Ferromagnetic-to-antiferromagnetic transition in UCu_2Ge_2: Magnetoresistance study

A. K. Nigam
Tata Institute of Fundamental Research, Homi Bhabha Road, Bombay 400005, India

S. B. Roy
Centre for Advanced Technology, Indore 452012, India

Girish Chandra
Tata Institute of Fundamental Research, Homi Bhabha Road, Bombay 400005, India

(Received 6 July 1993)

We report a detailed study of magnetoresistance in the intermetallic compounds UCu_2Ge_2 and $(U_{0.95}Ce_{0.05})Cu_2Ge_2$ in the temperature range of 4.5 to 140 K and in magnetic fields up to 50 kOe. The present measurements support the earlier conjecture drawn from the magnetic studies that the ferromagnetic to antiferromagnetic transition takes place gradually over a wide temperature range. The magnetic-field dependence of magnetoresistance shows that the low-temperature antiferromagnetic state is more fragile in UCu_2Ge_2 than in $(U_{0.95}Ce_{0.05})Cu_2Ge_2$.

I. INTRODUCTION

The magnetic study of the intermetallic compound UCu_2Ge_2 (with the $ThCr_2Si_2$ type crystal structure) shows interesting features as a function of temperature. This system orders ferromagnetically below 100–110 K.[1–3] However, the magnetic ordering at low temperatures (certainly below 20 K) seems to be antiferromagnetic as is indicated from neutron scattering measurements.[1,4] There is no clear consensus about the temperature where the transition from ferro- to antiferromagnetic ordering takes place. It is claimed to be 25–30 K,[1] 43 K,[2] or 65 K (Ref. 3) by different authors through their measurements. It has been argued recently that the ferro- to antiferromagnetic transition in UCu_2Ge_2 is probably a gradual one taking place over a broad temperature range.[5] Theoretically, such a possibility of gradual magnetic transition and coexistence of ferro- to antiferromagnetism has been considered in strongly interacting itinerant-electron systems.[6] The system UCu_2Ge_2, where the uranium $5f$ wave functions are quite extended and hybridize substantially with the conduction band providing both itinerant character and strong electronic correlation (reminiscent of the strongly correlated electrons in the isostructural compound $CeRu_2Si_2$, a heavy fermion system), seems to possess essential requirements for the occurrence of such phenomena. Further support in this regard has been obtained from recent measurements of nonlinear susceptibility in UCu_2Ge_2.[7] From these measurements, it is quite clear that in a broad temperature regime of 55 K–80 K, there exists no clear-cut characterisitics of either ferromagnetic or antiferromagnetic ordering. In fact the observed features in that temperature regime are more reminiscent of random magnetic ordering or spin glass type of behavior. Also, it is to be noted here that in the specific heat study, while the onset of T_c is marked with a distinct maxima at 108 K, no sharp anomaly has been observed in the lower-temperature regime.[4]

To shed further light on the interesting magnetic properties of UCu_2Ge_2, we undertook a detailed study of the magnetoresistance measurements in this compound as well as in $(U_{0.95}Ce_{0.05})Cu_2Ge_2$. This study has revealed more interesting aspects related to magnetic phases of this unusual system. In subsequent sections, we present the results of our magnetoresistance measurements and correlate our findings with the existing experimental picture.

II. EXPERIMENTAL DETAILS

The alloys were prepared by argon arc melting from metals of at least nominal 99.99% purity and suction chill casting in copper moulds to produce square cross section rods. The samples were subjected to metallographic analysis to investigate the possible presence of second phase. It is to be noted here that the samples have not been annealed. However, as far as the resistivity and magnetization studies are concerned, there seems to be not much difference between our sample[5] and a well annealed one.[2] Even the subtle structure in resistivity around 60 K could be observed in our sample. It seems that unlike in UCo_2Ge_2,[4] the physical properties of UCu_2Ge_2 do not depend on the heat treatment.

The longitudinal magnetoresistance measurements were carried out in the temperature range of 4.5 K–140 K up to a field of 50 kOe generated by a home-built superconducting magnet. The magnetoresistance was measured using standard four probe dc technique. The electrical contacts to the sample were made with indium solder using ultrasonic soldering. The resistivity changes of the order of 50 ppm could be detected in the present setup. The temperature of the sample was controlled

0163-1829/94/49(2)/1127(4)/$06.00

and monitored by a Lake Shore carbon glass sensor (in magnetic field) up to 60 K and by a silicon diode sensor beyond 60 K, employing a Lake-Shore DRC-82C temperature controller. The measurements were automated using an IBM compatible PC/AT through an IEEE-488 interface.

III. RESULTS AND DISCUSSION

A. UCu_2Ge_2

Figures 1(a)–1(f) give the magnetic-field (H) dependence of magnetoresistance at various temperatures in the UCu_2Ge_2 compound. The sample was cooled to 4.5 K in zero external magnetic field and the data were recorded by increasing the field in steps at discrete intervals up to around 50 kOe. The field was then decreased in steps and the data recorded. For subsequent temperatures above 4.5 K, the samples were heated to those temperatures in zero field and the data taken in the same manner as described above. Figures 1(a)–1(c) show that this compound shows a metamagnetic transition indicated by a sharp increase in $|\Delta\rho/\rho|$ in a certain field range. Magnetization measurements on UCu_2Ge_2 at 4.2 K showed a metamagnetic transition around 5.5 kOe.[1] In the present study, we find that $\Delta\rho/\rho$ is almost negligible up to around 5.5 kOe which is expected in a weak antiferromagnetic system. The increase in $\Delta\rho/\rho$ after the above field is similar to the behavior of magnetization in the same field range. Also, $\Delta\rho/\rho$ shows strong hysteresis effect as has also been seen in magnetization measurements.[1] It is interesting to note that while decreasing the field at 4.5 K,

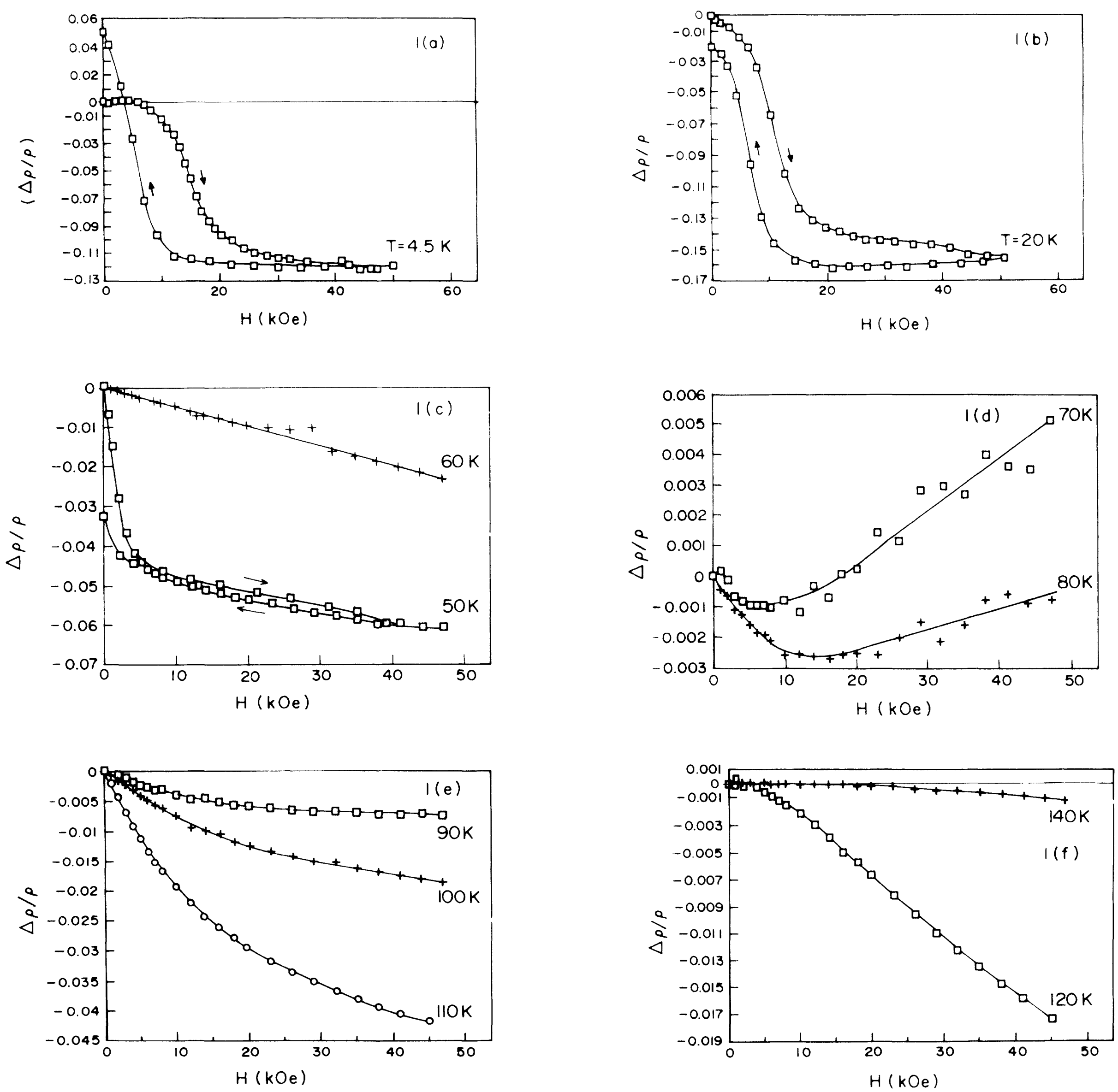

FIG. 1. (a)–(f) Magnetic field (H) dependence of magnetoresistance ($\Delta\rho/\rho$) in UCu_2Ge_2 at different temperatures. The arrows indicate the data taken in increasing and decreasing fields.

$\Delta\rho/\rho$ crosses the initial curve (obtained during increasing field) and becomes positive below about 3 kOe which is at present difficult to explain. At all the other temperatures below 50 K, the decreasing field curve is always below that taken in the increasing field which is expected when the sample has a hysteresis effect. The appearance of metamagnetic transition up to 50 K as shown in Fig. 1(c) indicates that antiferromagnetic state exists below this temperature range. This is also confirmed by magnetization measurements where a maximum is seen in the temperature range of 50–60 K in fields of few hundred gauss.[1] In the temperature range of 10–40 K, one finds that at low fields, before the metamagnetic transition, $\Delta\rho/\rho \propto H^n (n > 1)$ as can be seen at 20 K in Fig. 1(b). This dependence is similar to that observed in random magnetic systems such as spin glasses, etc.[8] It therefore implies that there is coexistence of both ferromagnetic and antiferromagnetic interactions leading to a random magnetic state. The behavior of $\Delta\rho/\rho$ as a function of magnetic field in the temperature range of 60–80 K shown in Figs. 1(c) and 1(d) is quite interesting. At 60 K, the metamagnetic transition disappears and almost a linear dependence of $\Delta\rho/\rho$ on H is observed. The magnetic field dependence of $\Delta\rho/\rho$ at 70 and 80 K shows initially a decrease of $\Delta\rho/\rho$ and then takes an upturn with increasing field. At 70 K, $\Delta\rho/\rho$ is positive at fields above around 20 kOe. The magnetoresistance in this temperature range (60–80 K) is quite small. The dc magnetization data of this compound in the above temperature range exhibits a weak temperature dependence.[3,5] Also the magnetic-field dependence of magnetization in this temperature range is typical of a ferromagnet.[3] In such a case, the magnetoresistance due to spin-disorder scattering would be very small. The observed upturn in $\Delta\rho/\rho$ at 70 K and 80 K could therefore result from the dominating influence of magnetic field on the Fermi surface. However, recent measurements of nonlinear susceptibility on UCu_2Ge_2 indicate a spin-glass-like state between 55 K and 75 K which might occur as a result of competing ferro- and antiferromagnetic interactions.[7] If such a spin glass state has strong random magnetic structure, then also the magnetoresistance due to spin-disorder scattering would be weak. Under this situation also, one would observe the above mentioned magnetoresistance behavior but then it is difficult to explain the observed field dependence of magnetization mentioned above.[3]

The field dependence of magnetoresistance in the temperature range of 90 K and 110 K is shown in Fig. 1(e) and is very much similar to that of an inhomogeneous ferromagnet such as AuFe above the percolation limit.[9] In this temperature range, $|\Delta\rho/\rho|$ is found to be maximum at 110 K which is close to the Curie temperature. The compound seems to have short range ferromagnetic correlations since even at 120 K, $\Delta\rho/\rho$ does not depend quadratically on H [Fig. 1(f)]. It is to be noted here that the resistivity of UCu_2Ge_2 shows distinct negative curvature above T_c,[2,5] which is also indicative of the presence of spin fluctuations. At 140 K, $\Delta\rho/\rho$ is very small and depends nearly quadratically on H indicating a paramagnetic state.

B. $(U_{0.95}Ce_{0.05})Cu_2Ge_2$

Figures 2(a)–2(c) show the magnetic-field dependence of $\Delta\rho/\rho$ at different temperatures $(U_{0.95}Ce_{0.05})Cu_2Ge_2$. The behavior is very similar to that of UCu_2Ge_2 except that its temperature dependence has the following salient features different from UCu_2Ge_2.

(1) The metamagnetic transition is found up to 90 K as against up to 50 K in UCu_2Ge_2. Also the fields at which metamagnetic transition occurs at different tem-

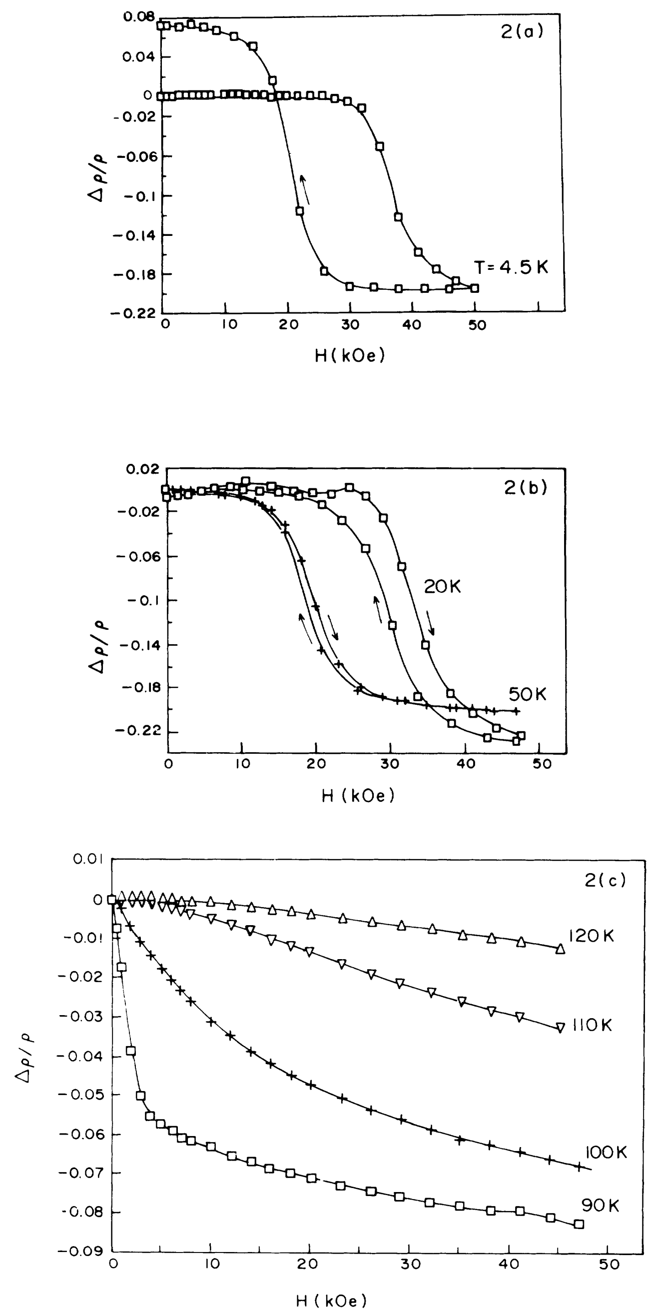

FIG. 2. (a)–(c) Magnetic field (H) dependence of magnetoresistance ($\Delta\rho/\rho$) in $(U_{0.95}Ce_{0.05})Cu_2Ge_2$ at different temperatures. The arrows indicate the data taken in increasing and decreasing fields.

peratures are much higher than those in UCu_2Ge_2. For example, at 4.5 K, it is around 6 kOe for UCu_2Ge_2 while in $(U_{0.95}Ce_{0.05})Cu_2Ge_2$ it is around 30 kOe. These observations lead us to believe that the antiferromagnetic state persists up to much higher temperatures in $(U_{0.95}Ce_{0.05})Cu_2Ge_2$ and is more stable and stronger than in UCu_2Ge_2 as had also been inferred by earlier magnetic measuremments.[5]

(2) Only at 100 K does $\Delta\rho/\rho$ show a field dependence similar to that of inhomogeneous ferromagnets as shown in Fig. 2(c). At 110 K, $\Delta\rho/\rho$ at low fields tends towards quadratic behavior indicating paramagnetic state setting in. It therefore implies that the ferromagnetism appears in a narrower range of temperature (between 90 and 110 K) in $(U_{0.95}Ce_{0.05})Cu_2Ge_2$ as compared to that of UCu_2Ge_2. This also confirms the results of magnetization measurements on the same compound where the onset of spin canting has been found to occur at 90 K.[5] However, it seems that the transition from antiferromagnetic to ferromagnetic state is more distinct in $(U_{0.95}Ce_{0.05})Cu_2Ge_2$ than in UCu_2Ge_2 where $\Delta\rho/\rho$ is quite small and also changes sign at certain fields in the temperature range of 60–80 K.

IV. CONCLUSIONS

The present magnetoresistance study supports the earlier conjecture drawn from magnetic studies that the ferromagnetic-to-antiferromagnetic transition takes place gradually over a broad temperature range. In UCu_2Ge_2, the nature of magnetic ordering in the temperature range between 50 K and 80 K still remains controversial since magnetoresistance is very small and changes sign. Such a behavior could result either from a ferromagnetic state or from a strongly random magnetic structure. The field dependence of magnetoresistance shows that the low-temperature antiferromagnetic state in UCu_2Ge_2 is quite fragile and is easily turned into a ferromagnetic one by the application of a field of a few kOe. On the other hand, the cerium substitution weakens the ferromagnetic state as can be seen by the metamagnetic transition taking place at much higher field in $(U_{0.95}Ce_{0.05})Cu_2Ge_2$.

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

[2] A.J. Dirkmaat, T. Endstra, E.A. Knetsch, A.A. Menovsky, G.J. Nieuwenhuys, and J.A. Mydosh, J. Magn. Magn. Mater. **84**, 143 (1990).

[3] S.P. McAlister, M. Olivier, and T. Siegrist, Solid State Commun. **69**, 113 (1989).

[4] T. Endstra, J.A.M. Mentink, G.J. Nieuwenhuys, and J.A. Mydosh, in *Frontiers in Solid State Sciences*, edited by L.C. Gupta and M.S. Multani (World Scientific, Singapore, 1992).

[5] S.B. Roy and B.R. Coles, Philos. Mag. **64**, 741 (1991).

[6] T. Moriya and K. Usami, Solid State Commun. **23**, 935 (1977).

[7] A. Chakravarti, R. Ranganathan, and S.B. Roy, Phys. Rev. B **46**, 6236 (1992).

[8] A.K. Nigam and A.K. Majumdar, Phys. Rev. B **27**, 495 (1983); S.P. McAlister and C.M. Hurd, *ibid.* **20**, 1273 (1979).

[9] A.K. Nigam, Girish Chandra, and S. Ramakrishnan, J. Phys. F **16**, 1255 (1986).