CONF-771127--15

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Prepared for

23rd Conference on Magnetism and Magnetic Materials
November 8-11, 1977
Minneapolis, MN

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Operated under Contract W-31-109-Eng-38 for the

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MAGNETIC SUSCEPTIBILITY OF $AnRe_2$ COMPOUNDS*

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ABSTRACT

The compounds $AnRe_2$, where An = Th, U, Np, and Pu all have the $MgZn_2$-type hexagonal Laves phase structure. Measurements of their magnetic susceptibilities have been made from 2-300 K. Whereas $ThRe_2$, URe_2, and $PuRe_2$ have essentially temperature-independent susceptibilities, $NpRe_2$ is ferromagnetic with T_c = 47 K. The paramagnetic data for $NpRe_2$ follow a modified Curie-Weiss law, $\chi - \chi_o = C/(T - \Theta)$ from 60-230 K with Θ = 46.9 K and C equivalent to an effective moment, p_{eff} = 2.11 μ_B. The ordered moment μ_o = 0.90 μ_B and the results are consistent with other actinide compounds having good local moment behavior. The electronic specific heat of URe_2 = 34 mJ/(mole-K^2) is large enough to indicate the possibility of spin-fluctuation behavior in this compound.

INTRODUCTION

Many actinide intermetallic compounds with composition AnX_2 form Laves phase-type structures. The largest number of these occur in the $MgCu_2$-type cubic phase structure. Systematic studies of the magnetic properties of $MgCu_2$-type compounds have been made, and reasonable correlations between magnetic ordering behavior and An-An separation have been presented for cases where X is not a 3*d* transition element [1]. Although a number of An Laves-phase compounds also form in the hexagonal $MgZn_2$ structure, magnetic data have been reported only for UNi_2, which orders ferromagnetically at T_c = 30 K and μ_o = 0.13 μ_B [2]. This paper presents the results of a magnetization study of $AnRe_2$ compounds where An = Th, U, Np and Pu. All of these compounds form the $MgZn_2$ structure. Although URe_2 does form in the $MgZn_2$ structure above 453 K, below that temperature it transforms to an orthorhombic structure which is a distortion of the $MgZn_2$ structure at lower temperatures. In view of the similarity of the low temperature structure to the other $AnRe_2$ structures, the results for URe_2 are also included.

RESULTS

Figure 1 shows the temperature dependence of the molar susceptibility for the compounds between 2-300 K.

The susceptibilities are essentially temperature-independent for $ThRe_2$, URe_2 and $PuRe_2$. However, the data for $NpRe_2$ are indicative of ferromagnetic ordering, and these results are replotted as $1/\chi$ versus T in Fig. 2. The results nearly fit a Curie-Weiss law. The curvature may be due to a temperature-independent term, χ_o, or to crystal-field effects. Both approaches have been used previously to explain curvature in $1/\chi$ versus T plots for actinides. In the absence of a crystal-field calculation for this structure, no attempt is made here to evaluate the crystal-field effects. Figure 2 also shows the effect of assuming $\chi_o = 1.21 \times 10^{-3}$ emu/mole and a straight line is obtained.

The modified Curie-Weiss law, $\chi - \chi_o = C/(T - \Theta)$, for $NpRe_2$ yields $\Theta = +47.4$ K and $p_{eff} = 2.11\ \mu_B$. The existence of ferromagnetic ordering at 47 K is supported further by the magnetization data in the vicinity of T_c. These are plotted in Fig. 3 as σ^2 versus H/σ and yield $T_c = 47$ K.

DISCUSSION

The $NpRe_2$ data in Fig. 2 are strongly indicative of local moment behavior. Below T_c, plots of σ versus $T^{3/2}$, σ versus T^2 and a Brillouin function for $J = 1/2$ all yield $\mu_o = 0.899 \pm .002\ \mu_B$ upon extrapolation to 0 K. Simple Russell-Saunders coupling would indicate $0.84\ \mu_B$ and $2.70\ \mu_B$ for p_{eff} of $5f^5$ and $5f^4$ configurations, respectively. In the well documented case of NaCl-type actinide compounds [3] intermediate coupling which involves full j-j manifold interactions, the effective moments are 1.41 and 2.59 for $5f^5$ and $5f^4$, respectively. The ratio $\mu_o/p_{eff} = 0.44$ for $NpRe_2$ is in the range found for NaCl-type compounds. These considerations are very qualitative in the absence of crystal-field calculations for actinides with the $MgZn_2$ structure. Whereas no attempt will be made to use the results to assign a ground state configuration to the Np atom, the results are all suggestive of good $5f$ local moment character in $NpRe_2$. This is in contrast to $NpOs_2$ (cubic Laves phase) which is an itinerant ferromagnet [4,5].

The small break in the data for $NpRe_2$ at 250 K (best seen in the $1/\Delta\chi$ versus T plot in Fig. 2) may be due to a lattice distortion of the type found in URe_2 at 453 K. Similar effects are found in the susceptibility data for the cubic Laves phase compounds $NpOs_2$, $NpRu_2$ and $NpIr_2$ [4]. The room temperature x-ray diffraction pattern for the URe_2 sample used here showed the presence of both the hexagonal Laves phase (major) and the low-temperature orthorhombic phase (minor). However, the absence of a temperature-dependent susceptibility or of a strongly temperature-dependent magnetic

impurity contribution indicate that both URe_2 structures are nonmagnetic.

The strong magnetism of the Np compound, in comparison to the U and Pu compounds, is not uncommon. Consider the sequence $ThAl_2$, UAl_2, $NpAl_2$ and $PuAl_2$. $ThAl_2$ is nonmagnetic; UAl_2 is a spin fluctuation compound [6]; $NpAl_2$ is a good ferromagnet with T_c = 56 K [4]; and $PuAl_2$ may be magnetic but only below 4 K [7]. Similarly, USn_3 is likely to be a spin fluctuation compound [8]; $NpSn_3$ is an itinerant antiferromagnet [9]; and $PuSn_3$ is nonmagnetic [8]. These results may be understood in terms of a strong spin-orbit splitting. When that splitting is larger than the 5*f* band-width, the 5*f* band splits into j = 5/2 and j = 7/2 sub-bands [10]. This approach can explain the lack of magnetism in Am, as well as the stronger magnetism for Np versus U and Pu in some cases.

Most certainly $ThRe_2$ is nonmagnetic and a superconductor due to the lack of 5*f* electrons below the Fermi level [11]. Despite the temperature-independent paramagnetism of URe_2 and $PuRe_2$, they may still exhibit spin-fluctuation behavior. The specific heat of the URe_2 sample measured at low temperatures yields an electronic term $\gamma = 34$ mJ/(mole-K^2). It is observed that all actinide compounds having $\gamma \geq 20$ mJ/(mole-K^2) are magnetic or show spin fluctuation behavior. The simplest way to show this would be via a resistivity $\propto T^2$ at low temperatures. However, the two-phase nature of URe_2 samples currently available makes this approach suspect. The possibility of spin fluctuation effects may be tested in $PuRe_2$, which is single phased. However, the lower susceptibility of $PuRe_2$, $[\chi(PuRe_2)/\chi(URe_2)] = 0.5$, makes it a less likely candidate for spin fluctuations than is URe_2.

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*Work supported by the U.S. Energy Research and Development Administration.

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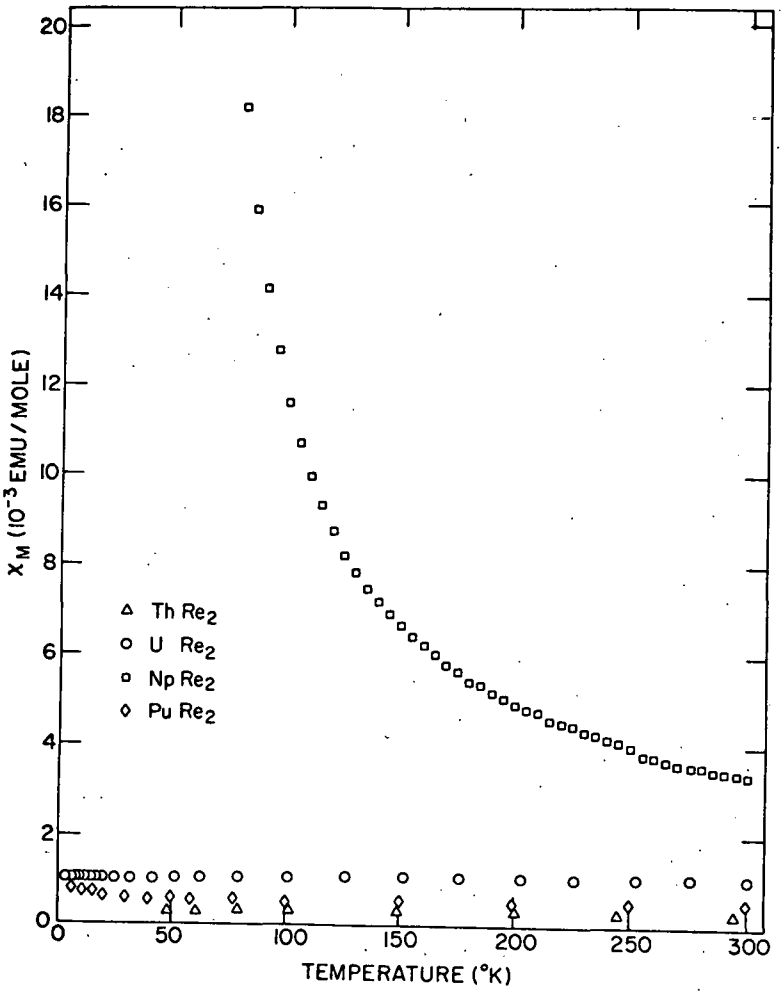

Fig. 1. Molar susceptibilities vs. temperature for $AnRe_2$ compounds.

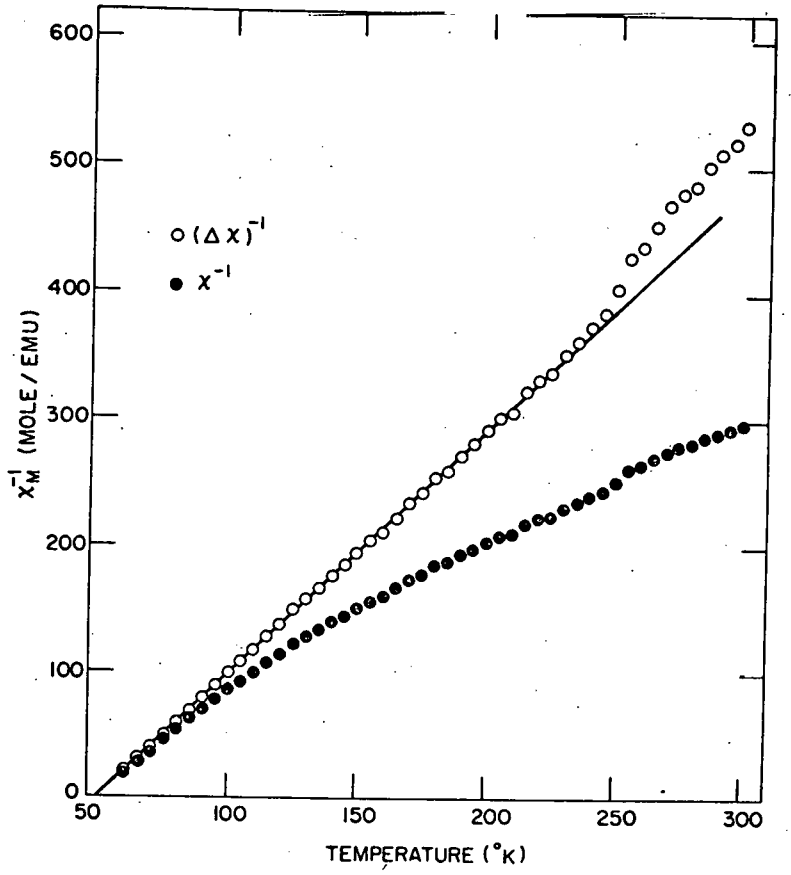

Fig. 2. Inverse molar susceptibility and corrected susceptibility vs. temperature for $NpRe_2$.

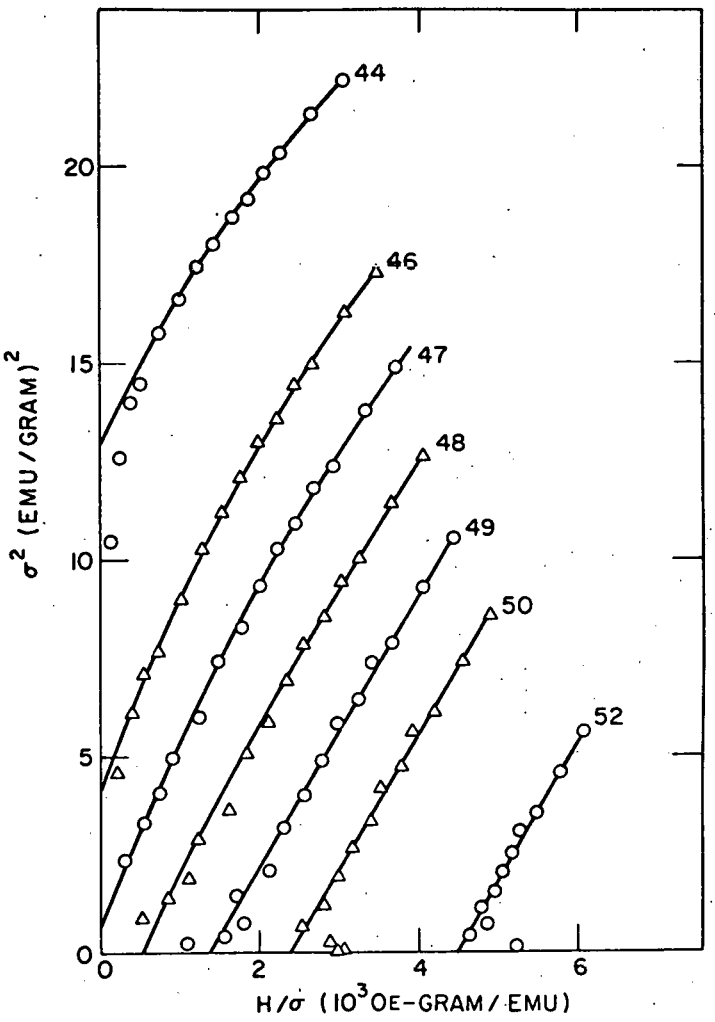

Fig. 3. Arrott plots for $NpRe_2$.