Magnetoresistance in a well-annealed sample of UCu_2Ge_2

A. K. Nigam
Tata Institute of Fundamental Research, Homi Bhabha Road, Mumbai 400 005, India

S. B. Roy and P. Chaddah
Centre for Advanced Technology, Indore 452 019, India

(Received 12 February 1999)

Results are presented of magnetoresistance measurements in a well-annealed sample of UCu_2Ge_2. Between the as cast sample and the annealed sample, a significant difference in the low temperature behavior has been observed. A fairly large positive magnetoresistance is observed in the temperature regime below ~50 K in the annealed sample whereas earlier study on the as cast sample had shown a metamagnetic behavior at low temperatures. The measurements suggest the presence of antiferromagnetic interactions in this compound, even in the well-annealed sample, which has been otherwise characterized as a ferromagnet.
[S0163-1829(99)11725-9]

I. INTRODUCTION

In UCu_2Ge_2, the onset of ferromagnetism occurs at a fairly high temperature (~105 K) as compared to the other isostructural uranium compounds. The magnetic measurements on the as cast and short time annealed samples have shown that ferromagnetic (FM) ordering around 105 K is followed at a lower temperature by the onset of antiferromagnetic (AF) ordering around 45–50 K.[1–3] This conjecture had the support of neutron scattering experiments.[1,2] The FM to AFM transition, however, seems to be taking place over a broad temperature range,[4] accompanied by various interesting properties in the crossover regime.[5,6] Our measurements of magnetoresistance on as cast sample were supportive of the AF state which is on the verge of FM instability as it showed metamagnetic transition at low fields.[6] There has been some controversy over the nature and existence of low temperature AF phase. The neutron diffraction studies carried out by some other groups reported that their samples remain ferromagnetic down to 5 K.[7–9] They attributed this to the sample preparation procedure of other authors where the measurements were carried out on the unannealed samples. It was argued that such samples might have second phase giving rise to the low temperature AF transition. It is worth pointing out that the Endstra *et al.*[2] carried out their neutron scattering studies on a well annealed and characterized sample[10] in which the impurity phase was below 1 vol% and exhibited AF transition below 55 K. Whereas the impurity phase reported in Ref. 7 for their sample was higher at 2% in which the low temperature AF transition had not been observed. Our recent study of the effect of heat treatment on the magnetization of UCu_2Ge_2, revealed that the lower temperature AF phase is quite marginal in nature and led us to conclude that its existence depends crucially on the heat treatment.[11] In the long time annealed sample, there is a small change in the unit cell volume and any signature suggestive of AF transition has been absent in bulk magnetization measurements. On the other hand, the change in the Cu/Ge stoichiometry has no effect on the existence of low temperature AF state in the as cast samples.[11]

In the recent neutron depolarization and neutron diffraction study, it has been suggested that the low temperature magnetic phase is that of randomly canted ferromagnet.[9] The observed distinct drop in the magnetization measurements at low temperatures on zero field cooled (ZFC) samples has often been taken as a signature of re-entrant spin glass or canted magnetic state. Such a behavior could also be explained in terms of the difficulty of domain wall motion and/or domain rotation. This last picture gains much strength especially when the field cooled magnetization (MFC) shows Brillouin type behavior typical of a ferromagnet.[11] In order to resolve the controversy about the low temperature phase of UCu_2Ge_2, we performed detailed magnetoresistance measurements on a well annealed sample. In recent years, the magnetoresistance measurements have shown that it is a fairly sensitive probe in distinguishing between various kinds of magnetic orders.[12,13] In the $CeFe_2$ based pseudo-binary compounds, the magnetoresistance studies could unambiguously identify ferro- to antiferromagnetic transition. These measurements have the advantage of probing the microscopic spin correlations occurring over the mean free path length. Also, unlike magnetization and susceptibility, magnetoresistance is not sensitive to small amount of impurity phases (which evades detection by standard x-ray diffraction technique) and represents the behavior of the majority phase primarily. The results of the magnetoresistance presented here on the well annealed sample of UCu_2Ge_2 have revealed that there exists distinct difference between the as cast and well annealed samples as far as the low temperature behavior (below 50 K) is concerned. The observation of large positive magnetoresistance in the low temperature range indicates the presence of antiferromagnetic interaction in the compound.

II. EXPERIMENTAL DETAIL

The samples were prepared by argon arc melting of constituent elements having 99.99% purity. They were subjected to annealing at 700 °C for 3 days followed by at 750 °C for five days and ultimately at 900 °C for 13 days. The details of

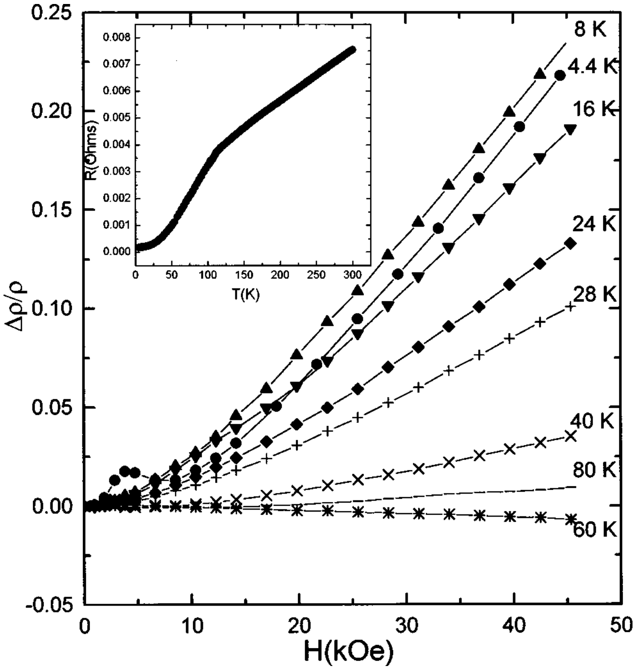

FIG. 1. Magnetic field (H) dependence of magnetoresistance ($\Delta\rho/\rho$) in UCu_2Ge_2 in the temperature range of 4.4 and 80 K. The inset shows the temperature dependence of resistance of the sample which shows a knee at the paramagnetic to ferromagnetic transition temperature around 110 K.

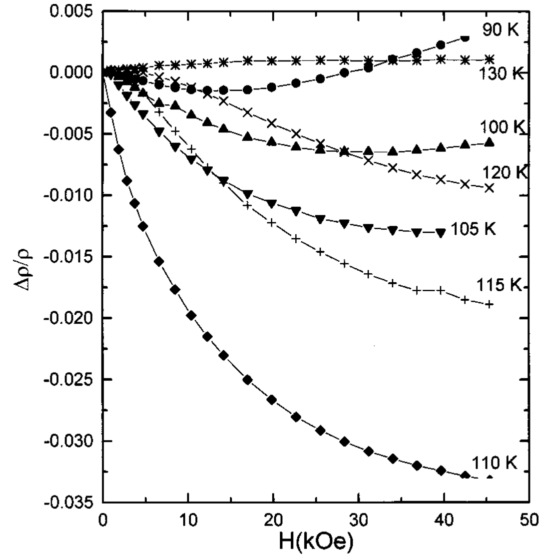

FIG. 2. Magnetic field dependence of magnetoresistance ($\Delta\rho/\rho$) in UCu_2Ge_2 in the temperature range from 90 to 130 K.

the sample characterization have been described in an earlier publication.[11]

The longitudinal magnetoresistance measurements were carried out in the temperature range of 4.5 K to 150 K in magnetic fields up to 45 kOe generated by a home-built superconducting magnet. The magnetoresistance was measured using a 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 set-up. The temperature of the sample was controlled and monitored by a Lake Shore carbon glass sensor (in magnetic field) up to 60 K and by a silicon diode sensor above 60 K, employing a Lake Shore DRC-82C temperature controller.

III. RESULTS AND DISCUSSION

In Figs. 1 and 2, the magnetic field (H) dependence of magnetoresistance $[\Delta\rho/\rho=(\rho(H)-\rho(0))/\rho(0)]$ has been plotted at various temperatures in the range 4.5 to 130 K. The inset in Fig. 1 shows the temperature (T) dependence of the electrical resistivity of the annealed sample which shows distinct knee close to the Curie temperature ($\approx$105 K) determined earlier in the same sample from the low field magnetization measurement.[11] In the magnetoresistance measurements the sample was cooled in zero external field and the data have been recorded by increasing the field in steps at discrete intervals up to 45 kOe.

In the temperature range 4.4 to 50 K, $\Delta\rho/\rho$ is found to be positive. It is recalled here that in the as cast sample a distinct metamagnetic transition was observed in this temperature regime.[6] In other words, $\Delta\rho/\rho$ was very small up to the metamagnetic transition field, above which it was negative due to the field induced ferromagnetism in the as cast sample. The maximum value of negative $\Delta\rho/\rho$, in this temperature range, was found to be around 16%. On the contrary, $\Delta\rho/\rho$ becomes positive in the annealed sample of UCu_2Ge_2. The magnetic field dependence of $\Delta\rho/\rho$ at low temperatures (between 4.4 and 40 K) could be described as

$$\Delta\rho/\rho \sim \alpha H+\beta H^2,$$

where α and β are constant dependent on temperature. The value of α is found to be almost two orders of magnitude higher than β. It implies a stronger linear dependence on H as against a quadratic dependence expected due to Fermi surface effects. In manganese also, a linear term in field dependence was seen in high fields (above 10 kOe) and attributed to be a characteristic of antiferromagnetic phase of α-Mn. The magnitude of $\Delta\rho/\rho$ is fairly large at low temperatures with a maximum value of 24% (in a field of 40 kOe) at 8 K. This large value of $\Delta\rho/\rho$ can not be attributed to the influence of magnetic field on the Fermi surface. It is interesting to note that the positive $\Delta\rho/\rho$ drops relatively rapidly with increasing temperature as one approaches the temperature regime of 50 K. It is to be recalled here that in the as cast sample, the ferro to antiferromagnetic transition was observed around 50 K. A similar large positive value of $\Delta\rho/\rho$ has been reported recently in antiferromagnetic compounds of $RE_2Ni_3Si_5$ where RE=Sm, Tb, and Nd.[14]

At 60 K, $\Delta\rho/\rho$ is negative and shows an almost linear field dependence. This small negative magnetoresistance could be due to the suppression of spin fluctuation which may be significant due to competing antiferro and ferromagnetic exchange interactions. The field dependence of $\Delta\rho/\rho$ in the temperature range 80–90 K shows initially a decrease of $\Delta\rho/\rho$, which then takes an upturn with the increase in field. The temperature dependence of magnetization in this regime is found to be that of a ferromagnet and is relatively weak.[11] The magnetoresistance due to spin disorder scattering is expected to be small in such a situation. As was in the case of the as cast sample of UCu_2Ge_2,[6] we conjecture that the positive magnetoresistance at 80 and 90 K results from the dominating influence of magnetic field on the Fermi surface.

In the temperature range 100 to 120 K (see Fig. 2), $\Delta\rho/\rho$ is found to be negative having a maximum value at 110 K, which is close to the Curie temperature $T_c \approx 105\,\mathrm{K}$, as determined from the low field magnetization measurements. This behavior is very similar to what had been observed in the as cast sample of UCu_2Ge_2 in the same temperature range.[6] The increase of negative $\Delta\rho/\rho$ above 90 K up to T_c is presumably due to increased influence of magnetic field in suppressing the spin disorder. This increased influence could be due to decrease (or absence) of AF interactions, which in turn reduces the frustration of spins, in this temperature range. In inhomogeneous ferromagnets, such as *Au*Fe just above the percolation concentration of long range magnetic order, similar behavior has been observed.[15] Signature of short range ferromagnetic correlations exists even at $T=115$ and 120 K where $\Delta\rho/\rho$ does not show the typical quadratic field dependence expected in a paramagnet (see Fig. 2).

In Fig. 3, the temperature dependence of $\Delta\rho/\rho$ is shown at 40 kOe. One observes a rapid decrease of $\Delta\rho/\rho$ up to around 50 K followed by a weak T dependence up to around 100 K. The negative magnetoresistance shows a maximum at T_c, as expected for a ferromagnet. The rapid increase of $\Delta\rho/\rho$ below around 50 K is qualitatively similar to what has been observed in α-Mn below the AF transition temperature.[16] It was attributed to the modification of the Fermi surface by the new Brillouin zone planes (formed as a result of AF ordering).

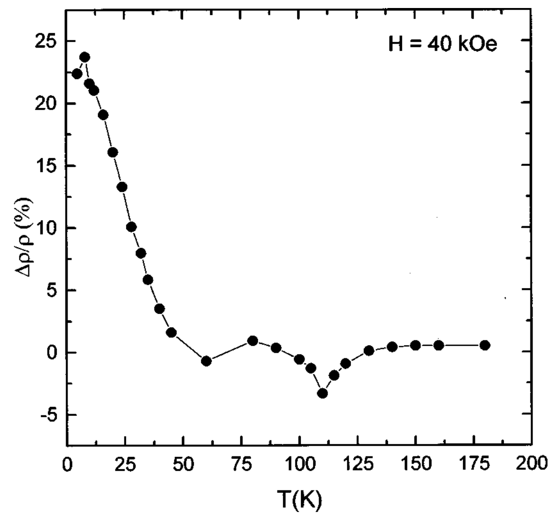

FIG. 3. Temperature dependence of magnetoresistance ($\Delta\rho/\rho$) in UCu_2Ge_2 at a field of 40 kOe. A maximum in the negative magnetoresistance is observed close to the Curie temperature at 110 K.

IV. CONCLUSION

The bulk magnetization on the well annealed sample of UCu_2Ge_2 revealed a typical ferromagnetic response in the temperature regime 5 K to 105 K.[11] The present observation of large positive magnetoresistance on the same sample in the temperature regime below 50 K, however, indicates the presence of distinct antiferromagnetic correlation over the mean free path length. While the recent neutron diffraction study on annealed samples of UCu_2Ge_2 suggested a randomly canted spin structure in the temperature regime $T \leqslant 50\,\mathrm{K}$,[9] such a structure is a far cry from an antiferromagnetic structure which is inferred from the observation of large positive magnetoresistance in the present study on a well annealed sample. One should also note here that in the as cast sample, the low temperature antiferromagnetic state (which was clearly observed both in the bulk magnetization and the neutron diffraction experiments) was quite sensitive to the applied magnetic field and a field induced ferromagnetic state (metamagnetic transition) was clearly observed in the magnetoresistance measurements in an earlier study.[6] So the present observation of fairly large positive magnetoresistance in the well annealed sample of UCu_2Ge_2 with no indication of any low temperature AF order in the bulk magnetization study, is certainly intriguing and demands more microscopic magnetic measurements.

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

[2] T. Endstra, S. 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), p. 167.

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

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

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

[6] A. K. Nigam, S. B. Roy, and Girish Chandra, Phys. Rev. B **49**, 1127 (1994).

[7] M. Kuznietz, H. Pinto, and M. Melamud, J. Magn. Magn. Mater. **83**, 321 (1990).

[8] M. Kuznietz, H. Pinto, H. Ettedgui, and M. Melamud, Phys. Rev. B **46**, 3183 (1993).

[9] S. M. Yusuf, L. Madhav Rao, and P. Raj, Phys. Rev. B **53**, 28 (1996).

[10] 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).

[11] S. B. Roy, A. K. Pradhan, P. Chaddah, and J. G. Park, Philos. Mag. B **75**, 303 (1997).

[12] S. Radha, A. K. Nigam, S. B. Roy, and G. Chandra, Phys. Rev. B **50**, 6866 (1994).

[13] H. P. Kunkel, X. Z. Zhou, P. A. Stampe, J. A. Cowen, and Gwyn Williams, Phys. Rev. B **53**, 15099 (1996).

[14] Chandan Majumdar, A. K. Nigam, R. Nagarajan, C. Godart, L. C. Gupta, B. D. Padalia, G. Chandra, and R. Vijayaraghvan, Appl. Phys. Lett. **68**, 3647 (1996).

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

[16] D. Griffiths and B. R. Coles, Proc. Phys. Soc. London **82**, 127 (1963).