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LETTER TO THE EDITOR

Structural transformation in UNi_2Sn: a magnetic, transport, thermal and x-ray study

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Abstract. We have studied the magnetisation, resistivity and specific heat of a new U-based Heusler compound UNi_2Sn. This compound exhibits a structural transition around $T = 220$ K where both the magnetisation and the resistivity show a large jump and a hysteresis of approximately 30 K between cooling and heating. X-ray diffraction confirms that the transition is structural and to a crystallographic phase of much lower symmetry. Below the transition, indications of Kondo-like behaviour are observed.

The rare-earth ternary Heusler alloys have attracted much attention because of the rich variety in their physical properties [1, 2]. Structural transformations, superconductivity and magnetism are commonly observed in these compounds. In U- and Ce-based Heusler alloys both structural transformations and heavy-fermion behaviour were found. For example, $CeCu_2In$ [3] is a heavy-fermion compound, while in $CeAu_2In$ a martensitic transformation was revealed at around $T = 210$ K [2] along with a relatively large γ of 130 mJ mol^{-1} K^{-2} at low temperature [4]. UAu_2In was characterised as a localised moment antiferromagnet with Kondo-type spin fluctuations [5]. Recently it was shown that UPd_2In exhibits both a structural transformation (at $T = 180$ K) and heavy-fermion behaviour (with $\gamma = 200$ mJ mol^{-1} K^{-2}) at low temperatures [6].

We have prepared a new cubic U-based Heusler alloy, UNi_2Sn, and we have examined its electronic properties via magnetisation, resistivity and specific heat measurements. At around 220 K the compound exhibits a structural transformation to a crystallographical phase of much lower symmetry. The latter is clearly revealed by x-ray diffraction at room and liquid nitrogen temperatures. The magnetic behaviour is also influenced by the structural transformation: a Kondo-like state appears at low T, whereas the high-temperature phase is Pauli paramagnetic.

The sample was prepared by arc-melting in an atmosphere of purified argon gas. After arc-melting, the sample was wrapped in Ta foil and vacuum-annealed inside a quartz tube for four weeks at 800 °C. Electron probe microanalysis disclosed the sample to be predominantly single phase with the proper 1–2–1 composition. Magnetisation in fields up to 5 T was measured using a foner-type vibrating-sample magnetometer.

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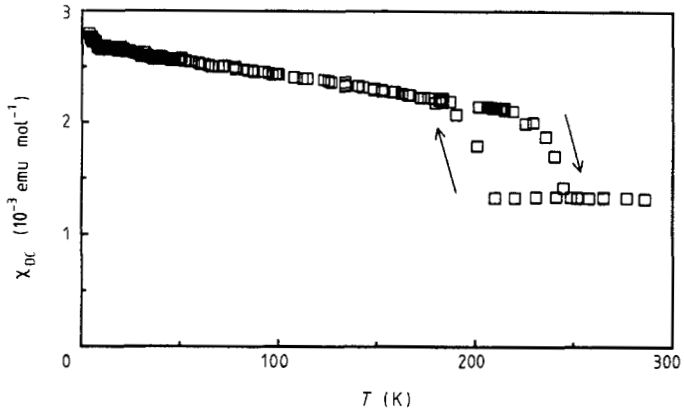

Figure 1. DC susceptibility ($\chi = M/H$) versus temperature for UNi_2Sn, measured in a field of $\mu_0 H = 2$ T.

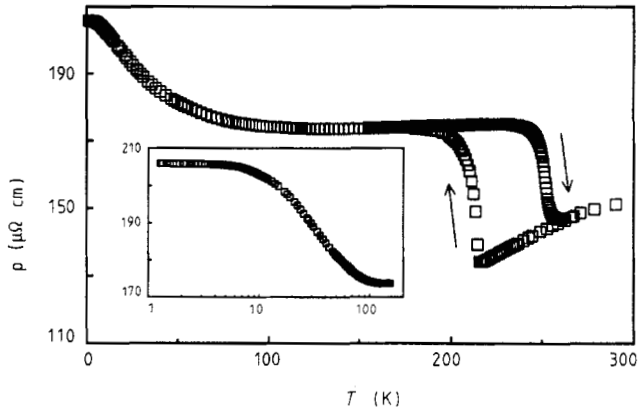

Figure 2. Resistivity versus temperature for UNi_2Sn. The inset plots the data on a logarithmic T-scale.

Resistivity was determined with a standard four-point DC technique, and the specific heat using the adiabatic heat-pulse method.

The magnetisation measured in a magnetic field of $\mu_0 H = 2$ T is shown in figure 1. Its magnitude is rather small and its temperature dependence does not obey a Curie–Weiss law. A step-like increase in the magnetisation is seen at 210 K upon cooling, and a sharp drop at 240 K upon heating. Temperature sweeps around the transition performed with different magnetic fields (0.4 T, 2 T and 5 T) show that the magnetic field neither influences the magnitude of the susceptibility ($\chi = M/H$) nor the temperatures of the transitions. Furthermore the magnetisation versus field isotherms (not shown) do not exhibit any structure and are nearly linear both at $T = 2$ K and at 215 K (on the upper as well as on the lower branch of the magnetisation curve).

The resistivity is displayed in figure 2 over the entire temperature range. The room temperature resistivity of 152 $\mu\Omega$ cm is comparable to that of UPd_2Sn: 190 $\mu\Omega$ cm [7] and UPd_2In: 130 $\mu\Omega$ cm [6]. Upon cooling, the resistivity first decreases, but below

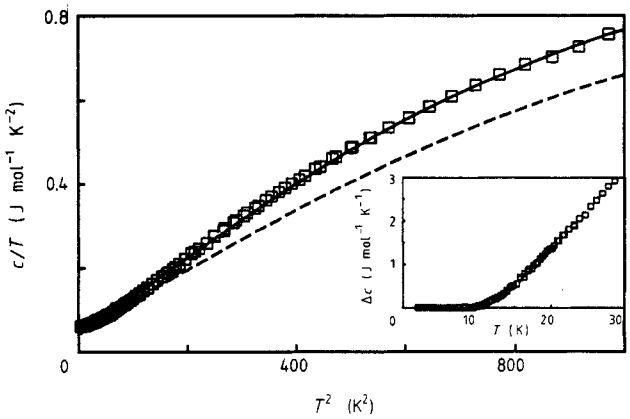

Figure 3. Specific heat divided by the temperature versus temperature squared for UNi_2Sn. The full curve is the best overall fit, using electronic plus full Debye contributions. The broken curve is a better two-contribution fit to the low-T (<10 K) data. The inset shows the difference between this fit and the data (see the text).

215 K there is a steep increase of ≈30%. Then ρ remains almost constant until below 80 K where it starts rising again. Finally below 5 K, $\rho(T)$ saturates at a value of 206 $\mu\Omega$ cm. Upon heating the resistivity is fully reversible up to ≈180 K and it sharply returns to the high-temperature branch at ≈250 K. The slightly different transition temperatures found in the magnetisation and resistivity measurement may reflect the different strains in the samples caused by the spark erosion forming. The inset of figure 2 gives the resistivity on a logarithmic temperature scale. Between the two T-independent regimes $\rho(T)$ is proportional to $-\log T$ for $10\,\text{K} \leqslant T \leqslant 70\,\text{K}$. Further analysis of the magnetic resistivity, without knowing the phonon contribution, is ambiguous.

The specific heat plotted as c/T versus T^2 from $T = 2$ K to $T = 31$ K is shown in figure 3. The linear electronic term as obtained by fitting the data with a linear term plus a *full* Debye function amounts to $\gamma = 39.7\ \text{mJ mol}^{-1}\,\text{K}^{-2}$. This fit yields a Debye temperature of $\theta_D = 203$ K. Although the above fit seems reasonable on the scale of figure 3, there are deviations, especially at the lowest temperatures (see below).

Figure 4 shows the x-ray (Cu Kα) diffraction patterns at room temperature and at 80 K. The room temperature diffractogram could be indexed on the basis of a cubic Heusler structure with lattice parameter $a = 6.459$ Å, yielding a U–U interatomic distance of 4.567 Å. In contrast, the low-temperature spectrum (below the transformation) reveals a large number of diffraction lines, indicating a lower-symmetry crystal, but an exact determination of its structure by fitting all the lines was not possible. It is inconceivable that the many new lines are due to a simple tetragonal distortion of the lattice. Instead a much lower symmetry is inferred. We are currently examining the possibility of an incomplete transformation, such that at low temperature a combination of reflections from a slightly distorted cubic and another phase are observed.

Although the U–U separation in UNi_2Sn is large and comparable to that in UPd_2In (4.81 Å [6]), no local moment behaviour is observed in UNi_2Sn. Above the transition, the perfectly flat susceptibility and the positive temperature coefficient of the resistivity indicate that UNi_2Sn in the cubic Heusler phase is a normal Pauli paramagnet. However, both the suceptibility and the resistivity show a dramatic change in behaviour at the

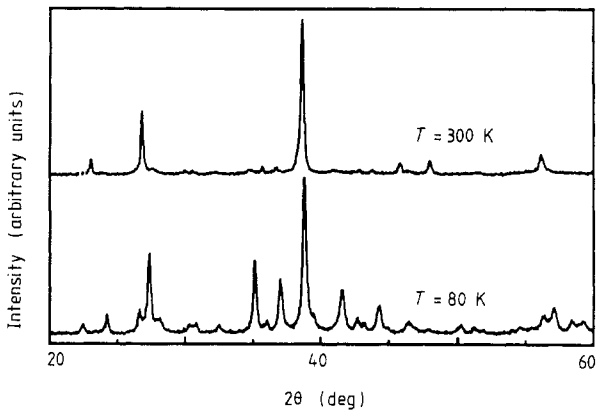

Figure 4. X-ray diffractograms of UNi_2Sn at 300 K and at 80 K.

transition. The increase at the transition of the susceptibility and its dependence on T below, illustrate that the low-T phase is 'more magnetic' than the high-T one. The logarithmic increase of the resistivity suggests that below the transition Kondo screening of the moments takes place. The interatomic distances play an important role in the formation of lcoal moments, which is a well known feature of uranium compounds. Since the U–U interatomic distance is large in this compound, the unexpected Pauli paramagnetic behaviour at high temperature is probably due to hybridisation of the U 5f bands with the Ni 3d bands. It is possible that the U–Ni separation increases during the crystallographic transformation or that the U environment completely changes. This then could give rise to the formation of local moments, which however become Kondo screened by the conduction electrons as the temperature is lowered.

The hysteresis of approximately 30 K observed in the susceptibility and resistivity is remarkably large and indicates the stability or pinning of the new phase against thermal excitations. Until now such a large hysteresis has not been reported for U or Ce compounds.

As mentioned above, our given fit of the specific heat deviates from the measured low-T data. An alternative approach restricts the upper bound of the fit to 10 K and attributes the high-temperature deviations to a magnetic contribution. This extrapolated low-T fit (with $\gamma = 50.1$ mJ mol^{-1} K^{-2} and $\theta_D = 220$ K) is shown as a broken curve in figure 3, while the inset shows the corresponding excess (magnetic) specific heat. It is unclear whether this extra contribution can be attributed to the Kondo effect, because of the limited temperature region of the measurement and since the low-temperature tail of a possible Kondo peak will modify our low-T, two-parameter (γ and θ_D) fit. Moreover, crystal-field effects, giving rise to Schottky contributions, cannot be excluded.

In conclusion, we have found that the new Heusler alloy UNi_2Sn undergoes a structural, symmetry-lowering transition at around 220 K, which strongly influences the susceptibility and the resistivity. No magnetic order was found down to 1.3 K, but indications of Kondo-like behaviour were found below the phase transition.

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