Suppression of simple antiferromagnetism in UNi_2Si_2 under high pressure

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UNi_2Si_2 appears successively in three magnetically ordered phases with decreasing temperature below $T_N = 124$ K. The intermediate phase, which exists in a temperature range of $43 < T < 103$ K at ambient pressure is the simple antiferromagnetic (AF1) phase. The temperature dependence of the electrical resistivity in this compound shows distinct anomalies at related magnetic-phase transitions at T_N, $T_1 = 43$ K and $T_2 = 103$ K. We have measured the electrical resistance for current parallel to the c axis ($i \| c$) in a UNi_2Si_2 single crystal under high pressures up to 4.0 GPa. With increasing pressure, T_1 increases and T_2 decreases at rates of 8.6 K/GPa and -1.3 K/GPa, respectively, which yields gradual shrinking of the temperature range of stability of the AF1 phase. The critical pressure for suppression of simple antiferromagnetism in UNi_2Si_2 has been determined as 3.2 GPa. On the other hand, T_N seems to be almost pressure independent. From these results, a more complete pressure-temperature magnetic phase diagram is constructed.

UNi_2Si_2 crystallizes in the tetragonal $ThCr_2Si_2$ type structure, which consists of the U and Ni_2Si_2 layers alternating along the c axis.[1] This compound orders magnetically at 124 K in incommensurate longitudinal spin density wave (ILSDW) propagating along the c axis. By decreasing temperature below T_N, it exhibits a second magnetic phase transition at $T_2 = 103$ K to a simple body-centered antiferromagnetic (AF1) state and finally at $T_1 = 53$ K it orders in a uncompensated antiferromagnetic (UAF) state with $q = (0,0,2/3)^2$ ($T_1 = 43$ K according to Refs. 3 and 4). Only U magnetic moments are present in the UNi_2Si_2 ordered magnetic phases always parallel along the c axis and ferromagnetically coupled within the basal plane. Coupling along the c axis varying with temperature then yields the different magnetic phases mentioned above.

Electrical resistivity is highly anisotropic, which reflects anisotropy in coupling of U moments. The three distinct anomalies in the resistivity (ρ) along the c axis can be associated with the magnetic phase transitions, which makes resistivity measurements particularly suitable for pressure-induced effects on magnetic phase transition temperatures.[5]

Recently suppression of the AF1 phase with a critical pressure of ~ 2.4 GPa has been predicted based on the electrical resistivity measurements up to 1.2 GPa.[6] On the other hand, linear extrapolation of $T_1(p)$ and $T_2(p)$ obtained from our previous measurements under pressures up to 2.2 GPa led us to a conclusion that the AF1 phase is to vanish at about 4 GPa.[4]

In the present work, we have measured the electrical resistance of UNi_2Si_2 along the c axis under high pressures up to 4 GPa in order to determine the stability of these magnetic phases at high pressure and construct a p-T magnetic phase diagram of this compound.

The single crystal of UNi_2Si_2 was grown by means of the triarc Czochralski technique. The sample cut parallel to the crystallographic c axis was used for electrical resistivity measurements with $i \| c$. Measurements under high pressures up to 2.2 GPa using a Cu-Be piston cylinder device were performed at Kumamoto University.[7] Above 2.0 GPa, the experiment was carried out at ISSP, the University of Tokyo using a cubic anvil-type high-pressure cell.[8] In both cases, the pressure transmitting medium was a 1:1 mixture of Fluorinert FC70:FC77. The electrical resistivity was measured by means of a dc four-probe method with electrical current applied parallel to the c axis. The sample dimensions were $1.2 \times 0.3 \times 0.2$ mm^3 in the low-pressure ($p \leqslant 2.2$ GPa) experiment and $0.7 \times 0.1 \times 0.1$ mm^3 for the high-pressure ($p \geqslant 2.0$ GPa) study. The samples were cut into the bar along the c axis.

The temperature dependence of the electrical resistivity from two experiments is shown for selected pressures in Figs. 1(a) and 1(b), respectively. The inset shows a temperature derivative of electrical resistivity in heating process at ambient pressure. Above 130 K, the $\rho(T)$ curve exhibits a gradual upturn with decreasing temperature at all pressures, i.e., $d\rho/dT$ is negative. Below T_N, the resistivity starts to

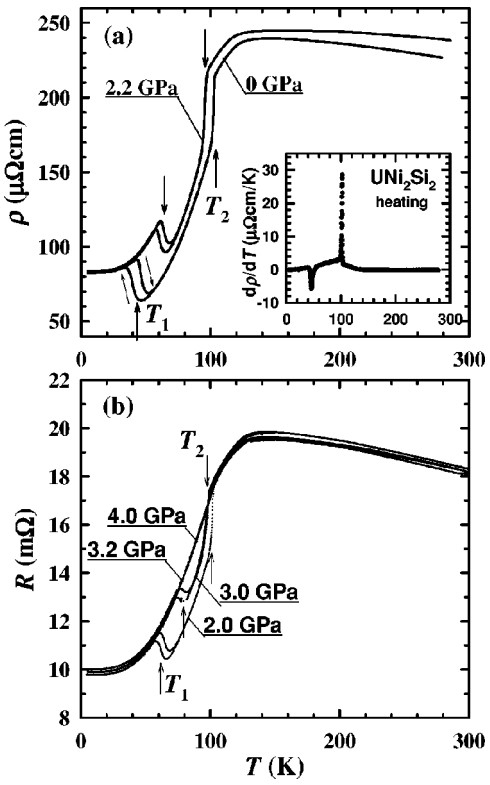

FIG. 1. (a) Temperature dependence of the electrical resistivity in a wide temperature range at 0 and 2.2 GPa. The inset shows the $d\rho/dT$ vs T plot at ambient pressure. The magnetic phase transitions are indicated by arrows. (b) The temperature dependence of the electrical resistance in the same temperature range under high pressure up to 4.0 GPa.

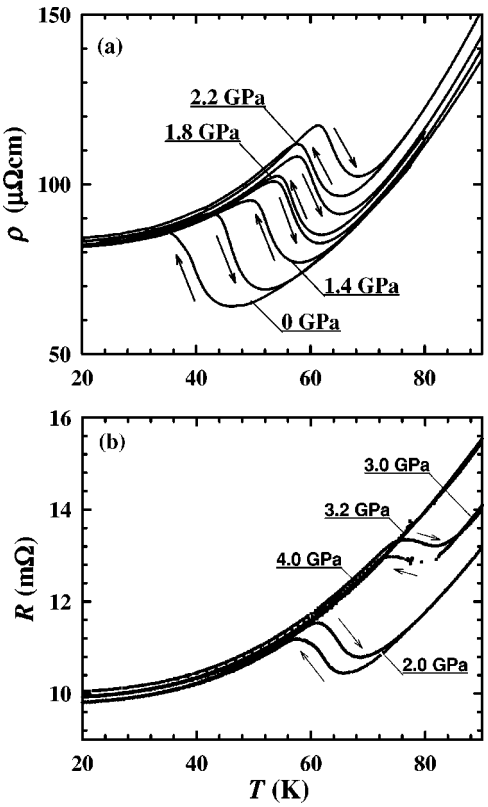

FIG. 2. Effect of pressure on the temperature dependence of the electrical resistance around T_1 in cooling and heating processes [(a) $p \leqslant 2.2$ GPa and (b) $2.0 \leqslant p \leqslant 4.0$ GPa]. The hysteresis region becomes narrower with increasing pressure.

decrease due to the magnetic ordering (ILSDW) and $d\rho/dT$ vs T dependence has a shoulder at T_N. As temperature decreases, the resistivity drops suddenly at $T_2 = 103$ K. This is connected with order-order magnetic phase transition from ILSDW to the simple AF1 phase and reflects the dramatic change of the periodicity of the magnetic structure along the *c*-axis. The temperature of this phase transition T_2 can be easily determined owing to a spike in the $d\rho/dT$ vs T plot.

The phase transition from the AF1 phase to the UAF structure at T_1 is accompanied by a pronounced increase of the resistivity. This transition manifests itself as a sharp minimum in the $d\rho/dT$ curve and exhibits a large temperature hysteresis. In the magnetic phase diagram presented below, we took T_1 as an average of phase transition temperatures observed within cooling and heating processes, respectively. At ambient pressure, the hysteresis appears in the range of $32 < T < 54$ K and T_1 is determined to be 43 K. These features are consistent with the data published previously.[2] The residual resistivity ratio (RRR) in the low-pressure experiment was $\simeq 3$ when measured at 2.2 GPa, whereas at 2.0 GPa in the high-pressure experiment RRR $\simeq 2$. This discrepancy may be attributed to additional lattice defects introduced by shaping and polishing the tiny small sample for the latter experiment.

More detailed plots focused on pressure effects on the transition at T_1 and T_2 are displayed in Figs. 2 and 3, respectively. The transition at T_1 is rapidly pushed to higher temperatures with increasing pressure, $dT_1/dp = 8.6$ K/GPa. Thus the UAF phase becomes stabilized with applying pressure. Simultaneously, the width of hysteresis becomes narrower ($72 < T < 82$ K at 3.0 GPa, which is about a half of that observed at ambient pressure), nevertheless, the hysteresis persists up to 3.0 GPa.

On the other hand, T_2 decreases with pressure at a rate of -1.3 K/GPa and the related $\rho(T)$ anomaly becomes gradually broadened. The resistivity changes at both transitions become gradually reduced and vanish above 3.2 GPa, which indicates that the intermediate AF1 phase is completely suppressed. At 3.2 GPa only a small anomaly appears on the $R(T)$ curve around 100 K, which we attribute to the order-order magnetic phase transition between the ILSDW and UAF phases. Since this anomaly shifted to higher temperature with increasing pressure ($T_2 = 102$ K at 4.0 GPa), the UAF phase seems to become stabilized also on account of the ILSDW. We expect that above 4.0 GPa also the ILSDW phase becomes completely suppressed and the UAF phase remains below T_N.

These results allow us to complete the pressure-temperature magnetic phase diagram of UNi_2Si_2 as shown in Fig. 4. As described above the temperature interval of stability of the UAF phase becomes gradually enlarged with pressure on account of the AF1 phase and at higher pressures also on account of the ILSDW. This result is in a qualitative agreement with the proposed p-T magnetic phase diagram in Ref. 6, but the AF1 phase disappears at 3.2 GPa, which is different from that in Ref. 6. From the thermodynamical point of view, the stability of UAF phase is also in good

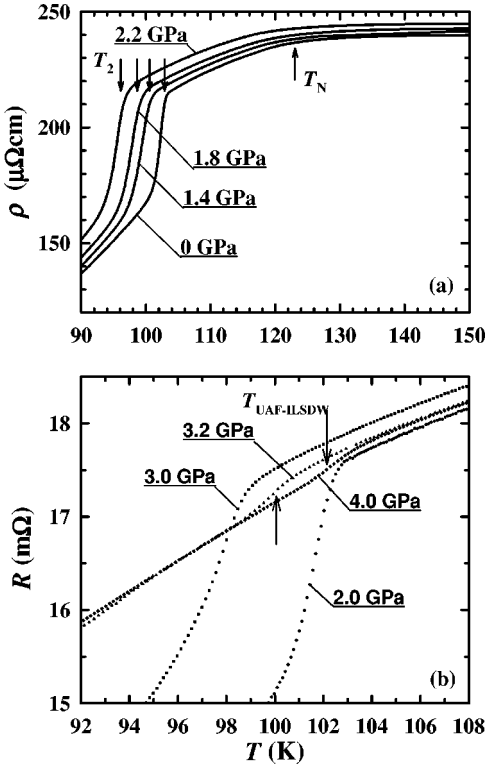

FIG. 3. Temperature dependence of the electrical resistance around T_2 and T_N under high pressure (a) $p \leqslant 2.2$ GPa and (b) $2.0 \leqslant p \leqslant 4.0$ GPa. A new anomaly, which is indicated by arrows in the electrical resistance, appears around 100 K reflecting the magnetic order-order phase transition between ILSDW and UAF phases.

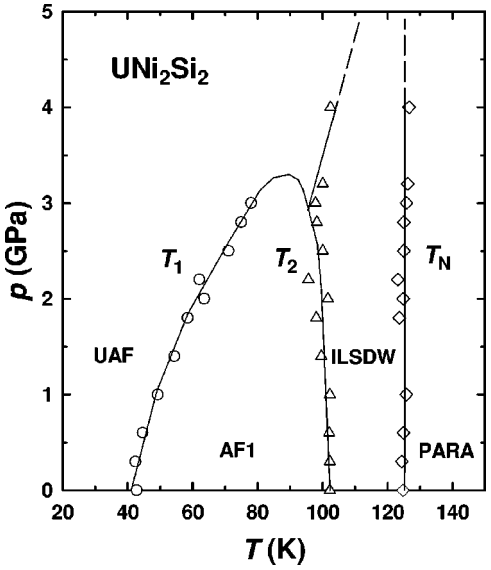

FIG. 4. A p-T magnetic phase diagram of UNi_2Si_2 up to 4.0 GPa. The lines are guides to the eye. T_1 increases and T_2 decreases with increasing pressure. The intermediate antiferromagnetic AF1 phase disappears at $\simeq 3.2$ GPa. The borderline of this phase boundary is quite flat. On the other hand, T_N is almost pressure independent up to 4.0 GPa.

agreement with our previous thermal expansion study at ambient pressure,[9] i.e., the UAF phase has the smallest volume among all magnetic phases of this compound because the external pressure usually stabilizes low-volume (high-density) state.

This p-T magnetic phase diagram is formally very similar to the magnetic-field-temperature (B-T) phase diagram at ambient pressure.[3,10,11] That is to say that UAF phase is stabilized and AF1 phase is suppressed by both external forces, say pressure and magnetic field. This indicates that there is a close relation between lattice and magnetic properties in this compound. In order to clarify the magnetoelastic properties of UNi_2Si_2, thermal expansion measurement under high pressure is in progress. Neutron-scattering studies are also strongly desirable in order to check the pressure-induced variations of the ILSDW propagation along the c axis. When magnetic field is applied along the c axis, the c axis component of q_{ILSDW} is gradually approaching to q_{UAF}.

In conclusion, we have constructed a p-T magnetic phase diagram under high pressure up to 4.0 GPa. The intermediate simple antiferromagnetic (AF1) phase, which exists in a range $43 \leqslant T \leqslant 103$ K at ambient pressure is suppressed by an external pressure of 3.2 GPa. The ground state UAF phase tends to stabilize at high pressure.

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