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Transport properties and phase diagram of UNi_2Si_2

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Abstract. The resistivity and Hall coefficient of single-crystal UNi_2Si_2 have been studied in detail for the temperature range 4.2–300 K. The resistivity of UNi_2Si_2 is largely due to magnetic scattering and the phonon scattering contribution is estimated to be about 14% at room temperature. At low temperatures, the resistivity can be described by a gapped spin-wave model plus a T^2 term. The temperature dependence of the Hall coefficient is accounted for by a theoretical model invoking skew scattering of conduction electrons by localized magnetic moments. Among the three magnetic phase transition temperatures, the two lower ones are found to be magnetic field dependent and shift with the field applied along the tetragonal c axis. Using the resistivity measurement in an applied magnetic field, a field–temperature phase diagram of UNi_2Si_2 is presented.

1. Introduction

A recent study of UNi_2Si_2 by neutron scattering measurements [1] has revealed that UNi_2Si_2 has three magnetically ordered phases at moderately low temperatures. In all the ordered phases, the magnetic moments are aligned along the tetragonal c axis. Below 53 K, the system has an ordered state that can be described as a longitudinal squared-off wave, in which every uranium atom carries the same moment of $2.2\mu_B$ and for every moment along the $+z$ direction there are two moments along the $-z$ direction. The magnetic structure has a periodicity of $3c$, where c is the lattice constant along the c axis. Because of the imbalance of the moments along the $+z$ and $-z$ directions, there is a net ferromagnetic moment along the c axis in this phase. It is interesting to note that the ordered structure in this phase can also be described by a commensurate longitudinal spin-density-wave (LSDW) model with the magnetic moment on the nth uranium atom at position $\boldsymbol{R}_n$ given by

$$\mu_n = [\mu_0 \cos(2\pi \boldsymbol{R}_n \cdot \boldsymbol{q}) + \mu_z]\boldsymbol{z} \tag{1}$$

where $\boldsymbol{z}$ is the unit vector along the c axis, $\mu_0 = (2.7\pm0.3)\mu_B$ and $\mu_z = (1.0\pm0.3)\mu_B$. These two models are equivalent within the experimental uncertainty of the neutron scattering measurements [1]. For clarity of discussion in this paper, we shall adopt the squared-off wave description and define this phase as the SQW phase. Between 53 and 103 K, the system is a simple body-centred tetragonal antiferromagnet (AFI). Between

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103 and 123 K, the system takes on an incommensurate longitudinal spin-density-wave (ILSDW) structure, with a periodicity slightly smaller than $4c$. Above 123 K, there is a paramagnetic state. In addition to the neutron scattering studies, these phase transitions have also been detected by transport and magnetic susceptibility measurements [2, 3]. The previous resistivity and Hall effect data [2] are extended in this paper with the measurement of the resistivity with an applied magnetic field and the Hall effect with the current perpendicular to the c axis. The temperature dependence of the resistivity and Hall effect are explained with reasonable theoretical models. A magnetic field dependence of the transition temperatures detected in the resistivity is used to determine a field–temperature phase diagram.

2. Experimental details

The single crystal used in this study was prepared from U, Ni and Si ingots using the Czochralski technique, with the detailed procedure described elsewhere [2, 3]. The samples were oriented and cut along 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 resistivity was measured with the standard four-probe method. For the Hall coefficients, a four-contact geometry (two for the Hall voltage and two for the current) was used. The Hall effect was measured with an electromagnet capable of generating a field of 1.8 T. Measurements of the magnetic field dependence of the resistivity were carried out with an Oxford superconducting magnet which was able to generate fields up to 7.5 T.

3. Results and discussion

Figure 1 shows the temperature dependence of the resistivity, in the directions parallel ($\rho_{\|}$) and perpendicular ($\rho_{\perp}$) to the tetragonal c axis. The resistivity is highly anisotropic. The room-temperature value of the resistivity is 226 $\mu\Omega$ cm along the c axis and 156 $\mu\Omega$ cm perpendicular to the c axis. At 4.2 K, the c-axis resistivity $\rho_{\|}$ decreases to 69 $\mu\Omega$ cm and the value of $\rho_{\perp}$ drops to 20 $\mu\Omega$ cm. The temperature coefficient $d\rho/dT$ is positive at all temperatures for $\rho_{\perp}$ but is negative for $\rho_{\|}$ down to 150 K. There are several distinct features that are associated with the three magnetic phase transitions. As the temperature is lowered, a drastic decrease in the resistivity along both directions occurs at about 123 K. A second feature occurs at about 103 K where the slope of $\rho_{\|}$ becomes much steeper while the resistivity perpendicular to the c axis develops a knee. As the temperature is lowered further, a local maximum occurs in $\rho_{\|}$ between 56 and 35 K, while the resistivity $\rho_{\perp}$ is very smooth in the same temperature range. The local maximum in $\rho_{\|}$ shows a temperature hysteresis of 4.5 K, as shown in figure 1.

The temperature dependence of the resistivity above 123 K is comparatively weak. Such a behaviour indicates that electron–phonon scattering does not dominate the contributions to the resistivity in this system and scattering with a magnetic origin is more important. If one takes the linear variation in the resistivity $\rho_{\perp}$ above 150 K as due to phonon scattering, then at room temperature the phonon contribution only makes up about 14% of the total resistivity ρ. Above 150 K, the resistivity $\rho_{\|}$ has

a negative slope and is characteristic of the Kondo behaviour. A best fit with the expression

$$\rho = \rho_0 + AT - B \ln T \tag{2}$$

however, requires a negative phonon contribution to the resistivity which is not physical. Such a result indicates that the resistivity in this temperature range is in a broad crossover region from Kondo behaviour at higher temperatures to non-Kondo behaviour, and one would need to extend the measurement to much higher temperatures to have a meaningful fit.

The resistivity at low temperatures does not follow a simple T^2 behaviour and, instead, it can be described by a gapped spin-wave term [4] plus a T^2 term:

$$\rho = \rho_0 + AT^2 + BT(1 + 2T)/\Delta) \exp(-\Delta/T) \tag{3}$$

where ρ_0 is the residual resistivity, Δ is the energy gap in the spin-wave (magnon) spectrum, and A and B are constants. The best fit with this model is shown in figure 2 for the temperature range 4.2–45 K. The best-fit parameters are summarized in table 1. Because of the large magnetic anisotropy in the system, as has been demonstrated by the magnetic susceptibility [3] and neutron scattering measurements [1], it is almost certain that an energy gap should be present in the spin-wave excitation spectrum. Therefore it is not surprising that the gapped spin-wave model works as well as it does. Note that the gap values Δ are comparable with those of URu_2Si_2 [5, 6] and UNi_2Ge_2 [7] obtained through the same procedure, indicating a similar magnetic anisotropy in these systems.

Table 1. Best-fit parameters obtained from fitting the low-temperature resistivity of UNi_2Si_2 with equation (3) in the text.

	Temperature range (K)	ρ_0 ($\mu\Omega$ cm)	A ($\mu\Omega$ cm K^{-2})	B ($\mu\Omega$ cm K^{-1})	Δ (K)
$I \parallel c$	4.2–45	70.03	1.2×10^{-3}	1.03	100
$I \perp c$	4.2–45	19.67	5.9×10^{-4}	0.38	102

The strongest anisotropy of the resistivity occurs in the vicinity of the 53 K phase transition where a local maximum is seen in $\rho_{\parallel}$ while $\rho_{\perp}$ remains very smooth. This type of resistivity anomaly at the magnetic phase transition has been observed in several systems including Cr, α-Mn [8], URu_2Si_2 [5, 6] and UNi_2Ge_2 [7]. In all the cases, the origin of the anomalous increase has been attributed to gapping of the Fermi surface (by the formation of a magnetic Brillouin zone) and critical scattering. The effect of gapping is to reduce the effective number of conduction electrons along the order direction which results in an increase in the resistivity just below the transition temperature [9, 10]. Because of the drastic change in the magnetic periodicity along the c axis at 53 K, gapping of the Fermi surface may well occur in UNi_2Si_2. However, the situation here is different; the phase transition at 53 K is of first order, as is indicated by the temperature hysteresis, and the term due to critical scattering should be absent. Furthermore, one can no longer use the critical approach to handle the temperature dependence of the energy gap at the Fermi surface which in turn gives rise to the T dependence of the gap resistivity. In fact, for a first-order

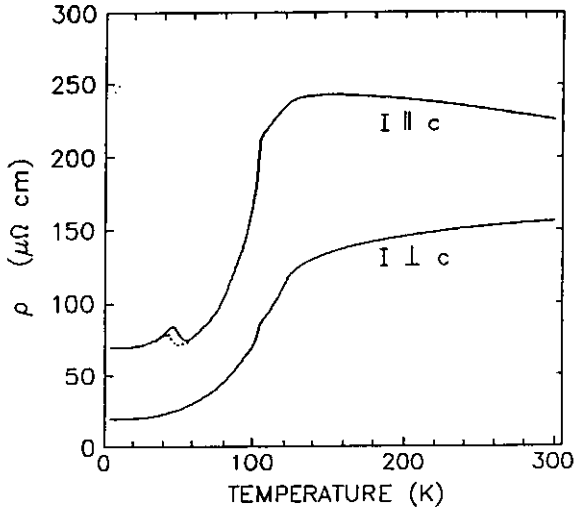

Figure 1. Temperature dependence of the resistivity of UNi_2Si_2: ·····, curve obtained with decreasing temperature to show the thermal hysteresis.

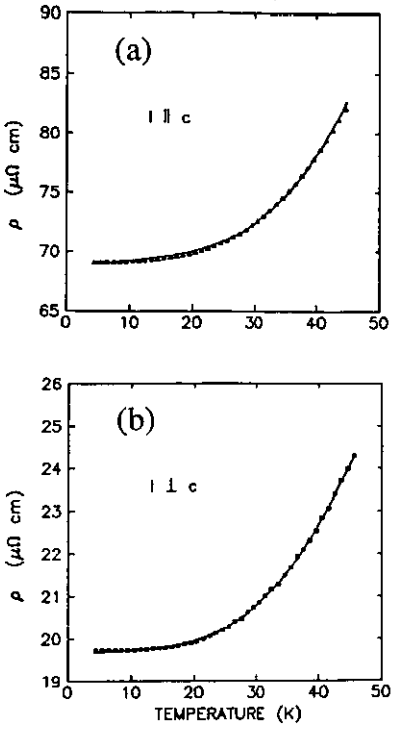

Figure 2. Resistivity of UNi_2Si_2 in the temperature range 4.2–45 K for (*a*) $I \parallel c$ and (*b*) $I \perp c$: ——, best fit using equation (3) in the text. The fitting parameters are summarized in table 1.

phase transition, the energy gap opened up at the Fermi surface should be a step function of temperature and goes from zero to a finite value at the phase transition temperature. In a real system, however, the phase transition and hence the step function of the energy gap may be broadened considerably, as in the case of UNi_2Si_2 [1]. Despite these differences, the anomalous increase in the resistivity $\rho_{\parallel}$ in the vicinity of 53 K can be qualitatively explained by gapping of the Fermi surface due to the folding of the magnetic Brillouin zone along the c axis.

A very interesting phenomenon is that, when a magnetic field is applied along the c axis, the two lower phase transition temperatures change significantly. No such change in the 123 K phase transition is seen within the experimental uncertainty (2 K in a field of 3.5 T). Figure 3 shows the resistivity $\rho_{\parallel}$ taken at 0 and 2.5 T for the temperature range 15–140 K. One can note immediately that the anomaly associated with the 53 K transition (SQW–AF1) in zero field moves to a higher temperature by about 20 K in a 2.5 T field. Also, the feature at 103 K in zero field moves to a lower temperature in an applied field. In contrast, the feature at 123 K remains virtually unchanged. A quantitative and also more sensitive way of describing these changes is to identify the features of the phase transitions in the temperature derivative $d\rho_{\parallel}/dT$ of the resistivity. The SQW–AF1 transition is identified as the negative minimum in $d\rho_{\parallel}/dT$, the antiferromagnetic transition is identified as the peak in $d\rho_{\parallel}/dT$, and

the 123 K transition is identified as the shoulder in $d\rho_{\parallel}/dT$, as indicated by the three arrows in figure 4. In doing this, a field–temperature phase diagram of UNi_2Si_2 is mapped out which is shown in figure 5. The highest field at which the two lower-temperature phase transitions can be identified is 3.5 T. At still higher fields, these two features are no longer observable. The fact that the transitions are not seen in the resistivity measurements above 3.5 T indicates that the SQW–AF1 transition does not occur above 3.5 T and the phase boundaries around the AF1 region are connected in figure 5. Consequently, there must be another phase boundary emerging somewhere to separate the SQW phase from the ILSDW phase. Such a phase boundary has indeed been observed by Lin and Collins [11] in their neutron scattering measurements carried out in an applied magnetic field. In fact, the phase boundary between the AF1 phase and the ILSDW phase splits at a field of 2.5 T, as is shown in figure 5. This new phase boundary separating the SQW and ILSDW phases was not seen in the resistivity measurement, suggesting a very small change in the scattering process of conduction electrons. From a thermodynamic point of view, an increase in the SQW–AF1 transition temperature with increasing applied magnetic field is expected, as the SQW phase has a net ferromagnetic moment and can lower its energy by having the net moments aligned along the direction of the applied field. Similarly, this argument can also be applied to explain the increase in the SQW–ILSDW transition temperature and the decrease in the AF1–SQW transition temperature above the splitting point.

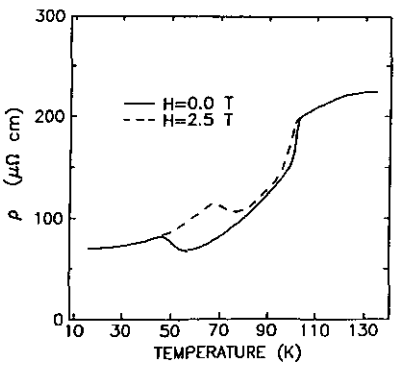

Figure 3. Resistivity of UNi_2Si_2 measured with $I \parallel c$ and a magnetic field applied along the c axis: ——, $H = 0$ T; – – –, $H = 2.5$ T.

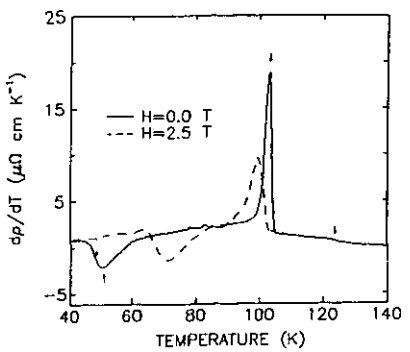

Figure 4. Derivative of the resistivity of UNi_2Si_2 measured with $I \parallel c$ and a magnetic field applied along the c axis: ——, $H = 0$ T; – – –, $H = 2.5$ T.

Figure 6 presents the temperature dependence of the Hall coefficient of UNi_2Si_2 measured in a field of 1.6 T. As in many of the Kondo lattice and heavy-fermion compounds, the Hall coefficient of UNi_2Si_2 has a strong temperature dependence and is mostly positive except at its minimum where small negative values are observed. Again, the behaviour of the Hall coefficient is highly anisotropic in both the magnitude and the temperature dependence. To account for the anomalous Hall effect in Kondo lattice and heavy-fermion compounds, Fert and Levy [12] have devised a theoretical model invoking magnetic skew scattering which can be expressed as the following:

$$R_H = R_H^o + R^s \rho_m \chi \tag{4}$$

where R_H^o is the ordinary Hall coefficient, R^s is the pre-factor of the skew scattering term which is related to the phase shifts of the conduction electrons during the scattering process [12] and χ is the magnetic susceptibility. To carry out a best fit of the Hall coefficient with the above expression, one needs to know the magnetic part ρ_m of the resistivity. Since the temperature dependence of the resistivity is dominated by magnetic scattering, it is a reasonable approximation to neglect the phonon contribution and to replace ρ_m with the total resistivity $\rho(T)$. As we do not know how much of the residual resistivity is of magnetic origin, we simply let the magnetic residual resistivity ρ_{0m} vary and be determined by the best-fit procedure. The best fit with this model is shown in figure 6 as the full curves, and the fitting parameters are summarized in table 2. It is interesting to note that, for the case $H \perp c$ and $I \parallel c$, it is necessary to break the data into two temperature regions at 53 K to obtain a reasonable fit of the data. This situation has also been encountered in URu_2Si_2 [6] and UNi_2Ge_2 [7] and may be interpreted as another signature of a Fermi surface modification.

Table 2. Parameters obtained through the best-fitting procedure of the Hall coefficient with equation (4) in the text.

	Temperature range (K)	ρ_{0m} ($\mu\Omega$ cm)	R_H^o (10^{-3} cm^3 C^{-1})	R^s (cm^2 mol Ω^{-1} emu^{-1})
$H \parallel c$, $I \perp c$	4.2–45	4.5	-0.03 ± 0.03	$(1.40 \pm 0.10) \times 10^3$
$H \perp c$, $I \parallel c$	4.2–53	3.5	0.10 ± 0.01	$(-0.30 \pm 0.05) \times 10^3$
$H \perp c$, $I \parallel c$	53–300	3.5	-0.05 ± 0.01	$(1.59 \pm 0.04) \times 10^3$

4. Conclusions

We have studied the resistivity and Hall coefficient of UNi_2Si_2. The resistivity is dominated by magnetic scattering, and the phonon scattering contribution is estimated to be about 14% of the total resistivity at room temperature. At low temperatures, the resistivity can be described well by a gapped spin-wave model plus a T^2 term. The energy gap in the spin-wave spectrum inferred from the resistivity measurement is about 100 K. The Hall coefficient can be accounted for by the model of Fert and Levy which invokes magnetic skew scattering of the conduction electrons. The two lower phase transition temperatures are field dependent and can shift when a magnetic field is applied along the c axis. By monitoring the c-axis resistivity anomaly and features

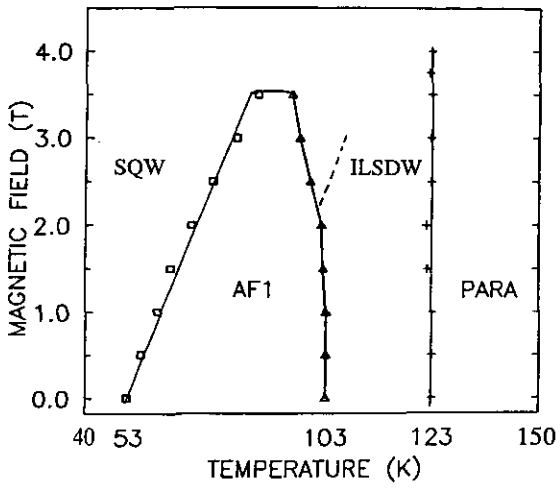

Figure 5. Phase diagram of UNi_2Si_2 mapped out by monitoring the field dependence of the transition temperatures: – – –, result of the neutron scattering measurement of Lin and Collins [11].

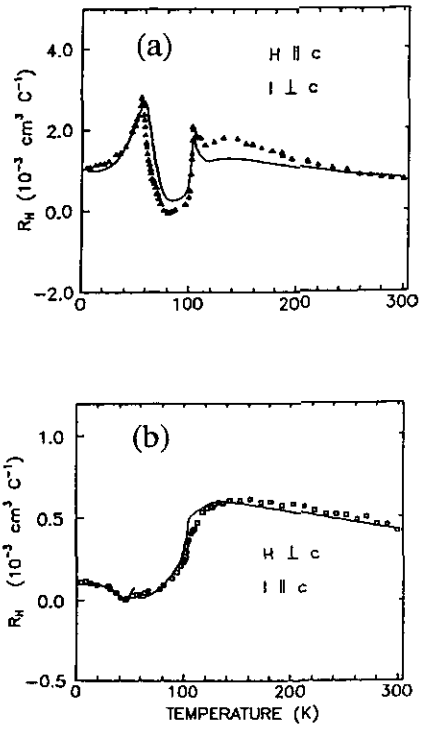

Figure 6. Temperature dependence of the Hall coefficient measured in a field of 1.6 T for (*a*) $\boldsymbol{H} \parallel c$ and $\boldsymbol{I} \perp c$ and (*b*) $\boldsymbol{H} \perp c$ and $\boldsymbol{I} \parallel c$: ——, best fit using equation (4) in the text. The fitting parameters are summarized in table 2.

at the three phase transitions in an applied magnetic field, a field–temperature phase diagram of UNi_2Si_2 has been mapped out. The SQW–AF1 phase transition temperature increases linearly with increasing applied magnetic field. At a field of about 2.5 T, the phase boundary between the AF1 phase and the ILSDW phase splits into two branches. At a field of 3.5 T, the AF1 phase becomes unstable against the SQW phase and is eventually taken over by the SQW phase. These features of the phase diagram of UNi_2Si_2 can be understood thermodynamically in terms of the energy gain of the SQW phase in an applied magnetic field along the tetragonal *c* axis.

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