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Magnetic phase diagram of $NdRh_{2-x}Ru_xSi_2$ in high magnetic field

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Abstract

The high field magnetization and a.c. susceptibility of $NdRh_{2-x}Ru_xSi_2$ compounds were investigated up to 140 kOe. The *(H, T, x)* magnetic phase diagram was determined. Increases in the concentration x and magnetic field caused changes in the magnetic properties of the $NdRh_{2-x}Ru_xSi_2$ compounds.

1. Introduction

Ternary RT_2X_2 compounds (R ≡ rare earth; T ≡ "*n*d" transition element; X ≡ Si or Ge) with the tetragonal $ThCr_2Si_2$-type structure exhibit many interesting physical, particularly magnetic, properties [1, 2].

$NdRh_2Si_2$ and $NdRu_2Si_2$ crystallize in the same tetragonal structure of the $ThCr_2Si_2$ type but their magnetic properties are different [3, 4]. $NdRh_2Si_2$ orders antiferromagnetically (AF) below 53 K with a magnetic structure of the AFI type [5]. $NdRu_2Si_2$ displays a complicated magnetic behaviour. Below $T_N = 24$ K it develops a sine wave modulation of the magnetic moments which are along the c axis. At about 15 K a square magnetic structure occurs which persists down to 2.8 K. At $T < 10$ K a ferromagnetic order coexists with the square-modulated phase [6].

Magnetization curves of $NdRu_2Si_2$ measured along the c axis at various temperatures also indicate a complicated character of the magnetic properties. At 4.2 K the magnetization increases rapidly at very low field and almost saturates at 0.3 kOe [7]. The saturation moment is 2.8 μ_B/per formula unit, which is smaller than the theoretical Nd^{3+} moment ($gJ = 3.27$ μ_B). In the temperature range $10 < T < 15$ K the magnetization process exhibits a one-step metamagnetic transition at a very low critical field ($H_{c1} = 3$ kOe at 12 K). Above $T = 15$ K the one-step magnetization process changes to a two-step one. The critical fields are $H_{c1} = 5$ kOe and $H_{c2} = 7$ kOe at 16 K [7].

$NdRh_{2-x}Ru_xSi_2$ solid solutions exist in the whole region of concentration x. All compounds have the $ThCr_2Si_2$-type crystal structure. The magnetic properties depend on the ruthenium concentration. A collinear antiferromagnetic ordering of the AFI type for $x \leqslant 0.5$ changes via sine modulation to a ferromagnetic ordering with increasing x [8].

In this work the results of a.c. susceptibility and high field magnetization measurements on $NdRh_{2-x}Ru_xSi_2$ compounds are reported.

2. Experimental details and results

The experiments were carried out on polycrystalline samples as reported in a previous paper [8].

The a.c. susceptibility was measured using a mutual inductance bridge. The magnetization of the samples was measured by means of a vibrating sample magnetometer in high magnetic fields up to 140 kOe produced in a "solenoid" installation. Measurements in magnetic fields up to 50 kOe were made in a Foner magnetometer.

The temperature dependences of the a.c. and d.c. magnetic susceptibilities exhibit a maximum which is characteristic of a phase transition from the antiferro- to the paramagnetic state.

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The results of magnetization measurements on samples oriented in a magnetic field at different temperatures and in magnetic fields up to 140 kOe are presented below.

2.1. $NdRh_2Si_2$

Figure 1 shows the magnetization curves at various temperatures. At $T=4.2$ K a one-step metamagnetic transition with the critical field $H_c=140$ kOe is observed, The character of the magnetization curve and the value of the critical field are in agreement with data for single-crystal $NdRh_2Si_2$ [9]. The magnetization curve along the c axis measured in magnetic fields up to 400 kOe at $t=4.2$ K indicates a two-step process with the critical fields $H_{c1}=143$ kOe and $H_{c2}=187$ kOe. The upper critical field is above the maximal value of the fields applied in our experiments.

In the temperature range 4.2–32.7 K the magnetization process is similar to that observed at 4.2 K. An increase in temperature causes a decrease in critical field. In the magnetization curve measured at $T=42.4$ K two critical fields are observed at $H_{c1}=122$ kOe and $H_{c2}=134$ kOe. The magnetization curves measured at $T=45$ and 51 K have only one critical field. The critical fields are determined from the field dependence of the differential magnetization, dM/dH (see Fig. 2). The temperature dependences of the critical fields are shown in Fig. 3. From these data the magnetic phase diagram is determined.

2.2. $NdRh_{1.5}Ru_{0.5}Si_2$, $NdRh_{1.2}Ru_{0.8}Si_2$ and $NdRhRuSi_2$

The magnetization curves for these compounds have a one-step metamagnetic character (see Fig. 4). The critical fields determined at different temperatures are presented in Fig. 5. The critical field decreases with increasing ruthenium concentration. At $H=140$ kOe and $T=4.2$ K the magnetic moment is close to 2.4 μ_B which is similar to those observed for $NdRh_2Si_2$ [9] and $NdRu_2Si_2$ [7].

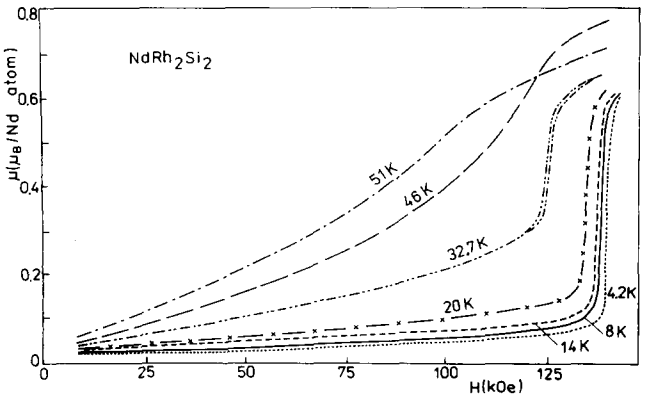

Fig. 1. High field magnetization curves at different temperatures for $NdRh_2Si_2$.

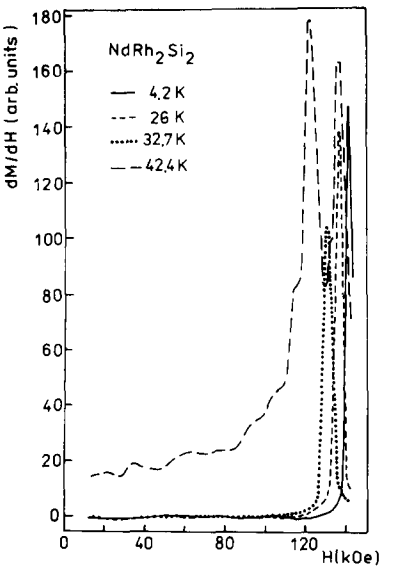

Fig. 2. Magnetic field dependence of the differential magnetization of $NdRh_2Si_2$ at different temperatures.

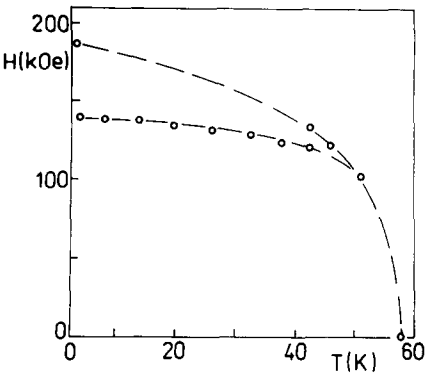

Fig. 3. Temperature dependence of the critical fields of $NdRh_2Si_2$.

2.3. $NdRh_{0.5}Ru_{1.5}Si_2$, $NdRh_{0.2}Ru_{1.8}Si_2$ and $NdRu_2Si_2$

The magnetization curves for these compounds are presented in Fig. 6. The magnetization of $NdRh_{0.5}Ru_{1.5}Si_2$ increases slowly with increasing magnetic field and saturates above 80 kOe. For $NdRh_{0.2}Ru_{1.8}Si_2$ at $t=4.2$ K the magnetization is saturated at $H\approx 10$ kOe. The observed hysteresis of the magnetization is typical for ferromagnetic materials.

The magnetization curves of $NdRu_2Si_2$ measured at different temperatures do not detect the metamagnetic phase transition observed for the $NdRu_2Si_2$ single crystal [7].

The temperature dependence of the magnetization of $NdRh_{0.5}Ru_{1.5}Si_2$ and $NdRh_{0.2}Ru_{1.8}Si_2$ (see Fig. 7) indicates two magnetic phase transitions: first, from the ferro- to the antiferromagnetic state at $T_c=16$ K for both samples; secondly, from the antiferro- to the paramagnetic state at $T_N=20.5$ K for $NdRh_{0.5}Ru_{1.5}Si_2$ and at $T_N=23.5$ K for $NdRh_{0.2}Ru_{1.8}Si_2$.

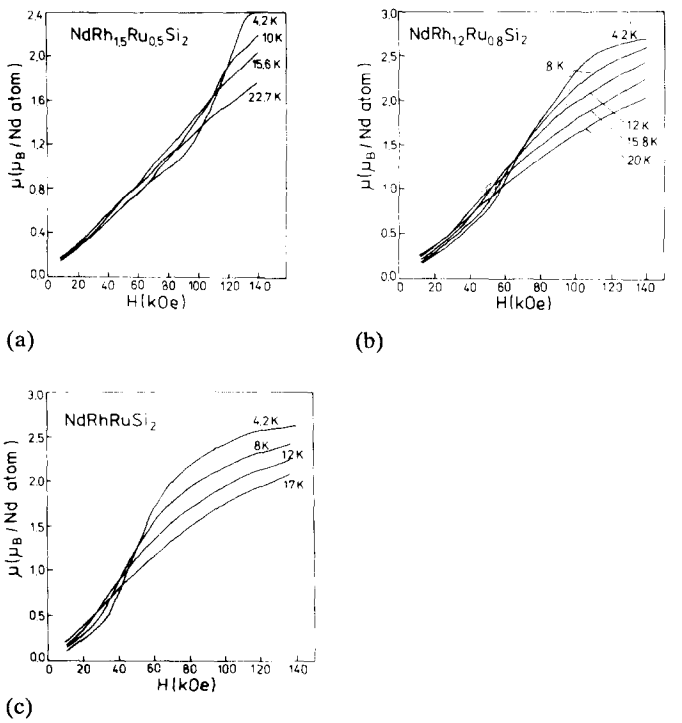

Fig. 4. High field magnetization curves at different temperatures for $NdRh_{1.5}Ru_{0.5}Si_2$, $NdRh_{1.2}Ru_{0.8}Si_2$ and $NdRhRuSi_2$.

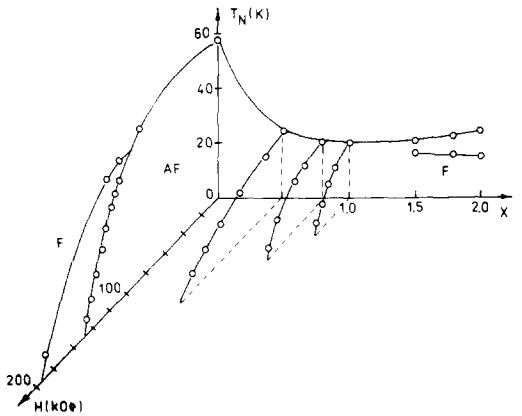

Fig. 5. The (H, T, x) magnetic phase diagram for the $NdRh_{2-x}Ru_xSi_2$ system.

3. Discussion

From the measurements of the $NdRh_{2-x}Ru_xSi_2$ system presented in this work, we obtained a phase diagram as a function of concentration x and magnetic field H as shown in Fig. 5. Depending on x, the phase diagram is divided into three regions. In the region $0 \leqslant x \leqslant 0.5$ a sharp decrease in the Néel temperature from 57.5 to 22 K is observed. In the region $0.5 < x < 1.5$ the Néel temperature is at first constant, then increases to 26.5 K for $NdRu_2Si_2$. For $x > 1.5$ in the low temperature region, ferromagnetic ordering is observed.

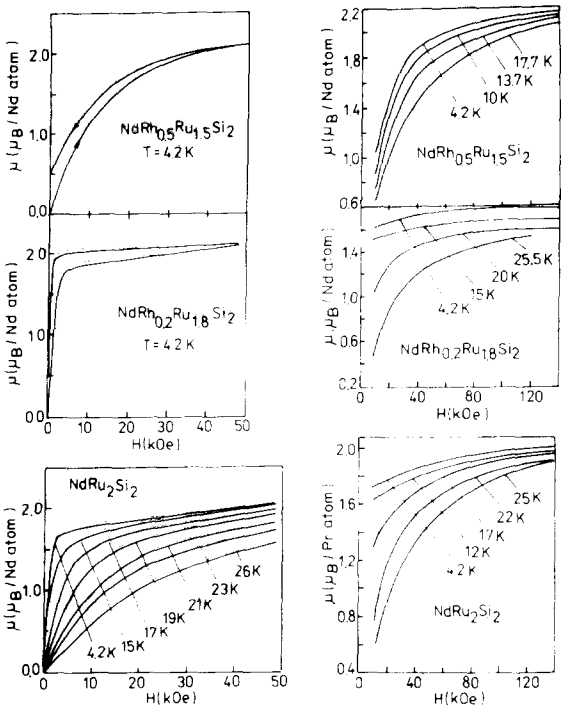

Fig. 6. Magnetization curves at different temperatures for $NdRh_{0.5}Ru_{1.5}Si_2$, $NdRh_{0.2}Ru_{1.8}Si_2$ and $NdRu_2Si_2$.

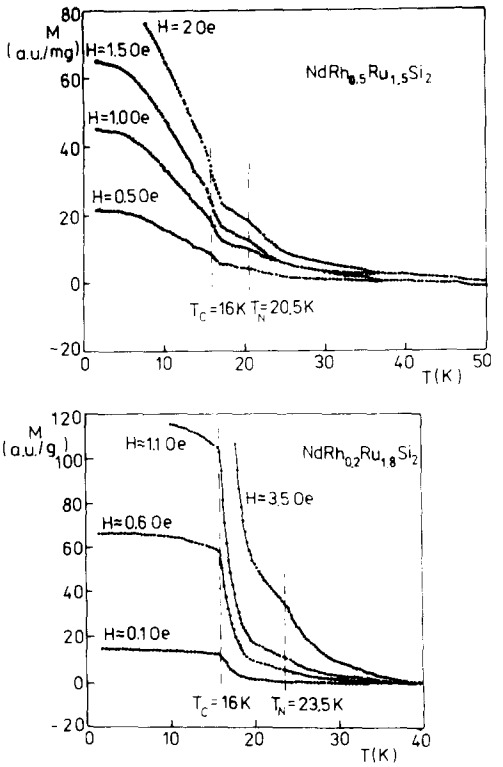

Fig. 7. Temperature dependence of the magnetization at low magnetic field for $NdRh_{0.5}Ru_{1.5}Si_2$ and $NdRh_{0.2}Ru_{1.8}Si_2$.

Neutron diffraction data indicate that $NdRh_2Si_2$ has the AFI-type structure. This structure is characterized by the commensurate propagation vector [0, 0, 1] and moments along the ⟨001⟩ direction.

An increase in concentration x in the $NdRh_{2-x}Ru_xSi_2$ system at 4.2 K revealed the following magnetic structures. At $x=0.5$ a coexistence of the collinear AFI-type structure and a modulated structure with propagation vector $\boldsymbol{k}=(0, 0, 0.838)$ is observed. At $x=0.8$, 1.0 and 1.5, modulated structures appear with the wave vector $\boldsymbol{k}=(0, 0, k_z)$. The k_z value decreases with increasing x from $k_z=0.838$ for $x=0.5$ to $k_z=0.231$ for $x=1.8$. At $x=1.8$ a coexistence of the ferromagnetic structure and a modulated structure is seen.

The concentration dependence of the Néel temperature observed for the $NdRh_{2-x}Ru_xSi_2$ system is similar to that observed in other systems, *e.g.* $CeRh_{2-x}Ru_xSi_2$ [10], $TbRh_{2-x}Ru_xSi_2$ [11] and $URh_{2-x}Ru_xSi_2$ [12]. In this system changes in the value of the Néel temperature *vs.* temperature are observed. Also in other systems, *e.g.* $Gd(Rh_{1-x}Ru_x)_3B_2$ [13] and $Ce(Cu_{1-x}Ni_x)_2Ge_2$ [14], changes in the magnetic properties are observed as a function of concentration x.

The magnetic interactions in these compounds are of the RKKY type [15]. Analysis of the Mössbauer spectroscopy data for isostructural GdT_2Si_2 compounds indicates an influence of the transition metal d-band on many properties of these compounds [16, 17].

A change in concentration x probably causes a change in the filling of the d-band. The anomalous dependence of the electronic specific heat coefficient on x for $CeRh_{2-x}Ru_xSi_2$ [18] and $URh_{2-x}Ru_xSi_2$ [12] is connected with this effect.

Photoemission studies of the $Ho(Rh_{1-x}Ru_x)_4B_4$ system, in which changes in the superconducting and magnetic properties with changes in concentration x are observed, indicate a shift of the Fermi energy E_F to lower energies with increasing x [19].

The influence of the external magnetic field causes the change in magnetic properties. For $NdRh_2Si_2$ a two-step metamagnetic process is observed. A similar dependence of the magnetization is observed for isostructural $TbCo_2Si_2$ [20], $DyCo_2Si_2$ [21] and $TbRh_2Si_2$ [22] compounds. For all these compounds below H_{c1} the antiferromagnetic collinear structure of the AFI type is observed [1, 2]. In the intermediate region, $H_{c1}<H<H_{c2}$, ferromagnetic ordering in a $+++-$ sequence [21] or a modulated structure [23] is detected, while for $H>H_{c2}$ the ferromagnetic ordering is stable.

Radwański and Franse [24] interpreted this type of two-step metamagnetic process in terms of the effect of crystalline electric field interactions of the 4f ions.

The magnetic properties of these compounds may be described by molecular field theory. The investigation by Katsura and Narita [25] shows that we should take

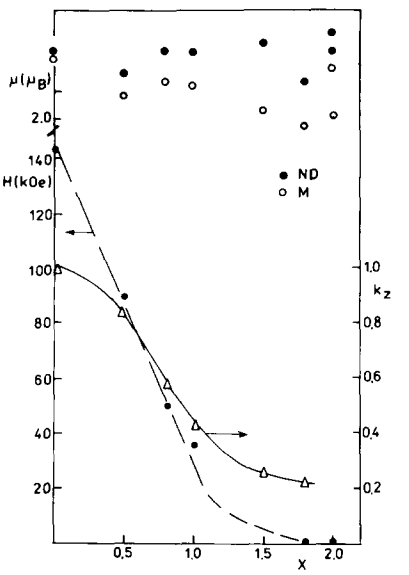

Fig. 8. Concentration dependence of magnetic moment μ, critical field H_c and wavevector k_z for the $NdRh_{2-x}Ru_xSi_2$ system.

the exchange integrals J_n at least up to $n=3$ to obtain the appearance of the structure with the $+++-$ sequence in the intermediate region.

With increasing Ru concentration x (for $x\geqslant 0.5$), the magnetization curves change from two-step curves, while the critical field values decrease. The decrease in critical field with increasing concentration x is correlated with changes in the wavevector (see Fig. 8).

In ref. 8 the change in the wavevector is analysed in terms of the Anisotropic next-nearest-neighbour Ising (ANNNI) model [26], in which the wavevector is given by $\cos(\pi k)=-J/4J_2$ (where J_1, J_2 are the exchange integrals between first and second nearest neighbours). Substitution of rhodium by ruthenium causes a change in J_i values. Owing to the fact that H_c is also correlated with the values of the exchange integrals [27], the above correlation is normal.

The values of the magnetic moment hardly change as a function of the concentration x. A difference between values determined by neutron diffraction and magnetic measurements is observed (see Fig. 8).

Measurements on single-crystal samples are necessary to determine the details of the magnetic properties of this system.

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