Magnetic properties of polycrystalline UNi_2Ge_2: irreversibility and metastable behaviour

1994 J. Phys.: Condens. Matter 6 5155

(http://iopscience.iop.org/0953-8984/6/27/023)

J. Phys.: Condens. Matter 6 (1994) 5155–5160. Printed in the UK

Magnetic properties of polycrystalline UNi_2Ge_2: irreversibility and metastable behaviour

S B Roy, A K Pradhan and P Chaddah

Low Temperature Physics Group, Centre for Advanced Technology, Indore 452 013, India

Received 11 November 1993, in final form 4 March 1994

Abstract. We present a detailed magnetization study on the intermetallic compound UNi_2Ge_2 which orders antiferromagnetically around $T_N \sim 78$ K. Distinct irreversibility and metastable behaviour are observed below T_N in applied fields $H \leqslant 1$ T. With small Ce doping, T_N is slightly suppressed but the irreversibility becomes more prominent. Our study highlights the difficulty in defining a few specific macroscopic magnetic characteristics which can be used to distinguish an antiferromagnet unequivocally from a spin glass.

1. Introduction

The intermetallic compound UNi_2Ge_2 with $ThCr_2Si_2$ structure orders antiferromagnetically around $T_N \sim 78$ K. This magnetic transition shows up quite clearly in the measurements of thermodynamic and transport properties [1, 2, 3]. Specific heat measurements yield an electronic coefficient of specific heat $\gamma \sim 40$ mJ mol^{-1} K^{-2} below T_N at low temperatures while above T_N, γ was found to be ~ 84 mJ mol^{-1} K^{-2}. Such a change in electronic specific heat accompanying magnetic transitions has been observed in UCd_{11} , U_2Zn_{17} [4] and UIr_2Si_2 [5] and was attributed to the opening of an energy gap over part of the Fermi surface. Magnetization measurements with external fields $H \geqslant 0.3$ T revealed around 78 K a typical peak, commonly associated with antiferromagnetic ordering [1, 3]. The behaviour of the resistivity is interesting in that it shows a weak negative temperature coefficient. This feature along with the reduced (with respect to the free U ion value) magnetic moment $3.08\mu_B$ in the paramagnetic state [2, 3] hints at the possible role of Kondo screening in this system. Below about 80 K, the resistivity drops sharply indicating the strong reduction of spin disorder scattering as a result of the magnetic transition. Neutron measurements [1] performed at 4.2 K suggest an antiferromagnetic order of collinear AF1 type. This measurement also suggests that the magnetic moments are localized on uranium atoms, the moment directions being aligned along the c-axis; the ordered magnetic moment on the nickel ion, if it exists at all, is less than $0.3\mu_B$.

In this paper we shall present results of our detailed magnetization and resistivity measurements revealing new features, which to our knowledge have not been reported earlier.

2. Experimental details

The compounds (UNi_2Ge_2 and $(U_{0.9}Ce_{0.1})Ni_2Ge_2$) used in the present study were prepared by argon arc melting from metals of at least nominal 99.99% purity, subsequently subjected

0953-8984/94/275155+06$19.50

to annnealing at 700 °C for 5 d and characterized by x-ray and metallographic studies. While x-ray study revealed only lines of tetragonal $ThCr_2Si_2$ structure, metallography indicated that the amount of second phase if any was less than 5% of the bulk matrix. The resistivity measurements were performed using the standard DC four-probe method. The magnetization measurements to be reported here were performed using a commercial SQUID magnetometer (MPMS5, Quantum Design). The sample used in the present measurements was a square cross-section rod of dimension 5 mm ×1mm ×1 mm and it was mounted firmly in the sample holder with its long axis parallel to the external magnetic field. The same sample position and configuration was strictly adhered to, during all the measurements to be reported here. This step is necessary to eliminate the orientation effect in our polycrystalline sample due to the presence of inherent anisotropy. The scan length we used was 4 cm and measurements were averaged over three scans each containing 32 data points. This relatively short scan length minimizes the inhomogeneity of the magnetic field through which the sample travels. Before each measurement special care was taken to reduce the trapped field in the superconducting magnet of the SQUID magnetometer. We followed the procedure suggested in the instruction manual (from Quantum Design) by ramping down the field to zero value in an oscillatory mode. This usually gives a trapped field of about ±0.3 – 0.4 mT. We found the magnetic response due to this trapped field was at least one order of magnitude less than that in our lowest field of measurement i.e. 10 mT.

3. Results and discussion

In figure 1, we present magnetization (M) versus temperature (T) plots of UNi_2Ge_2 in external fields $H = 10$ mT, 50 mT and 0.3 T in both the zero-field-cooled (ZFC) and field-cooled (FC) conditions. The behaviour for $H = 0.3$ T (see figure 1(c)) is essentially the same as what has already been reported [1, 3]. The peak temperature of 80 K agrees reasonably well with the literature value [1, 3]; however, the ZFC and FC magnetization splits below a temperature $T_{\mathrm{irrev}} \sim 55$ K. With lower applied fields, the irreversibility in magnetization becomes more prominent and T_{irrev} goes up in temperature. For $H = 10$ mT (see figure 1(a)) strong irreversibility crops up just below T_N. On the other hand with $H = 1$ T (not shown in figure 1) we could not observe any irreversibility (at least in the scale of figure 1(c)) down to 10 K. The observed irreversibility is much more drastic in the case of a 10% Ce doped alloy $(U_{0.9}\,Ce_{0.1})Ni_2Ge_2$ (see figure 2). Here the character in FC behaviour below T_N changes markedly with the external field. As shown in figure 2(a) the FC behaviour (at T_N and below) with $H = 10$ mT is more reminiscent of a ferromagnetic type rather than an antiferromagnetic transition, while that with $H = 50$ mT (figure 2(b)) is very similar to canonical spin glasses. FC behaviour acquires a conventional antiferromagnetic character with $H \geqslant 0.1$ T, however as shown in figure 2(c) substantial irreversibility remains even with $H = 0.3$ T (in comparison to UNi_2Ge_2 with the same external field). Magnetization (M) as a function of field (H) (see figure 3) measured at $T = 10$ K shows a linear behaviour (typical of an antiferromagnet) up to 5 T for both UNi_2Ge_2 and $(U_{0.9}Ce_{0.1})Ni_2Ge_2$.

Observation of such irreversibility indicates that at least in the field regime $H \leqslant 0.3$ T, the low-temperature magnetic response of UNi_2Ge_2 is metastable in character. To support this conjecture we have measured zero-field-cooled magnetization (M_{ZFC}) as a function of time; a representative measurement is shown in figure 4, with H kept fixed at 10 mT and T at 10 K. A distinct increase in M_{ZFC} is clearly observed. (During these measurements, the external field was in the persistent mode of the superconducting magnet, thus ruling out any fluctuations/instability from external sources).

In figure 5 we present the results of our resistivity measurements performed on UNi_2Ge_2

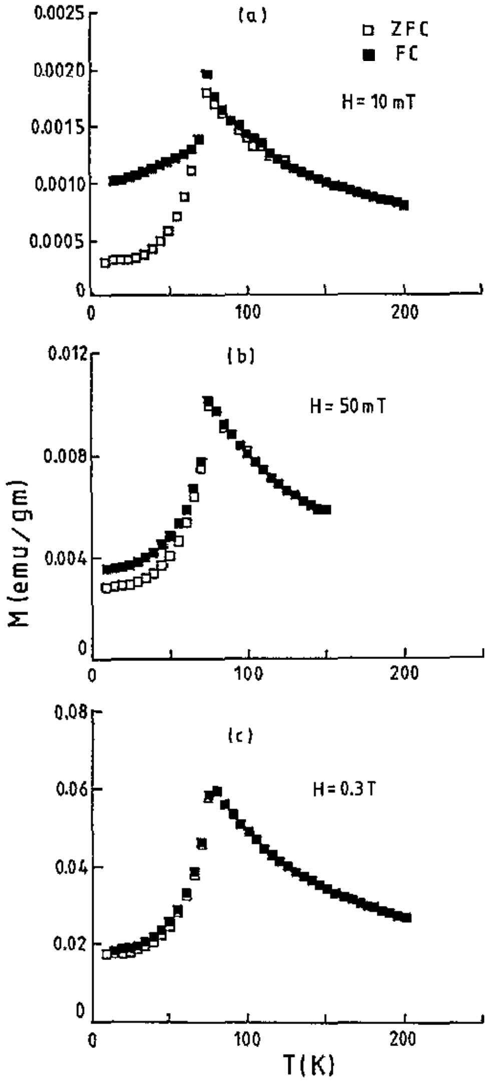

Figure 1. Magnetization (M) (both ZFC and FC) versus temperature (T) plots of UNi_2Ge_2 in various external magnetic fields.

and $(U_{0.9}Ce_{0.1})Ni_2Ge_2$. Note that the absolute value of resistivity of UNi_2Ge_2 is slightly higher in our case (in comparison to the reported value in the literature [3]), but the resistivity ratio ($R_{300\,K}/R_0$) is roughly the same. However there is a new feature in our resistivity results, which shows a very distinct increase in the resistivity of UNi_2Ge_2 at $T_N \sim 80$ K leading to a peak at 70 K before decreasing very rapidly. The same feature was observed in the 10% Ce doped alloy, with a reduced value of T_N. The other analogous uranium based antiferromagnets, i.e. UCd_{11} and U_2Zn_{17}, did not show such structure around T_N in their resistivity results [4]. This behaviour is rather reminiscent of the appearance of a superzone effect [6, 7, 8] due to the onset of SDW type antiferromagnetic ordering, as in the case of Cr. A structural distortion accompanying the magnetic ordering can also give rise to initial increase in resistivity below T_N. However in the absence of any structural study in this temperature regime, no further comment can be made in this regard.

The picture which emerges from the above study suggests that UNi_2Ge_2 is an antiferromagnet with certain features in the low-temperature regime and low-field regime,

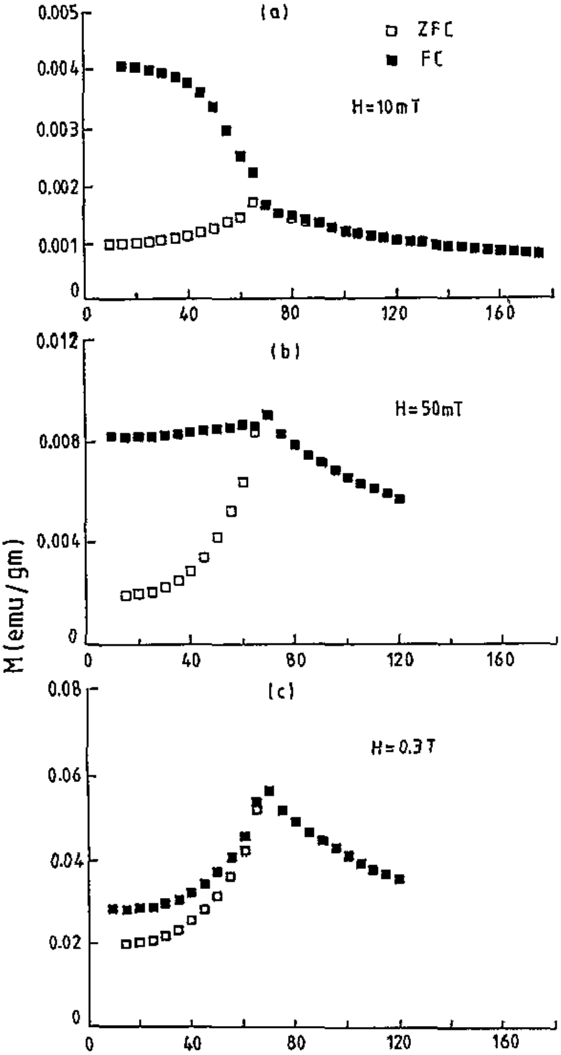

Figure 2. Magnetization (M) (both ZFC and FC) versus temperature (T) plots of $U_{0.9}Ce_{0.1}Ni_2Ge_2$ in various external magnetic fields.

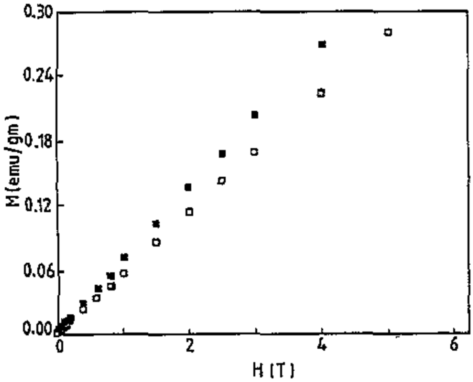

Figure 3. Magnetization (M) versus magnetic field (H) plot of UNi_2Ge_2 (□) and $U_{0.9}Ce_{0.1}Ni_2Ge_2$ (■) at $T = 10$ K.

namely irreversibility and time relaxation, which are generally associated with spin glasses

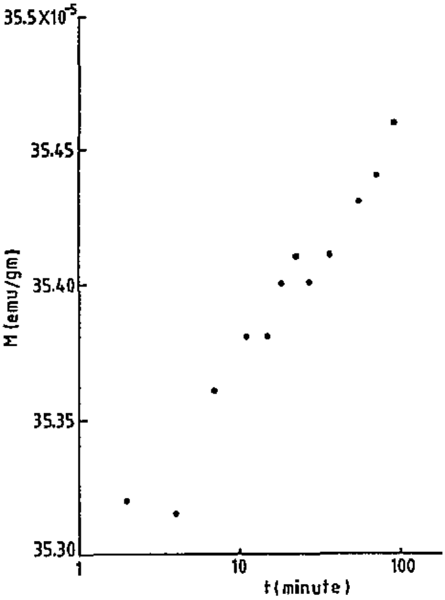

Figure 4. Zero-field-cooled (ZFC) magnetization (M) versus time (t) for UNi_2Ge_2 with H being kept constant at 10 mT and T at 10 K.

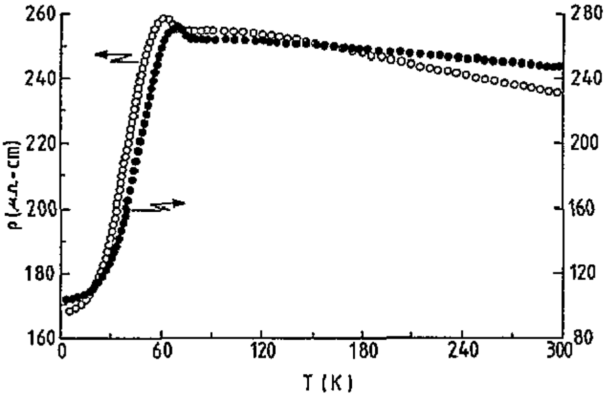

Figure 5. Resistivity (ρ) versus temperature (T) plots of UNi_2Ge_2 (•) and $U_{0.9}Ce_{0.1}Ni_2Ge_2$ (○).

and random field Ising systems [9]. At the first inspection, randomness and frustration, which are thought to be the essential ingredients for these spin-glass-like features, seem to be absent, at least in pure UNi_2Ge_2. So what is the source of these unusual features? The common character of the magnetic structures found in the U 1-2-2 compounds with $ThCr_2Si_2$ structure is the presence of ferromagnetically ordered moments in the (001) planes which are normal to the tetragonal axis i.e. the c-axis [2]. The $ThCr_2Si_2$ type structure suggests a highly anisotropic character of the magnetic interactions and the magnetic moments are aligned along the c-axis leading to an Ising like behaviour. The coupling between the planes is weaker than the coupling within the planes and the magnetic structure can be explained in terms of the ANNNI model [10]. Although the antiferromagnetic ordering of UNi_2Ge_2 finds a natural explanation within such a model, explanation of the thermomagnetic history effects observed in the present study is beyond the scope of the ANNNI model. It is now commonly accepted that the magnetization of U 1-2-2 compounds is the result of a complex interplay between the Ruderman–Kittel–Kasuya–Yosida (RKKY) type of interaction, Kondo screening of the moments and the anisotropy field. One possible source of the anisotropy field is the Dzyaloshinsky–Moriya (DM) interaction [11], which has its origin in the spin–orbit interaction. Such an anisotropy was considered by many authors to be essential for the existence of macroscopic irreversibility in metallic spin glasses and re-entrant spin glasses [12]. A polycrystalline sample of UNi_2Ge_2, with various orientations of the highly anisotropic crystallites, seems to emulate here many of the magnetic properties of spin glasses. A very similar situation also arose in minnesotaite $[Si_4]\{Fe_3^{2+}\}O_{10}(OH)_2$ which orders as a planar antiferromagnet below $T_N = 28$ K [13] with strong evidence of anisotropic interactions. It must be noted here that frustration in a Kondo lattice system might arise due to the RKKY interaction alone [14] hence suggesting the possibility of another source of macroscopic irreversibility in UNi_2Ge_2.

4. Conclusions

In conclusion we would like to say that the bulk property measurements in UNi_2Ge_2 reveal certain characteristics of the magnetic state which do not conform to a simple antiferromagnetic behaviour. Results presented here illustrate the difficulty in defining a few specific macroscopic magnetic characteristics which can be used to distinguish an antiferromagnetic system unequivocally from a spin glass. On completion of our present work we became aware of a magnetic and transport property study by Ning *et al* [15] on a single crystal of UNi_2Ge_2. The results of the resistivity measurements along the c-axis are very similar to ours. Regarding the magnetic measurements, both the qualitative and quantitative aspects at and around T_N agree well, but in our study we do not find any signature of a ferromagnetic transition down to 10 K. Ning *et al* [15] however did not report any study on the thermomagnetic history effects which form a major part in our present investigation. In the light of present findings, further macroscopic magnetic studies on single crystals of UNi_2Ge_2 and detailed neutron measurements will be very informative.

References

[1] Chelmicki L, Leciejewicz J and Zygmunt A 1985 *J. Phys. Chem. Solids* **46** 529
[2] Endstra T, Mentink S A M, Nieuwenhuys G J and Mydosh J A 1992 *Frontiers in Solid State Sciences* vol 2, ed L C Gupta and M S Multani (Singapore: World Scientific) p 167
[3] Dirkmaat A J, Endstra T, Knetsch E A, Menovsky A A, Nieuwenhuys G J and Mydosh J A 1990 *J. Magn. Magn. Mater.* **84** 143
[4] Stewart G R 1984 *Rev. Mod. Phys.* **56** 755
[5] Dirkmaat A J, Endstra T, Knetsch E A, Nieuwenhuys G J, Mydosh J A, Menovsky A A, de Boer F R and Tarnawski Z 1990 *Phys. Rev.* B **41** 2589
[6] Coles B R 1958 *Adv. Phys.* **7** 40
[7] Mackintosh A R 1962 *Phys. Rev. Lett.* **9** 90
[8] Suezaki Y and Mori H 1969 *Prog. Theor. Phys.* **41** 1177
[9] Fischer K H and Hertz J 1991 *Spin-Glasses* (Cambridge: Cambridge University Press)
[10] McAlister S P, Olivier M and Siegrist T 1989 *Solid State Commun.* **69** 113
[11] Moriya T 1963 *Magnetism* vol 85, ed G T Rado and H Suhl (New York: Academic)
[12] Campbell I A and Senoussi S 1992 *Phil. Mag.* **65** 1267
[13] Coey J M D, Ballet O, Bakes T and Mangin P 1990 *J. Appl. Phys.* **67** 5258
[14] Lacroix J 1989 *J. Magn. Magn. Mater.* **100** 90
[15] Ning Y B, Garrett J D, Stager C V and Datars W R 1992 *Phys. Rev.* B **46** 8201