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COMMENT

Comment on 'Commented review: UCu_2Ge_2 and UCu_2Si_2—compounds with only ferromagnetic ordering'

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Abstract
Certain experimental results pertaining to the low temperature magnetic state of UCu_2Ge_2, which are not covered in a recent 'Commented review' (Kuznietz 2003 *J. Phys.: Condens. Matter* **15** 8957), are discussed. These results need to be understood and explained within an accepted model of the magnetic state of this interesting compound.

The magnetic ground state of the intermetallic compound UCu_2Ge_2 is subjected to much scrutiny in a recent 'Commented review' [1]. One of the conclusions of that 'review' as well as an earlier 'Note' [2] is that the as-cast samples of UCu_2Ge_2 suffer from problems related to disorder and stoichiometry, and that this was the cause of the observed antiferromagnetic (AFM) state at low temperature. One of the present authors earlier expressed his reservations as regards this role of disorder and non-stoichiometry in the low temperature (T) magnetic state of as-cast UCu_2Ge_2 [3], and this is criticized in the 'Commented review' [1]. In that earlier work [3] it was also stated that 'Any argument that it [non-stoichiometry] does [play a role] will surely need support from studies of the effects of intentional variations, such as $Cu_{1.95}$ to $Cu_{2.05}$'. Indeed, such a study was subsequently taken up and the results of that study were published in an international journal [4]. In addition to that, a magnetoresistance study has been carried out on a well annealed sample of UCu_2Ge_2 [5]. The findings of these works (which are not mentioned in the 'Commented review' [1]) provide important information for building up a true balanced picture of the magnetic properties of UCu_2Ge_2, and they are discussed below.

A set of UCu_2Ge_2 samples were prepared by varying the Cu and Ge stoichiometry, and each of these samples was given a systematic heat treatment [4]. In the dc magnetization study all the as-cast samples and the samples annealed at 700 °C for three days showed the same qualitative features irrespective of their stoichiometry. Both the zero-field cooled (ZFC) and the field cooled (FC) magnetizations showed the low temperature downturn which was earlier identified with the onset of an antiferromagnetic (AFM) transition. The samples annealed at 750 °C for five days followed by annealing at 900 °C for thirteen days, irrespective of their stoichiometry, showed the typical behaviour of a ferromagnet (FM) in the temperature dependence of the FC magnetization. On the basis of these magnetization results, along with the x-ray diffraction (XRD) study, a conclusion was drawn that the non-stoichiometry cannot be the source of the qualitative difference in low temperature magnetic properties between as-cast and annealed samples of UCu_2Ge_2 [4]. Pechev *et al* [6], on the basis of a microprobe analysis of the as-cast UCu_2Ge_2 sample, reported one major phase with deviation from stoichiometry not larger than 1% and a parasitic phase, $U_3Cu_4Ge_4$. This parasitic phase is ferromagnetic in nature with a transition temperature of 71 K and gives rise to a bump in the temperature dependence of the magnetization of the as-cast UCu_2Ge_2 sample studied [6]. In our XRD measurement study, we had reported the presence of some faint lines which could not be identified with the $ThCr_2Si_2$ structure [4]. We did not observe any bump in the temperature dependence of the magnetization for our sample [4], which can probably be attributed to the sparsity of the data points. However, a large drop in magnetization in the low T regime [4, 6] cannot be explained in terms of a small amount of ferromagnetic impurity phase. Our XRD studies revealed a small change in volume on annealing the samples, and a possible role of this volume change in the magnetic properties of UCu_2Ge_2 involving interesting layered $ThCr_2Si_2$ structure was pointed out [4].

Following the dc magnetization measurements, a detailed magnetoresistance study was taken up to highlight the difference between the as-cast and annealed samples of UCu_2Ge_2. The annealed sample showed a significant positive magnetoresistance at low temperatures [5]. Such large positive magnetoresistance cannot be explained within a simple framework of a ferromagnetic ground state, and indicates the presence of antiferromagnetic correlation in some form even in the annealed sample of UCu_2Ge_2 [5]. It should be noted that, unlike magnetization and susceptibility, magnetoresistance is not sensitive to a small amount of impurity phase and represents the behaviour of the majority phase, primarily. Hence a marked difference in magnetoresistance behaviour between the as-cast [7] and annealed [5] samples cannot be attributed to a small amount of impurity phase.

Our interest in UCu_2Ge_2 was generated originally by the similarity in magnetic properties with Al-doped $CeFe_2$ alloys [8]. There was some confusion regarding the low T magnetic state of Al-doped $CeFe_2$ [9], but the AFM ground state of this compound is now well established [10, 11]. Some recent careful neutron scattering studies [12] have actually shown the presence of a dynamic antiferromagnetic correlation at low temperature in pure $CeFe_2$ itself, which has been identified as a ferromagnet since the early 1960s [13, 14]. It is worthwhile mentioning here that at low temperatures $CeFe_2$ shows positive magnetoresistance very similar to that observed for annealed samples of UCu_2Ge_2 [15].

In many classes of magnetic materials including the much studied manganites with colossal magnetoresistance, FM and AFM states are in competition, leading to a first-order phase transition [16]. This transition is accompanied by a FM–AFM phase coexistence and associated metastability over a large field (H)–temperature (T) regime [17]. Following a certain experimental H–T path it is possible to supercool the FM state down to the lowest temperature of measurement where, in the zero-field cooled condition, an equilibrium AFM state is observed [17]. In addition to that, sometimes the initial ZFC AFM state is lost after the first experimental cycle and the FM state remains (at least partially) frozen in all subsequent measurements [18–20]. It is now emerging that the intrinsic quenched-in disorder plays an important role in the phase transition process in various classes of magnetic materials, and that the magnetic states of such materials depend crucially on the experimental path followed in the H–T phase space [16]. In the light of this newer information it will be interesting to probe UCu_2Ge_2 more deeply, especially in its single-crystal form.

This discussion is by no means an attempt to bias the reader towards an AFM ground state (which may eventually turn out to be a mere diversion) for UCu_2Ge_2, and the present authors do not have any entrenched belief in this regard either. On the other hand, on the basis of present information on UCu_2Ge_2, a framework of a ferromagnetic ground state appears to be too simplistic to encompass all the available experimental results. And recent developments involving various classes of magnetic materials with competing FM–AFM interactions suggest that more experimental work on UCu_2Ge_2 is required before the matter can be deemed closed.

References

[1] Kuznietz M 2003 *J. Phys.: Condens. Matter* **15** 8957
[2] Kuznietz M, Pinto H and Melamud M 1993 *Phil. Mag.* B **68** 195
[3] Roy S B 1995 *Phil. Mag.* B **71** 105
[4] Roy S B, Pradhan A K, Chaddah P and Park J G 1997 *Phil. Mag.* B **75** 303
[5] Nigam A K, Roy S B and Chaddah P 1999 *Phys. Rev.* B **60** 3002
[6] Pechev S *et al* 1997 *J. Alloys Compounds* **243** 77
[7] Nigam A K, Roy S B and Chandra G 1994 *Phys. Rev.* B **49** 1127
[8] Roy S B and Coles B R 1991 *Phil. Mag.* B **64** 741
[9] Eynon J and Ali N 1991 *J. Appl. Phys.* **69** 5063 and references therein
[10] Kennedy S J and Coles B R 1990 *J. Phys.: Condens. Matter* **2** 1213
[11] Fernandez G J *et al* 1997 *J. Alloys Compounds* **261** 26 and references within
[12] Paolasini L *et al* 1998 *Phys. Rev.* B **58** 12117
Paolasini L *et al* 2003 *Phys. Rev. Lett.* **90** 057201
[13] Wallace W E 1964 *J. Chem. Phys.* **41** 3854
[14] Buschow K H J *Handbook of Ferromagnetic Materials* vol 1, ed E P Wohlfarth (Amsterdam: North-Holland) chapter 4
[15] Radha S, Roy S B and Nigam A K 2000 *J. Appl. Phys.* **87** 6803
[16] Dagotto E *et al* 2001 *Phys. Rep.* **344** 1
Dagotto E 2003 *The Physics of Manganites and Related Compounds* (Berlin: Springer) (and references within)
[17] Sokhey K J S *et al* 2004 *Solid State Commun.* **129** 19
Chattopadhyay M K *et al* 2003 *Phys. Rev.* B **68** 174404
[18] Manekar M A *et al* 2001 *Phys. Rev.* B **64** 104416
[19] Tang H *et al* 2004 *Phys. Rev.* B **69** 064410 and references therein
[20] Dho J and Hur N H 2003 *Phys. Rev.* B **67** 214414