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

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This paper contains a reply to Bansal *et al.* [first paper of this series, Phys. Rev. B **44**, 7111 (1991)].

In their comment, Bansal *et al.*[1] report on Mössbauer studies of the reentrant crystalline alloy $Cr_{74}Fe_{26}$ in order to disprove the existence of a real ferromagnetic phase transition at $T_0 = 180$ K in this alloy, and so cast doubt on the usual interpretation of the magnetic behavior of alloy systems in the reentrant region (for example, see Ref. 2). They argue that their interpretation is similar to that of Beck and co-workers[3,4] for the $Au_{82.5}Fe_{17.5}$ alloy. On the other hand, we have previously presented evidence[5] that, in the case of the metallic glass $(Fe_xCr_{1-x})_{75}P_{15}C_{10}$, three different types of experiments with widely different intrinsic time scales [bulk ac susceptibility, Mössbauer effect (ME) spectroscopy, and small-angle neutron scattering (SANS)] yield the same definition of T_c, and so indicating that a true phase transition takes place at T_c.

Recently, Mangin *et al.*[6] have discussed the SANS results of the different alloy systems showing reentrant behavior in great detail, and have presented results for the CrFe amorphous glass. They conclude that the SANS results prove the existence of a true phase transition at T_c, but that the ferromagnetic (FM) phase shows nonstandard behavior. This nonstandard behavior (which presumably is typical for reentrant systems) is evident in the low-field susceptibility, the spin-wave properties, and in SANS results. This last technique measures the instantaneous spin correlations and so is sensitive to the presence of magnetic inhomogeneities. Their results show that, in addition to critical scattering dominant at large transferred momentum q, there is a subcritical scattering at low q which increases with decreasing temperature below T_c. They propose a model phase diagram from susceptibility and SANS results with transitions from paramagnetic to a spin-glass phase at low, and to a ferromagnetic phase at larger Fe concentrations, both consistent with the known Mössbauer results. In the ferromagnetic region, there is a reentrant transition region with a different low-temperature state. Since they propose several possible low-temperature transitions, the correspondence to the Gabay-Toulouse transition[7] has not yet been fully established, and in-field Mössbauer results are lacking on equivalent samples in this region. They also compare their results to those for the CrFe crystalline system. This work will be restricted to a discussion of the Mössbauer results and analysis as presented by Bansal *et al.* with respect to the transition at T_c, and our understanding of the FM and reentrant phase transitions.

These results for the Cr-Fe glasses[5,6] are fully consistent with the results presented by Kaul[8] for the transition from paramagnetism (PM) to ferromagnetism in inhomogeneous systems with competing magnetic exchange interactions (frozen-in disorder). He shows that, in a wide range of FM metallic glasses including Cr-Fe glasses, a true critical region at T_c can be found. Kaul has also presented a phenomenological model for these inhomogeneous systems outside the true critical region, trying to account both for the temperature-induced spin disorder, and that due to spatial inhomogeneities, both topological and chemical. He concludes that chemical disorder, present in crystalline alloys as well, is more important than the topological inhomogeneities present in amorphous and glassy systems in determining the effective exponents outside the true critical region. His model for the inhomogeneous FM allows finite clusters of spins which are canted with respect to the direction of the spontaneous magnetization M_s. The spatial extent of these clusters is temperature dependent (see Fig. 20 of Ref. 8).

On the other hand, Sherrington and Kirkpatrick[9] (SK) have presented a model of reentrant FM spin-glass systems. This was extended to vector spins by Gabay and Toulouse (GT).[7] In the GT phase diagram, there is a second transition at a freezing temperature $T_f < T_c$ to a canted FM spin-glass-like phase. The upper FM phase is collinear. However, in this FM phase there is a strong reduction in the thermal expectation value of the average local magnetic moment. This reduction can be thought of as being due to the fact that the moments are instantaneously canted, but in this phase, they are precessing around the common direction of M_s so that only the projection along M_s can contribute both to M_s and to the local moment (as measured, for example, by the Mössbauer hyperfine field). The fact that, in such systems, the hyperfine field is unusually small in the FM phase and regains its normal value in the low-temperature spin-glass-like state (where this precession stops and the spins are then macroscopically noncollinear), can be taken as evidence for this model.[2] The main question is whether or not disorder destroys the transition at T_c, so that the transition at T_f is not a reentrant transition at all. The results of Kaul[8] clearly show that this is not the case for almost all of the metallic glasses: the transition at T_c is indeed a true transition. This does not mean, however, that all magnetic moments must participate in the phase transition at T_c. This is most clearly evident in insulating systems with short-ranged interactions: Even at concentrations above the percolation threshold, finite clusters are present, and these will be seen as "free" spins. In ME spectra, this is evidenced by a nonzero intensity at $B_{hf}=0$: $P(B_{hf}=0)>0$. The role of these finite clusters and free spins in determining the second transition at T_f is controversial, but several years ago the author pointed out[10] that such free spins are not present in the canonical reentrant Au-Fe system.

The system studied by Bansal *et al.*,[1] $Cr_{74}Fe_{26}$, is somewhat more complicated than the cases discussed above. Preliminary SANS results presented by the group of Shapiro[11] indicated that there were spin waves detectable below T_c, but that these disappeared as the transition to the spin-glass-like phase was approached. Later results from this group[12,13] show that this is not the case, and that spin waves could be detected down to about 13 K. They concluded that "the most natural extension of the data favors, but does not prove, the existence of spin waves at the lowest temperatures. . . ." The complete concentration range was investigated by Burke *et al.*[14–16] The magnetic phase diagram[15] shows regions of itinerant antiferromagnetic order (spin-density wave) for low, FM order for high, and spin-glass order for low Fe concentration. The FM order near the transition to the spin-glass regime shows "complex magnetic behavior"[15] which is now interpreted as a reentrant transition to a spin-glass-like order discussed below.

There have been several ME studies of the $Cr_{1-x}Fe_x$ system. Both the results of Dubiel *et al.*[17,18] and of Tsuge *et al.*[19] are consistent with the reentrant magnetic phase diagram as presented by Burke *et al.*[15] In the region of $x=0.26$ (Bansal's sample), there is a reentrant transition from FM to a spin-glass-like state (it is not known if this is a canted FM or pure spin glass) at about 30 K, and so well above the lowest temperatures for which spin waves could be detected.[13] The above ME results show an increase in the slope of the average hyperfine field $\langle B_{hf}(T)\rangle$ below T_f. There are some differences between the Dubiel *et al.* results and those of Tsuge *et al.*, indicating, in addition, some problems with the metallurgy in this system.

We now turn to the evidence presented by Bansal *et al.*[1] on the temperature dependence of the hyperfine fields found in their spectra for $Cr_{74}Fe_{26}$. Instead of reporting the average $\langle B_{hf}(T)\rangle$, they choose to report the thermal behavior of the secondary peaks found in the distribution function $P(B_{hf})$. They have calculated this distribution using the Window[20] method, a method which leads invariably to spurious secondary maxima in the calculated $P(B_{hf})$. It is known, however, that the Window method yields reliable values of the average hyperfine field and $\langle B_{hf}(T)\rangle$ does show the characteristic features of the canting transition in this system,[18,19] a point not discussed by Bansal *et al.* They have not tried the more reliable Hesse-Rübartsch[21] method as used previously.[19] Thus, the authors' main arguments are based on features of the calculated $P(B_{hf})$, which cannot be separated from spurious artifacts. They also do not compare their results to the known ME or SANS literature for the reentrant behavior in this system.

The main misunderstanding of this Comment has to do with what is to be expected for a magnetic transition in inhomogeneous systems in the presence of competing exchange interactions. The canting picture presented above accounts for the general trends and properties of these systems better than a description in terms of supermagnetism (Bansal *et al.*) or of canted finite clusters in an otherwise collinear matrix (Kaul). The SK model assumes infinite-ranged forces, so that there is no percolation threshold as found in many reentrant systems (see Ref. 8). But the model does account for the spin-glass phase where the competition between the exchange interactions dominate, and ferromagnetism when this is not the case. Between these two limits, there is a range of reentrant FM with a lower transition from FM to a canted FM spin-glass-like state. In the reentrant region, the FM phase is collinear (moments all parallel within one magnetic domain) but is characterized by nonstandard dynamical behavior and an unexpectedly small spontaneous magnetization (reflected in a small hyperfine magnetic field). These small moments are a reflection of the fact that there is competition in the magnetic exchange interactions. At a lower temperature, these competing interactions lead to a noncollinear state in which the moments regain their full value, but are now no longer all parallel. The analysis of the Mössbauer hyperfine results in terms of secondary maxima of the distribution function cannot show whether or not there is a true phase transition in these inhomogeneous systems. The $Cr_{1-x}Fe_x$ system is also not a good choice to test models of the inhomogeneous magnetic states in local moment systems because of the itinerant nature of this alloy system. Despite this, several Mössbauer studies already published do show the characteristic features of reentrant magnetism, as was found in $Au_{1-x}Fe_x$, in $(Ni_{1-x}Fe_x)$ metallic glasses, and diluted ferrites,[2] and in $(Cr_{1-x}Fe_x)$ metallic glasses.[14–19]

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