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Magnetic properties of the $U_{1-x}Gd_xNi_2$ system

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Abstract

The magnetic properties of $U_{1-x}Gd_xNi_2$ systems in the 4–600 K temperature range and fields up to 8 T were studied. UNi_2 is a weak itinerant ferromagnet; uranium shows a small magnetic contribution, and nickel in this system is not magnetic. Replacing gadolinium for uranium leads to an increase of spontaneous magnetisation and Curie temperatures up to 7 μ_B/f.u. and 86 K, respectively. Above the transition temperatures the systems show a Curie–Weiss-type paramagnetism. The effective magnetic moment per gadolinium atom in all composition ranges is close to that of gadolinium free ion. The magnetic properties were correlated with the structural data. © 1998 Elsevier Science S.A.

Keywords: Magnetisation; Susceptibility; Ferromagnetism; Curie temperature; Magnetic moment

1. Introduction

The UNi_2 compound crystallises in a hexagonal C-14 type structure with lattice parameters $a=4.966$ Å and $c=8.25$ Å. The system is a weak itinerant ferromagnet with the Curie temperature $T_C=23$ K [1,2]. The neutron diffraction studies [3] evidenced a very low spontaneous moment of 0.08–0.09 μ_B per uranium atom. The magnetic contribution of nickel may be neglected. The $GdNi_2$ compound has a cubic symmetry of $MgCu_2$ type with a lattice constant of 7.175 Å [4]. This compound is a ferromagnet having $T_C\approx 86$ K and saturation magnetic moment 7 μ_B [4].

We analyse the effect of uranium substitution by gadolinium on the structural and magnetic properties of the $U_{1-x}Gd_xNi_2$ system.

2. Experimental details

The samples were melted in an arc furnace in a purified argon atmosphere. To ensure a good homogeneity, the samples were several times remelted. The alloys were thermally treated in vacuum for 1 week at 1000 K. The X-ray analyses show for $x\geq 0.2$ the presence of a single phase having cubic symmetry of $MgCu_2$ type. The crystalline structure is changing from hexagonal to cubic in the composition range $0<x<0.2$. The lattice constants of the cubic samples linearly increase as gadolinium content increases from 7.11 Å for $x=0.2$ up to 7.175 Å for $GdNi_2$.

The magnetic measurements were performed in the 4–600-K temperature range. The spontaneous magnetisation values were determined from the magnetisation isotherms. The M_S values were calculated according to the relation: $M=M_S(1-a/H)+\chi_0' H$. Here a represents the coefficient of magnetic hardness, χ_0' is the Pauli-type contribution, M_S is the measured magnetisation and H is the external magnetic field.

3. Results

The low temperature measurements performed on the $U_{1-x}Gd_xNi_2$ system evidenced ferromagnetic ordering, and allowed us to obtain the spontaneous magnetisation and the transition temperature, T_C, values. The spontaneous magnetisation values as a function of temperature are presented in Fig. 1. An increase of M_S and of the Curie temperatures as x increases is evidenced. The composition dependence of the spontaneous magnetisation per formula unit is plotted in Fig. 2. In the same figure it is shown that the increase of x leads to an increase of the magnetic moment per Gd atom as well as T_C values.

The susceptibility values were obtained from the field dependence of the measured χ_m values according to Honda–Owen rule: $\chi_m=\chi+cM_S' H^{-1}$ by extrapolating to $H^{-1}\to 0$. In the paramagnetic range, the systems with

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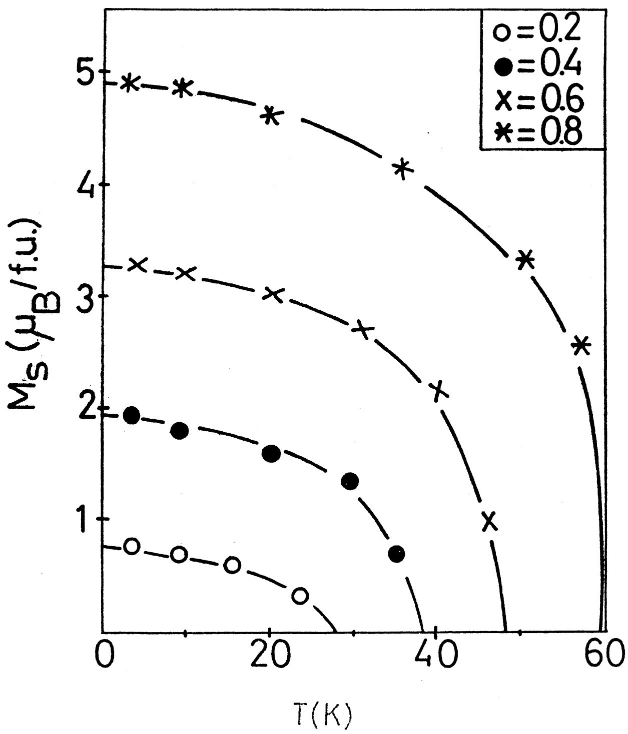

Fig. 1. The temperature dependence of spontaneous magnetisation for $U_{1-x}Gd_xNi_2$ systems.

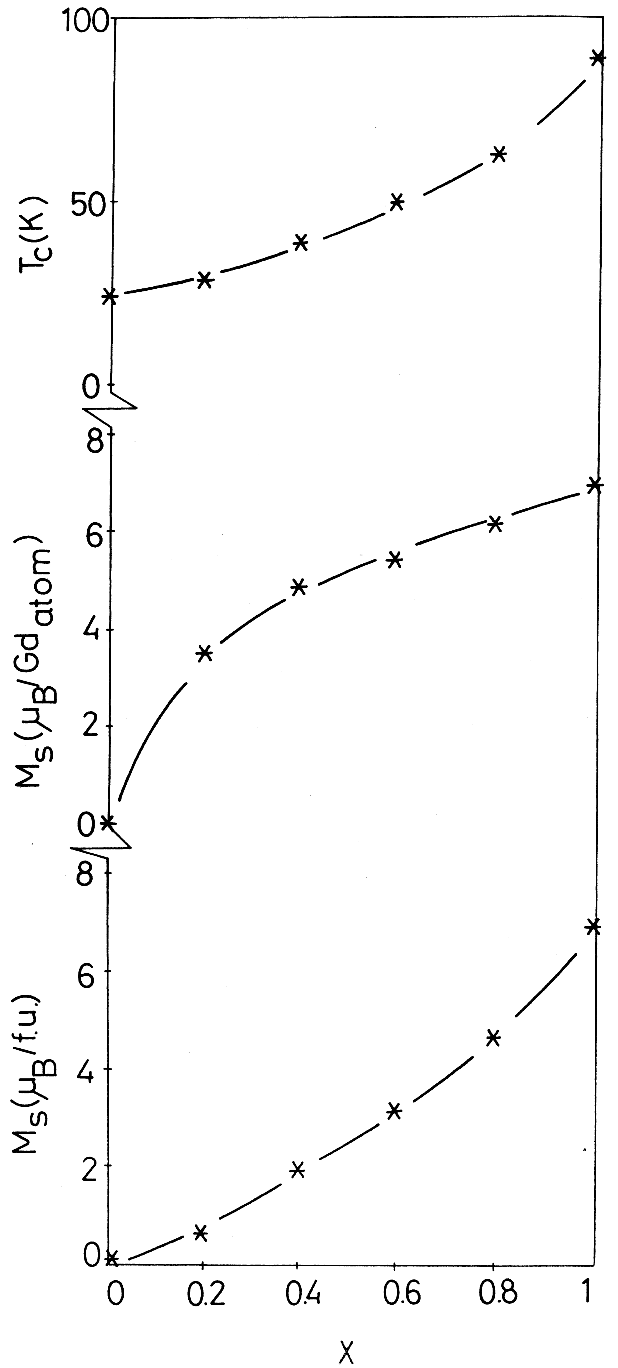

Fig. 2. The composition dependence of the spontaneous magnetisation per f.u. and gadolinium atom, respectively, and the transition temperatures, T_C.

$x \geq 0.2$ show a Curie–Weiss law, $\chi = C/(T-\theta)$, as is seen in Fig. 3. We denoted by χ the accurate values of the susceptibility, c is the assumed ferromagnetic impurity concentration and M'_S is its saturation magnetisation. The linear temperature dependence of the reciprocal susceptibility values is presented in Fig. 3. By fitting the experimental data, the molar Curie constants C and the paramagnetic Curie temperatures θ were determined. We present in Fig. 4 the composition dependence of θ values. The paramagnetic Curie temperatures increase as the substitution element content is increased, from ≈25 K corresponding to UNi_2 to ≈87 K, the value for $GdNi_2$. The molar Curie constants also increase as the gadolinium content increases; the effective magnetic moments μ_{eff} calculated per gadolinium atom has the constant value 8 μ_B in the whole composition range (Fig. 4).

4. Discussion

The UNi_2 compound is a weak itinerant ferromagnet. In the ordered phase the magnetic moments are due to uranium atoms, and they have very small values. Over T_C the susceptibility follows a modified Curie–Weiss law, where the temperature-independent term χ_0 is 10^{-3} emu mol^{-1} and represents the predominant term in the susceptibility; for $T > 200$ K the susceptibility becomes almost temperature independent.

The $U_{1-x}Gd_xNi_2$ systems present a cubic symmetry from $x \geq 0.2$. The spontaneous magnetisation per formula unit almost linearly increases with gadolinium content x up to 7 μ_B, the value reported for $GdNi_2$. Using the Hill criterion [5] for the magnetic ordering behaviour of uranium, the distances d_{U-U} between two nearest uranium neighbours are lower than the Hill limit (3.4–3.6 Å). These distances vary from 3.08 Å for $x=0.2$ to 3.09 Å for $x=0.8$, and indicate the 5f uranium electron itinerancy.

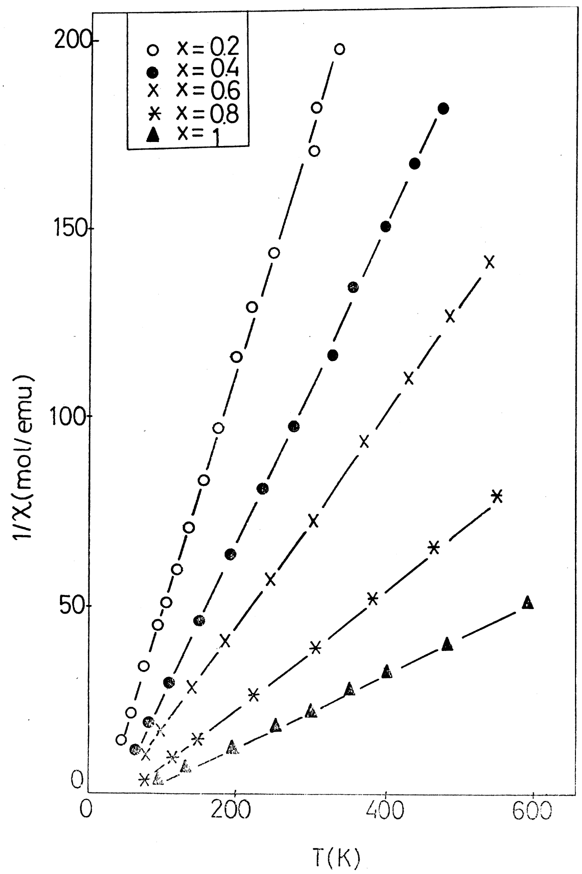

Fig. 3. The temperature dependence of the reciprocal susceptibilities for the $U_{1-x}Gd_xNi_2$ compounds.

The magnitudes of U magnetic moments are unaffected for all uranium concentrations with increasing dilution in other reported systems $U_{1-x}Gd_xGa_2$ [6]. The effective magnetic moment attributed to the gadolinium atom has the constant value 8 μ_B (the gadolinium free ion Gd^{3+}). The ratio $r=S_P/S_O$ between the number of spins obtained from the effective magnetic moments S_P and the saturation moments S_O, respectively, varies from 1.29 ($x=0.8$) to 2.66 ($x=0.2$). The increase in r values, as the T_C values decrease, proves a gradual increase of itinerancy degree. The increase of T_C and θ values when uranium is substituted by gadolinium proves the increase in exchange interactions between Gd atoms. A similar situation was reported for cobalt atoms in $xUCo_{5.3}(1-x)UNi_5$ [7].

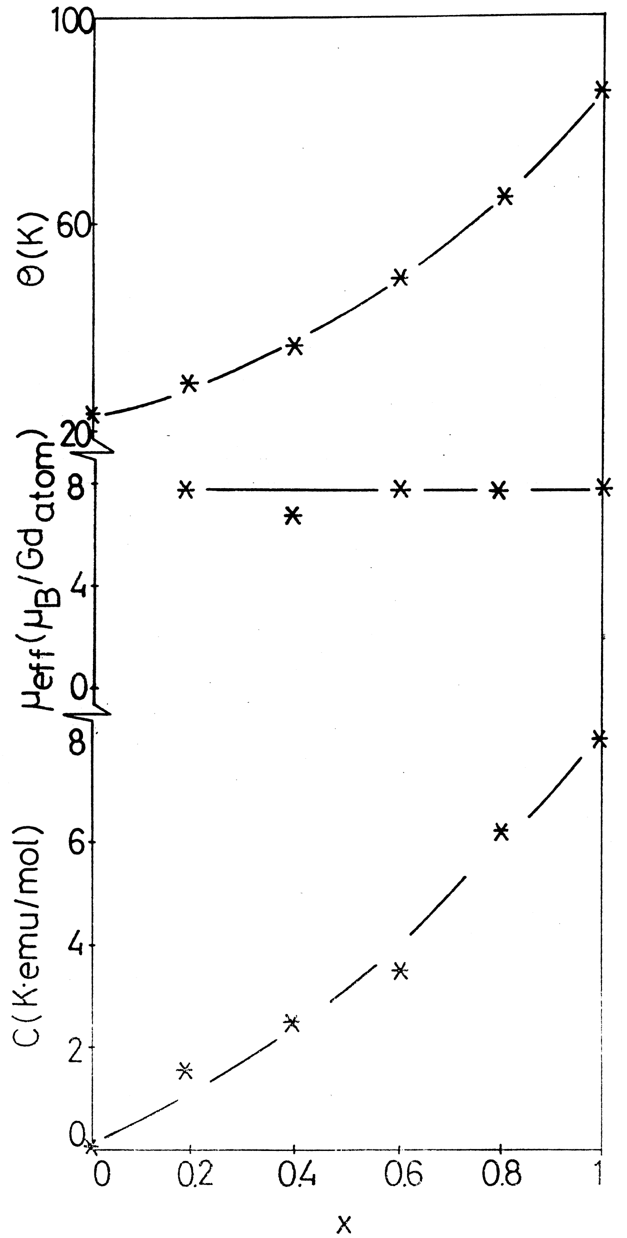

Fig. 4. The composition dependence of the molar Curie constants, the effective magnetic moments per gadolinium atom, and the paramagnetic Curie temperatures for the $U_{1-x}Gd_xNi_2$ compounds.

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