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Magnetic susceptibility, thermoelectric power and specific heat of UNi_2Si_2

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Abstract. The magnetic ternary compound UNi_2Si_2 has been studied by means of magnetic susceptibility, thermoelectric power and low-temperature (1.5–29 K) specific heat measurements. The magnetic susceptibility is Curie–Weiss-like above 130 K and becomes highly anisotropic at lower temperatures. Distinct features associated with the 103 and 53 K phase transitions are observed while deviation from Curie–Weiss behaviour occurs near the 123 K phase transition. The thermoelectric power from oriented samples is also anisotropic, with the *c*-component strongly coupled to the three magnetic phase transitions. The gamma value obtained from the specific heat measurements is 22 mJ mol^{-1} K^{-2} which indicates a small mass enhancement in this system.

1. Introduction

UNi_2Si_2 has the $ThCr_2Si_2$ crystal structure and has been attracting attention lately because of the various magnetic phase transitions observed in this system [1–4]. Early studies by neutron diffraction [1] revealed that the system undergoes a magnetic phase transition at $T_N = 103$ K to a collinear antiferromagnetically ordered state (AF1). At 53 K, the system undergoes a second phase transition into a commensurate longitudinal spin density wave (LSDW) state. The magnetization measurements [1, 2], however, show ferromagnetic ordering below 98 K and no evidence of a phase transition at 53 K. More recently, neutron diffraction studies of single-crystal samples by Lin *et al* [3] confirmed the presence of the two magnetically ordered phases and, in addition, established a third magnetically ordered phase, between 103 K and 123 K, which is an incommensurate longitudinal spin density wave state. The studies also revealed that the longitudinal spin density wave below 53 K is acompanied by a net ferromagnetic moment of $(1.0 \pm 0.3)\mu_B$ along the *c* axis on each uranium site. Above 123 K, the system is paramagnetic. These transitions have been studied recently by resistivity and Hall effect measurements [4]. The present work investigates the electronic properties of single-crystal UNi_2Si_2 via magnetization, thermoelectric power and specific heat measurements. Detailed comparisons are made between the results of this work and those of the previous neutron [3] and transport studies [4].

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2. Experimental details

The single crystal of UNi_2Si_2 used in this study was prepared from U, Ni, and Si ingots which were premelted, cleaned where appicable, and weighed. They were reacted and homogenized in an inert gas atmosphere in an arc furnace with a water-cooled hearth. The single crystal was grown by the Czochralski technique in a Reed-type triarc furnace which had been modified to include a water-cooled hearth and seed rod. Argon gettered with titanium was used at 100 kPa as the chamber atmosphere. The samples for various measurements were cut parallel and perpendicular to the c axis with a spark cutter. X-ray measurements established that the samples were single crystals with lattice parameters $a = 3.96$ Å and $c = 9.51$ Å. The magnetization was measured with a Quantum Design SQUID magnetometer. The thermoelectric power was measured by a differential method using high-purity lead wires as the reference. For the specific heat measurements, a conventional relaxation method was used [5].

3. Results and discussion

The magnetic susceptibility (M/H) measured in a magnetic field of 1.6 T is shown in figure 1 for fields applied parallel ($\chi_\parallel$) and perpendicular ($\chi_\perp$) to the c axis. The susceptibility is very anisotropic, with $\chi_\parallel$ exceeding $\chi_\perp$ by a factor of 2.8 at room temperature and 54 at 5 K. From room temperature down to about 130 K, the susceptibilities can be described by the Curie–Weiss law, as can be seen from the inset of figure 1. A fit for temperatures above 130 K yields $\theta_{CW} = -530$ K and $\mu_{eff} = 3.55\mu_B$ for $\chi_\perp$, $\theta_{CW} = -15$ K and $\mu_{eff} = 3.67\mu_B$ for $\chi_\parallel$. At about 130 K, where the system is about to enter a magnetically ordered phase (incommensurate LSDW), the susceptibilities start to deviate from Curie–Weiss behaviour. At 103 K, a sharp peak characteristic of an antiferromagnetic transition is observed in $\chi_\parallel$. A similar feature is also present in $\chi_\perp$ but the magnitude is much smaller and cannot be seen on this scale. Between 100 and 80 K, the magnetic susceptibility is a flat minimum. As the temperature is further lowered, the onset of ferromagnetism occurs at about 78 K for $H \parallel c$. The ferromagnetism coexists with the commensurate LSDW phase [3]; the apparent increase of the transition temperature is due to the magnetic field applied along the c axis [6]. The estimated ferromagnetic moment at 4.2 K is $0.62\mu_B$ per U atom which is in reasonable agreement with the neutron diffraction result [3] of $(1.0 \pm 0.3)\mu_B$. The magnetic susceptibility perpendicular to the c axis ($\chi_\perp$), however, shows a very different behaviour: it seems to increase at about 57 K and reaches a relatively broad maximum of 2.0×10^{-2} emu mol^{-1} at about 40 K. Below about 20 K, the susceptibility drops to 3.9×10^{-3} emu mol^{-1} and remains at this value at low temperatures.

One of the concerns in fitting the magnetic susceptibility with the Curie–Weiss law is that the parameters obtained may be obscured by the existence of a temperature-independent Pauli paramagnetic susceptibility. To ensure that this is not the case, a best fit of the data with the expression $\chi = \chi_{CW} + \chi_{Pauli}$, where χ_{CW} is the Curie–Weiss term and χ_{Pauli} is the Pauli paramagnetic term, was also carried out. For $\chi_\perp$, the fit yields $\theta_{CW} = 430$ K, $\mu_{eff} = 3.10\mu_B$ and $\chi_{Pauli} = 2.8 \times 10^{-4}$ emu mol^{-1}. For $\chi_\parallel$, the fit yields $\chi_{Pauli} = (0.0 \pm 1.0) \times 10^{-4}$ emu mol^{-1} which leaves the values of θ_{CW} and μ_{eff} unaltered. The results do not differ significantly from the fit of a simple Curie–Weiss term, suggesting that the large anisotropy in θ_{CW} is intrinsic to the system and a simple Curie–

Weiss term is adequate to describe the magnetic susceptibility in the paramagnetic phase.

The temperature dependence of the thermoelectric power parallel ($S_{\parallel}$) and perpendicular ($S_{\perp}$) to the tetragonal *c* axis is shown in figure 2. The thermoelectric power is also very anisotropic, with a larger magnitude perpendicular to the *c* axis. Above 123 K, both $S_{\parallel}$ and $S_{\perp}$ are weakly dependent on temperature and the ratio of $S_{\perp}/S_{\parallel}$ is in the range 2–4. The thermoelectric power is negative from room temperature down to about 20 K, and becomes positive at temperatures below 20 K. The positive thermoelectric power reaches a shallow but notable maximum at about 10 K, and then starts to diminish as the temperature is lowered further. For the thermoelectric power parallel to the *c* axis, there are sharp features at the three magnetic phase transitions at 123, 103 and 53 K. For the thermoelectric power perpendicular to the *c* axis, there is a broad peak centred at approximately 94 K. However, the only sharp feature associated with the phase transitions is the sudden increase of the magnitude at about 123 K. The features associated with the 103 K and 53 K transitions are rather weak compared with those observed in $S_{\parallel}$. Thus, the behaviour of the thermoelectric power is in good correspondence with that of the resistivities reported earlier [4] in which the resistivity parallel to the *c* axis shows distinct features at all three phase transitions while the resistivity perpendicular to the *c* axis shows no feature at 53 K and a comparatively weak feature (compared with the feature in the *c*-axis resistivity) at 103 K [7].

The fact that the thermoelectric power perpendicular to the *c* axis shows weak features at the 103 and 53 K transitions, together with the fact that the resistivity perpendicular to the *c* axis shows no feature at 53 K and a comparatively weak feature at 103 K [4], suggests that the transport electrons moving perpendicular to the *c* axis are rather insensitive to the two phase transitions at lower temperatures. On the other hand, both resistivity [4] and thermoelectric power parallel to the *c* axis shows strong features at the 123 K transition as well as the 103 and 53 K transitions. This may be understood by considering the fact that when the system undergoes the phase transitions from the incommensurate LSDW phase to the AF1 phase at 103 K and from AF1 phase to the commensurate LSDW phase at 53 K, only the periodicities of the magnetic superstructures along the *c* axis are altered [2, 3]. Hence we expect the phase transitions to be more influential on the transport properties along the *c* axis [8].

The specific heat is plotted as C_p/T versus T^2 from 1.5 K to 29 K in figure 3. For $T < 10$ K, the data can be fitted with a linear term plus a cubic term down to the lowest temperature measured (see the inset of figure 3). The fit yields a gamma value of 22 mJ mol^{-1} K^{-2} and a Debye temperature $T_D = 370$ K. This result suggests a rather small mass enhancement in this system. The high-temperature part ($T > 12$ K) of the specific heat, plotted as C_p/T versus T^2, is rather linear and shows no sign of curving down up to the highest temperature measured (29 K).

4. Conclusions

We have studied the electronic properties of UNi_2Si_2 via magnetization, thermoelectric power and specific heat measurements. Above 130 K, the magnetic susceptibility can be described by the Curie–Weiss law with $\mu_{eff} = 3.55\mu_B$ and $\theta_{CW} = -530$ K for $\chi_{\perp}$, $\mu_{eff} = 3.67\mu_B$ and $\theta_{CW} = -15$ K for $\chi_{\parallel}$. At 103 K, a cusp characteristic of an antiferromagnetic transition has been observed. At about 78 K, the onset of ferromagnetism is observed with the ferromagnetic moment along the *c* axis. The corresponding ferromagnetic

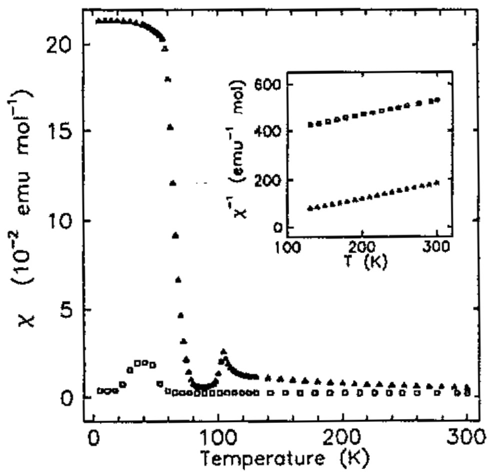

Figure 1. DC magnetic susceptibility of single-crystal UNi_2Si_2 measured in a field of 1.6 T. Δ: $\boldsymbol{H} \parallel c$; $\Box$: $\boldsymbol{H} \perp c$. The inset is the inverse of the magnetic susceptibility in the temperature range 130–300 K.

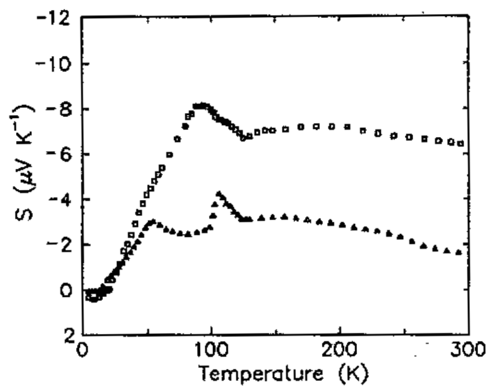

Figure 2. Thermoelectric power of single-crystal UNi_2Si_2. Δ: $\boldsymbol{\nabla T} \parallel c$; $\Box$: $\boldsymbol{\nabla T} \perp c$.

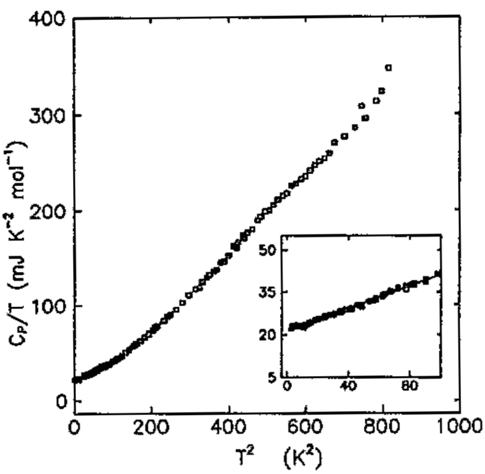

Figure 3. Specific heat divided by temperature versus temperature squared for UNi_2Si_2. The inset shows the linear fit of the low-temperature part (1.5–10 K).

moment is $0.62\mu_B$ per U atom. The thermoelectric power also shows anisotropic behaviour, with the *c*-axis component strongly coupled to the magnetic phase transitions. Low-temperature specific heat measurements indicate that the system has only a small mass enhancement ($\gamma = 22$ mJ mol^{-1} K^{-2}) and is not a heavy-fermion material.

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[6] The phase transition temperautre of the commensurate LSDW state increases by about 14 K for a 1.6 T field applied along the *c* axis. No change of this temperature was observed with the field perpendicular to the *c* axis. See also: Ning Y B and Datars W R 1990 (unpublished).

[7] The comparison was made here in terms of the amplitudes of the resistivity derivatives in [4].

[8] The Hall effect is an exception since it involves the magnetization perpendicular to the current direction.