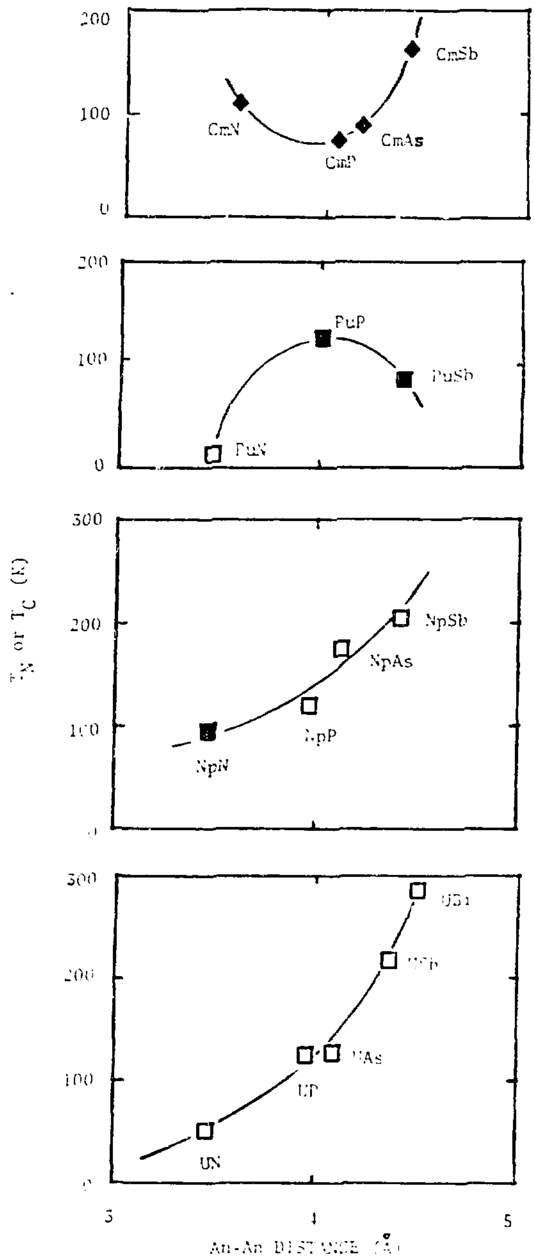

Fig. 2 : Magnetic transition temperature for the actinide monopnictides as a function of the An-An separation.

[1] D. J. Lam and A. T. Aldred, The Actinides: Electronic Structure and Related Properties. Academic Press, Vol. 1 (1974) p. 109-179.
[2] D. Damien, R. G. Haire, and J. R. Peterson, Journal of the Less-Common Metals 68 (1979) 159-65.
[3] B. Kanellakopulos, J. P. Charvillat, F. Maino, and W. Muller, Transplutonium Elements, North-Holland (1976) p. 181-189.
[4] P. G. Huray, S. E. Nave, J. R. Peterson and R. G. Haire, Physica 102B (1980) 217-220.
[5] S. E. Nave and P. G. Huray, Rev. Sci. Instr. 51(5), (1980) 7-12.