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

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REPLY

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

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Abstract

In a reply to the preceding Comment by Roy *et al* (2005 *J. Phys.: Condens. Matter* **17** 3113) on 'Commented review: UCu_2Ge_2 and UCu_2Si_2—compounds with only ferromagnetic ordering', published recently by the present author (Kuznietz 2003 *J. Phys.: Condens. Matter* **15** 8957), the assertion that only ferromagnetic ordering occurs in UCu_2Ge_2, as observed by means of neutron diffraction and other methods, is stated and documented. None of the variety of experimental results on UCu_2Ge_2 produced by Roy *et al* and summarized briefly, but without any new neutron diffraction data, can contradict or serve as a basis for disputing that there is only ferromagnetic ordering in UCu_2Ge_2 in zero and low applied magnetic fields, as observed by means of neutron diffraction and ac susceptibility, respectively. The comparison between UCu_2Ge_2 and some $Ce(Fe,M)_2$ solid solutions made by Roy *et al*, and the similarities of some of their magnetic properties, are claimed to be coincidental, and not to lead to conclusions regarding UCu_2Ge_2 magnetism. Only new neutron diffraction data could truly justify such a Comment on the 'Commented review'.

The present reply refers to the preceding Comment by Roy *et al* [1], made on an article: 'Commented review: UCu_2Ge_2 and UCu_2Si_2—compounds with only ferromagnetic ordering' published recently in this journal by the present author [2]. The 'Commented review' was published in response to and in criticism of an earlier *J. Phys.: Condens. Matter* paper entitled 'Properties of Cu-flux-grown UCu_2Si_2' by Fisk *et al* [3]. Magnetization measurements made by Fisk *et al* [3] on such single crystals, leading them to claim 'a 50 K antiferromagnetic transition

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below the 100 K ferromagnetic transition', were shown in the 'Commented review' [2] to have been misinterpreted owing to the omission of any reference to ferromagnetic domain structure. The 'Commented review' was followed by a 'Reply to commented review from Kuznietz' by Fisk *et al* [4], which did not add any justification for their previous misinterpretation, and therefore did not merit any response by the present author.

In the 'Commented review' [2], the assertion that there only occurs ferromagnetic ordering (below $T_C = 107$ K) of annealed UCu_2Ge_2, a compound crystallizing in the body centred tetragonal (BCT) $ThCr_2Si_2$-type structure, was thoroughly discussed, and several claims by various authors of a low temperature (LT) antiferromagnetic (AF) state were disputed, and related either to the use of inappropriate samples (as-cast, incorrectly annealed or non-stoichiometric) and/or misinterpretation of results (omission of reference to the ferromagnetic domain structure etc).

That there is only ferromagnetic ordering in UCu_2Ge_2 was initially observed at the Nuclear Research Centre—Negev (NRCN), Beer-Sheva, Israel, by means of neutron diffraction [5], for this compound as well as for the solid solution $U(Co_{0.25}Cu_{0.75})_2Ge_2$. That there is only ferromagnetic ordering was later confirmed at the Bhabha Atomic Research Centre (BARC), Mumbai, India, by neutron depolarization, neutron diffraction, ac susceptibility and magnetization investigations [6–9], for UCu_2Ge_2 as well as for the solid solution $(U_{0.95}Th_{0.05})Cu_2Ge_2$. That there is only ferromagnetic ordering in UCu_2Ge_2 was supported also by Pechev *et al* [10] using their susceptibility and magnetization measurements. This was reconfirmed very recently at the NRCN by neutron diffraction, ac susceptibility and magnetization studies [11], for UCu_2Ge_2 as well as for the solid solutions $U(Co_{0.25}Cu_{0.75})_2Ge_2$ and $U(Co_{0.30}Cu_{0.70})_2Ge_2$. This recent work was quoted in the 'Commented review' as an article at press, but it has since been published [11]. It clearly demonstrates that the $U(Co_{1-x}Cu_x)_2Ge_2$ solid solutions with no LT AF states, $x = 1$ (UCu_2Ge_2), 0.75 and 0.70, behave quite differently to those with LT AF states, $x = 0.60$ and 0.50.

The continuous interest of Dr Roy and his co-authors/co-workers in UCu_2Ge_2 since 1991 has produced a variety of experimental results on this interesting compound, some of them following or in response to the above work on the ferromagnetism of UCu_2Ge_2 [5–11], but without any new neutron diffraction data. However, none of their results has been able to contradict or provide a basis for disputing the assertion that there is only ferromagnetic ordering in UCu_2Ge_2 in zero and low applied magnetic fields, as observed by means of neutron diffraction and ac susceptibility, respectively [5–11].

Roy and Coles submitted in May 1991 a paper (published in December 1991) [12] on 'Magnetic and electric properties of UCu_2Ge_2', with no reference to the NRCN work on this material published early in 1990 [5]. Roy and Coles [12] reported that their UCu_2Ge_2 sample, which was not annealed after casting, showed, from their magnetization study in an applied field of 0.01 T, ferromagnetic ordering below $T_C = 107$ K and a 'gradual transition from a ferromagnetic to an AF state over a large temperature range (around 43 K)', the latter leaving no trace in their ac susceptibility measurements. The conclusions of Roy and Coles [12] brought a sharp reaction from the NRCN group in 'Note on the magnetism of UCu_2Ge_2', submitted in May 1992 and published only in August 1993 [13]. The NRCN group ascribed the absence or appearance of an AF state at LT to variations in final stoichiometry due to different annealing conditions (or absence of annealing), and the 'AF behaviour' of the magnetization was ascribed to ferromagnetic domain effects.

In the long delay in publication of the NRCN Note [13] Roy and co-workers further studied their above-mentioned UCu_2Ge_2 sample, which was not annealed after casting, and reported on spin-glass-like features [14] and again on the gradual ferromagnetic-to-AF transition, this time using magnetoresistance [15]. Only at this stage did Roy and co-workers start to study

annealed UCu_2Ge_2 samples, but they still reported a ferromagnetic-to-AF transition [16, 17], a metastable magnetic response [16] and magnetization relaxation [17], with almost no reference to ferromagnetic domain effects and, as before, with no mention of NRCN work on UCu_2Ge_2 ferromagnetism [5]. Roy reacted to the NRCN Note [13] and published 'Comments on "Note on the magnetism of UCu_2Ge_2"' [18], standing firmly behind the appearance of an AF phase at LT. The NRCN 'Countercomments to Roy's Comments' followed [19], adhering to the NRCN observation of there being only ferromagnetic ordering below T_C in UCu_2Ge_2.

As Roy and his co-workers started to study annealed samples of UCu_2Ge_2 and became aware of all the work indicating there being only ferromagnetic ordering of such samples [5–11], they reported on two investigations, which were known to the present author but not mentioned in the 'Commented review' [2]. In the article 'Magnetic irreversibility and a marginal phase transition in UCu_2Ge_2' Roy and co-workers [20] admitted that 'the LT ferromagnetic-to-AF transition is quite marginal and depends crucially on heat treatment', that ferromagnetic domains were responsible for the decrease in magnetization of ferromagnetic UCu_2Ge_2 at LT and that as-cast and annealed samples of UCu_2Ge_2 have different magnetic features. In their comment [1], Roy *et al* repeat their conclusion that 'non-stoichiometry cannot be the source of the qualitative difference in the LT magnetic properties between as-cast and annealed UCu_2Ge_2'. They are probably referring to the nominal stoichiometry, but what really affects the magnetic properties is the extent of the final (actual) stoichiometry following the heat treatment, i.e. the annealing process. A subsequent publication on 'Magnetoresistance in a well annealed sample of UCu_2Ge_2' [21] confirmed the difference in features of as-cast and annealed samples, but still hinted at the presence of AF interactions in annealed UCu_2Ge_2, 'otherwise characterized as a ferromagnet'.

In the preceding Comment [1], Roy *et al* testify that their interest in UCu_2Ge_2 was generated originally by the similarity of magnetic properties with Al-doped $CeFe_2$ alloys (which are not in fact alloys but rather pseudobinary $Ce(Fe,Al)_2$ solid solutions [12]). Actually, in the Comment, pertaining to UCu_2Ge_2 [1], 12 out of the 20 quoted references deal with non-UCu_2Ge_2 materials, mainly with pure $CeFe_2$ and $Ce(Fe,M)_2$ solid solutions (M = Al, Co, Ru). These latter materials are cubic and contain two kinds of magnetic atoms, the lanthanide (4f element) Ce and the 3d transition element Fe, on two different magnetic sites. The compound UCu_2Ge_2 crystallizes with the BCT $ThCr_2Si_2$-type structure, and contains only one magnetic atom, the actinide (5f element) U, on a single magnetic site. Any similarity between the two systems, in magnetic behaviour and in some of the magnetic properties, is therefore purely coincidental, and the comparison itself cannot lead to conclusions as regards the UCu_2Ge_2 magnetism.

The direct path that Roy and co-workers could follow in order to prove the existence of any LT AF phase in their annealed (stoichiometric or non-stoichiometric) UCu_2Ge_2 samples is to study them by means of neutron diffraction. Roy and co-workers might simply hand their UCu_2Ge_2 sample(s) to their compatriot scientists at BARC (or perhaps other colleagues elsewhere) in order to have the neutron diffraction study done. Only such straightforward measurement would seem to justify a Comment on the 'Commented review' [2].

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