Reply to "Magnetic behavior of $Cr_{74}Fe_{26}$ alloy investigated by Mössbauer spectroscopy"

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This paper comments on the preceding paper by C. Bansal, T. Kumaran, S. J. Campbell, and G. L. Whittle [Phys. Rev. B **44**, 7111 (1991)].

The Mössbauer results described in the preceding paper by Bansal *et al.*[1] represent a significant step toward understanding the unusual magnetic behavior of Cr-Fe alloys with less than 0.3 atomic fraction of Fe. Previously it has been widely accepted that, at Fe contents above a critical composition near 0.2 Fe, these bcc solid solutions are ferromagnetic. The recent work[1] furnishes clear evidence that the changeover of the Mössbauer spectrum on cooling of an alloy with 0.26 Fe from the paramagnetic type to the ferromagnetic type starts near 172 K, but that it is not complete even at 4.2 K (note in Fig. 1 of Ref. 1 that lines 1 and 6 of the 4.2 K spectrum are very weak in comparison with lines 3 and 4). This means that the change in the Mössbauer spectrum extends over a temperature range representing more than 97% of what has been sometimes referred to as the Curie temperature. Clearly, this is not a normal paramagnet-to-ferromagnet transition. It was already found some years ago by Burke and Rainford in an excellent small-angle neutron-scattering (SANS) study[2] that the temperature dependence of the susceptibility of a "ferromagnetic" Cr-Fe alloy with 0.195 Fe is very similar to that of a spin-glass alloy with 0.175 Fe. They concluded, from their results, that the dominant magnetic response of both alloys probably arises from finite magnetic clusters. Indeed, in the light of the results of the recent Mössbauer study,[1] it now appears very likely that both of those alloys, as well as the one with 0.26 Fe are indeed superparamagnetic at around 200 or 300 K and that, on cooling over a wide temperature range, they gradually develop spin-glass-like properties as the interaction between their cluster moments becomes more predominant at lower temperatures. The magnetic structures in this region of the Cr-Fe alloy system appear to be quite similar to those in the 0.15–0.25 Fe region of the Au-Fe system[3] and in the amorphous $(Cr_{0.35}Fe_{0.65})_{75}P_{15}C_{10}$ alloy.[4]

As briefly reviewed recently[5] in the answer to the comments in Ref. 4, a characteristic feature of the magnetic behavior of these alloys is that, at a certain temperature, the lines in the Mössbauer spectrum begin to broaden. Near this temperature a variety of "critical phenomena" appear which normally signify the onset of ferromagnetism. Hence, it is sometimes designated the Curie temperature, T_c. Although it does correspond to the onset of a hyperfine field, it does , in fact, not represent a Curie temperature in the usual sense of a transition to long-range ferromagnetism. This is shown by the very small initial value of the line broadening (and of the hyperfine field) just below T_c and by its very gradual increase over a wide temperature range. Although the critical peak of the susceptibility versus temperature observed by Sarkissian[6] in reentrant Au-Fe alloys at very low fields is quite sharp, the magnetic specific heat versus temperature[7] has no corresponding sharp peak.[8] A critical peak of the small-angle neutron scattering was reported by Boumazouza *et al.*[4] at the temperature of the onset of the hyperfine field in the amorphous Cr-Fe alloy mentioned above and such peaks were observed by Burke and Rainford for crystalline Cr-Fe alloys with 0.208–0.25 Fe (Fig. 6 of Ref. 2). All these critical effects strongly suggest the start of a cooperative transition normally associated with the Curie point of a ferromagnet. Yet the above-mentioned features of the Mössbauer spectra of $Cr_{74}Fe_{26}$ and the absence of sharp peaks in the magnetic specific heat for the reentrant Au-Fe alloys show that, in the alloys discussed, no phase transition into the long-range ferromagnetic state actually takes place at the so-called Curie temperature, T_C. It seems very likely that the observed changes in the Mössbauer spectra extending over wide ranges of temperature (see, for instance, Fig. 1 in Ref. 1) result from the gradual decrease of the frequency of the superparamagnetic fluctuations with decreasing temperature, as described by Rancourt and Daniels[9] following the calculations by Blume and Tjon.[10] At sufficiently low temperatures the slowing of the fluctuations can lead to Mössbauer spectra, which would be difficult to distinguish from the spectra for long-range ferromagnets. If the above interpretation in terms of superparamagnetic fluctuations is correct for $Cr_{74}Fe_{26}$, we may expect further changes below 4.2 K, as its Mössbauer-effect (ME) spectrum approaches even more closely that of a static long-range ferromagnet due to further slowing down of the superparamagnetic fluctuations.

The question arises why the usual transition to ferromagnetism here does not proceed to completion at the temperature where the various critical phenomena are observed. It seems to me reasonable to assume that the cooperative increase of the spin-correlation length in the superparamagnetic alloys considered becomes arrested very quickly after it starts due to the limitation of the spin-correlation length imposed by the small size of the regions capable of becoming ferromagnetic. In the so-called reentrant Au-Fe alloys, these regions are most likely the Fe-rich "platelets" of Guinier-Presten zones, which were shown by Dartyge, Bouchiat, and Monod by means of x-ray diffuse scattering[11] to be roughly two atomic layers thick and extending parallel to $(1\frac{1}{2}0)$-type planes to distances of approximately 30 Å. These platelets are surrounded and separated from one another by a gold-rich matrix (with the Fe-content estimated to be approximately 2 at. %, or less, for $Au_{82}Fe_{18}$), which is not expected to become ferromagnetic even at 4.2 K. Consequently, the superparamagnetic fluctuations may continue to temperatures well below "T_c".

It is to be hoped that sufficiently detailed Mössbauer studies will soon be made of many more so-called "reentrant" type of spin-glass systems so that one can find out how many of them are superparamagnetic, like $Cr_{74}Fe_{26}$, i.e., *without* the usually assumed two magnetic phase transitions into and out of a postulated long-range ferromagnetic phase. Recent SANS studies by Rhine and associates[12] on reentrant spin-glass alloys indicated the absence of a ferromagnetic phase below the "Curie temperatures" of these alloys.

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