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Resistivity and Hall effect of the magnetic ternary compound UNi_2Si_2

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Electrical-resistivity and Hall-effect measurements on single-crystal UNi_2Si_2 have been performed between 4.2 and 300 K. The resistivity ρ is highly anisotropic with a larger magnitude parallel to the c axis. Above 150 K, $d\rho/dT$ is small and positive in the ab plane and is negative along the c axis. Distinct features and anomalies have been observed in both the resistivity and the Hall-effect measurements. They are associated with the magnetic phase-transition temperatures at 123, 103, and 53 K observed by neutron diffraction and scattering experiments.

UNi_2Si_2 crystallizes in the $ThCr_2Si_2$ type of crystal structure and has been studied previously by neutron diffraction and magnetization measurements.[1,2] Neutron-diffraction studies[1] by Chelmicki, Leciejewicz, and Zygmunt showed that UNi_2Si_2 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 an antiferromagnetically ordered state with spin amplitude modulation along the order direction c (often referred to as LSDW —longitudinal spin-density wave). In contrast, magnetization studies[1,2] exhibited complex features and showed evidence of ferromagnetic ordering.

Very recently, Lin *et al.*[3] carried out detailed elastic neutron-scattering studies on single-crystal UNi_2Si_2 samples. The results clearly demonstrate the presence of a commensurate LSDW structure below 53 K and the AF1 state between 53 and 103 K. In addition, these authors have estblished a third magnetic phase between 103 and 123 K which is the LSDW type but incommensurate with respect to the lattice periodicity. Above 123 K, the system is paramagnetic. The various magnetic phases and phase-transition temperatures are summarized in Fig. 1. The commensurate LSDW phase has a periodicity of $3c$ and the incommensurate one has a periodicity slightly smaller than $4c$, where c is the c-axis lattice spacing.[3]

In this Rapid Communication, we present resistivity and Hall-effect measurements on single-crystal UNi_2Si_2 samples and make a direct connection and comparison with the features observed by the neutron-scattering studies.

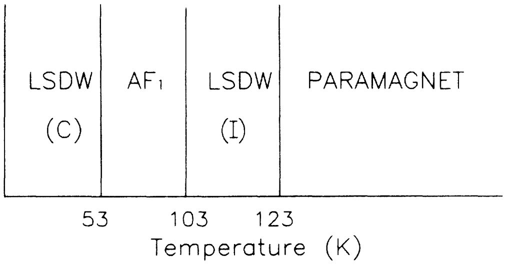

FIG. 1. Schematic phase diagram of UNi_2Si_2: LSDW, longitudinal spin-density wave; (C), commensurate; (I), incommensurate; AF1, collinear antiferromagnetic phase.

The single crystal used in this study was grown from U, Ni, and Si which were premelted, cleaned where applicable, and weighed. They were reacted and homogenized in an inert gas in a water-cooled-hearth arc furnace. The UNi_2Si_2 crystal was grown by the Czochalski technique in a Reed-type triarc furnace which had been modified to give a water-cooled hearth and seed rod. Argon gettered with titanium was used as the chamber atmosphere with a pressure of 100 kPa. The growth rate was 17 mm/h. The rod was rotated at 9 rpm clockwise and the hearth at 60–90 rpm counterclockwise. The samples were cut along and perpendicular to the c axis with a spark cutter. X-ray measurements established that the samples were single crystals with the lattice parameters $a = 3.96$ Å and $c = 9.51$ Å. Both the resistivity and the Hall effect were measured with the standard, four-probe method. The temperature was measured with calibrated carbon-glass and platinum thermometers.

Figure 2 shows the temperature dependence of the resistivity for two current directions, parallel to the ab plane and the c axis. The room-temperature resistivity is 156 $\mu\Omega$ cm in the ab plane and 226 $\mu\Omega$ cm along the c axis. The resistivities at 4.2 K decrease to 20 $\mu\Omega$ cm in

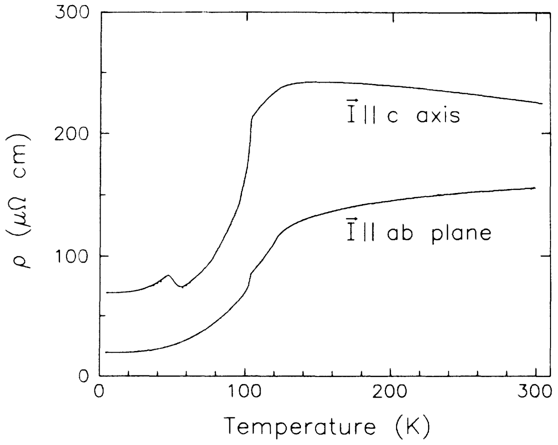

FIG. 2. Temperature dependence of the resistivity of single-crystal UNi_2Si_2 with the current parallel to the ab plane and the c axis. The solid curves were taken as the temperature was increasing and the dotted curve was taken as the temperature was decreasing.

the *ab* plane and 69 $\mu\Omega$ cm along the *c* axis. The temperature coefficient $d\rho/dT$ is positive in the *ab* plane at all temperatures but is negative down to 150 K along the *c* axis. There are several distinct "anomalies" in the resistivity below 130 K. As the temperature is lowered, a drastic decrease in the resistivity along both directions occurs at about 123 K. The second anomaly occurs at about 103 K where the slope of the resistivity parallel to the *c* axis becomes much steeper and the resistivity of the *ab* plane develops a knee. As the temperature is lowered further, the resistivities gradually level off and saturate below 50 K. For the resistivity along the *c* axis, a local maximum occurs between 56 and 35 K, with the peak centered at about 47 K. This feature shows a temperature hysteresis of about 4.5 K as shown in Fig. 2. This anomaly is reminiscent of that in the heavy-fermion system URu_2Si_2 at the Néel temperature.[4–7] To better describe these anomalies, we have computed the derivative $d\rho/dT$ as a function of temperature which is presented in Fig. 3. We note that all the distinct features occur in the vicinity of the three magnetic phase-transition temperatures[3] observed by Lin *et al.*, indicating that all the anomalies are intimately related to the phase transitions.

One of the characteristic features of Kondo-lattice systems is the occurrence of the coherent scattering of electrons by localized magnetic moments (Kondo coherence).[8] This is marked by the coherent temperature T_0 and the onset of this coherent scattering in general will lead to a decrease in resistivity. With the presence of magnetic phase transitions, the behavior of the resistivity becomes more complex, depending on how far the coherence has developed when the transitions occur and how much they modify the Fermi surface.[9] In URh_2Si_2 (Ref. 10) and UNi_2Ge_2,[11] the antiferromagnetic ordering occurs at about 130 and 77 K, respectively, and is accompanied by the onset of a fast decrease in resistivity. In UCu_2Ge_2,[11] the ferromagnetic ordering occurs at about 105 K at which a sharp decrease in resistivity is observed. The fact that the resistivities show sharp decreases at the magnetic ordering temperatures in these systems before showing any sign of onset of coherence seems to suggest that, if the magnetic ordering occurs above the anticipated coherent temperature T_0, this ordering will effectively suppress both the spin-flip scattering (Kondo scattering) and the spin-disorder scattering which in turn leads to a decrease in resistivity. Our results on UNi_2Si_2 do show decreases in resistivity at the two higher magnetic ordering temperatures (103 and 123 K) and are certainly consistent with the above observation. URu_2Si_2 on the other hand, shows a sharp increase in resistivity at the Néel temperature T_N which is superimposed on a rapidly falling background due to the onset of the Kondo coherence.[4–7] This sharp increase in resistivity is reminiscent of the Néel temperature anomaly in chromium—a spin density wave (SDW) antiferromagnet in which both the critical scattering and the gapping of the Fermi surface play important roles in determining the behavior of the resistivity near the Néel temperature.[12,13] In contrast to the heavy-fermion system URu_2Si_2, the resistivity anomaly in UNi_2Si_2 in the vicinity of 53 K is much broader and only occurs along the *c* axis while the resistivity in the *ab* plane is very smooth. There is, however, a fundamental difference between the phase transition at $T_N = 17.5$ K in URu_2Si_2 and that at 53 K in UNi_2Si_2. The former is an antiferromagnetic phase transition while the latter is a phase transition from one ordered state (AF1) to another (LSDW). The shape of the order parameter from neutron-scattering measurements[3] as well as the temperature hysteresis in resistivity strongly suggest that the 53-K phase transition in UNi_2Si_2 is of first order, yet the anomaly in the *c*-axis resistivity is not the usual type of first-order phase transition with a discontinuous jump. Previous theoretical studies[13–16] have often incorporated both the gapping effect (due to the magnetic superzone) and the critical scattering to explain the resistivity anomaly near the Néel temperature T_N. The gapping of the Fermi surface usually gives the sharp increase in the resistivity just below T_N while the critical scattering on the other hand gives a sharp decrease, yielding a peak just below the Néel temperature T_N.[13–16] Since the 53-K phase transition in UNi_2Si_2 is first order, as we have mentioned, therefore, critical scattering should be absent. Clearly, any attempt to explain the resistivity anomaly near the 53-K transition within the above frame would have to incorporate also other mechanisms (such as impurities, inhomogeneities, and induced strain effect, etc.) with the gapping effect.

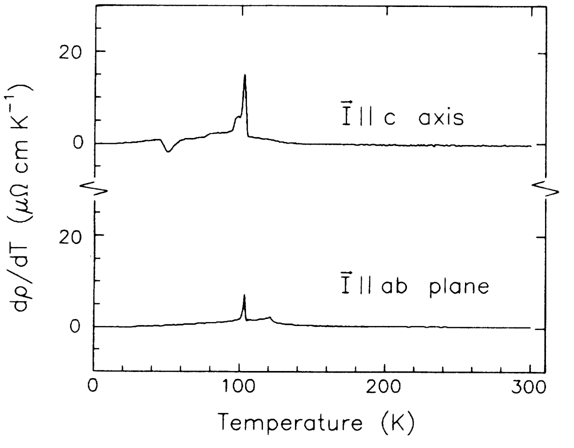

FIG. 3. Temperature dependence of the derivative of resistivity taken during warming of single-crystal UNi_2Si_2, with the current parallel to the *ab* plane and the *c* axis.

In Fig. 4, we present the temperature dependence of the Hall coefficient measured with the magnetic field *H* parallel to the *c* axis. Here we wish to emphasize the connection of the characteristic temperatures observed in our transport measurements and the various phase-transition temperatures observed in the neutron-scattering studies.[3] The Hall coefficient gradually increases upon cooling from room temperature down to about 130 K. In the region of the incommensurate LSDW phase, a small "dip" is developed which is centered at about 115 K. Between 56 and 103 K (the AF1 state), a giant *U*-shape dip in the Hall coefficient is observed which yields sharp peaks at 103 and 56 K, respectively. Below 56 K, the Hall coefficient monotonically decreases with decreasing tem-

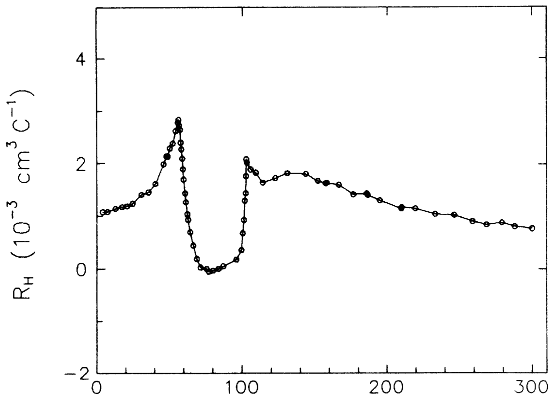

FIG. 4. Temperature dependence of the Hall coefficient with the current parallel to the *ab* plane and the magnetic field *H* along the *c* axis. Measurement was taken with a 1.6-T magnetic field. The line only serves as a guide to the eye.

perature. It is worth pointing out that 56 K is also the temperature at which the peak in the *c*-axis resistivity starts to develop. The Hall coefficient is 7.5×10^{-4} $\mathrm{cm}^3/^\circ\mathrm{C}$ at room temperature and has a value of 1.05×10^{-3} $\mathrm{cm}^3/^\circ\mathrm{C}$ at 4.2 K. The minimum of the Hall coefficient occurs at about 77 K with a value of -7.5×10^{-5} $\mathrm{cm}^3/^\circ\mathrm{C}$. Note that the value is small but negative in the vicinity of 77 K. The magnetic-field strength for the Hall-effect measurement was 1.6 T.

To conclude, we summarize our experimental findings as follows: (i) The resistivity is anisotropic over the entire temperature range investigated (4.2–300 K). The magnitude of the resistivity along the *c* axis is larger by a factor of 1.5 (room temperature) to 3 (4.2 K) than that in the *ab* plane. At high temperatures ($T > 150$ K), the temperature coefficient $d\rho/dT$ is small and positive for the *ab*-plane resistivity and is negative for the *c*-axis resistivity. This seems to suggest a stronger Kondo effect along the *c* axis. (ii) There is a drastic decrease in resistivity at about 123 K which is associated with the onset of the incommensurate LSDW. At the Néel temperature $T_N = 103$ K, the second anomaly develops which manifests itself as a "knee" in the *ab*-plane resistivity and a more drastic decrease in the *c*-axis resistivity. These are attributed to the suppression of spin-flip and spin-disorder scatterings by the magnetic ordering. (iii) At about 56 K, at which the system is about to undergo a phase transition from the AF1 state to a commensurate LSDW state, a peak starts to develop in the *c*-axis resistivity, much like the anomaly in the heavy-fermion compound $\mathrm{URu_2Si_2}$.[4–7] However, the peak in $\mathrm{UNi_2Si_2}$ is much broader and is absent in the *ab*-plane resistivity. Such results also illustrate the necessity of using a single-crystal sample since measurements on polycrystalline samples will be dominated by the behavior of the *ab*-plane resistivity and will show no anomaly. (iv) All the features and anomalies observed in the resistivity and Hall effect results are closely related to the presence and transitions of the three magnetic phases.[3] Also, the transport properties are similar to those of heavy fermion metals.[17] However, a definite conclusion as to whether $\mathrm{UNi_2Si_2}$ belongs to the family of heavy-fermion materials will have to wait until the specific-heat measurement becomes available.

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[1] L. Chelmicki, J. Leciejewicz, and A. Zygmunt, J. Phys. Chem. Solids **46**, 529 (1985).

[2] M. W. McElfresh, M. S. Torikachvili, H. Borges, and K. Reilly (unpublished).

[3] H. Lin, M. F. Collins, T. E. Mason, L. Rebelsky, and W. Buyers (unpublished); see also Proceedings of the 1990 Congress of Canadian Association of Physicists, May 1990, Abstracts, Physics in Canada Vol. 46 (unpublished), No. 3.

[4] M. B. Maple, J. W. Chen, Y. Dalichaouch, T. Kohara, C. Rossel, and M. S. Torikachvili, Phys. Rev. Lett. **56**, 185 (1986).

[5] T. T. M. Plastra, A. A. Menovsky, and J. A. Mydosh, Phys. Rev. B **33**, 6527 (1986).

[6] W. Schlabitz, J. Baumann, B. Pollit, U. Rauchschwalbe, H. M. Mayer, U. Ahlheim, and C. D. Bredl, Z. Phys. B **62**, 171 (1986).

[7] A. Ler Dawson, W. R. Datars, J. D. Garrett, and F. S. Razavi, J. Phys. Condens. Matter **1**, 6817 (1989).

[8] For a comprehensive review, see P. A. Lee, T. M. Rice, J. W. Serene, L. J. Sham, and J. W. Wilkins, Comments Condens. Matter Phys. **12**, 99 (1986).

[9] B. R. Coles, Contemp. Phys. **28**, 143 (1987).

[10] A. M. Umarji, J. V. Yakhmi, C. V. Tomy, R. M. Iyer, L. C. Gupta, and R. Vijayaraghavan, in *Theoretical and Experimental Aspects of Valence Fluctuations and Heavy Fermions,* Proceedings of the Fifth International Conference on Valence Fluctuations, Bangalore, India, 1987, edited by L. C. Gupta and S. K. Malih (Plenum, New York, 1987).

[11] A. J. Dirkmaat, Ph.D. thesis, Leiden, 1989 (unpublished).

[12] O. Rapp, G. Benediktsson, H. U. Astrom, S. Arals, and K. V. Rao, Phys. Rev. B **18**, 3665 (1978).

[13] Y. Suezaki and H. Mori, Phys. Lett. **28A**, 70 (1969); see also, Prog. Theor. Phys. (Kyoto) **41**, 1177 (1969).

[14] R. J. Elliott and F. A. Wedgwood, Proc. Phys. Soc. London **81**, 846 (1963).

[15] H. Miwa, Prog. Theor. Phys. (Kyoto) **29**, 477 (1963).

[16] M. Ausloos, J. Phys. F **6**, 1723 (1976).

[17] H. R. Ott, Ann. Rev. Mater. Sci. **17**, 13 (1987).