Comments Concerning "Metastable Phase Separation in Au-Fe Alloys"

A recent Letter by Violet and Borg[1] concerns (i) the nature of the short-range order in AuFe and (ii) the interpretation of the Mössbauer data in this system. We feel that both subjects are presented in a very unsatisfactory way which leads to confusion. Furthermore this presentation ignores recent work.[2,3]

The main source of confusion is the idea that one could represent the AuFe system in the presence of short-range order as a *strict* two-phase system in which the Fe atoms either belong or do not belong to a dilute or concentrated phase. The x-ray evidence[4] is that there exists a tendency for Fe to precipitate into 30×30-Å^2 platelets oriented along [420] planes and about two layers thick. This admittedly crude image has only a statistical value and should not be naively taken as gospel truth.[5] We note[6] that *even in this picture* because of the smallness of the Fe-rich platelets and their numerous mutual crossings (there exist twelve equivalent crystallographic planes) it is hazardous to ascribe a single value of the hyperfine field to the "Fe-rich phase," as the probability for a given atom to be on the border, or at a crossing point of two planes, is quite sizable. Furthermore, in the analysis attempted by Violet and Borg it is far from clear if, according to the hyperfine field value of 330 ± 8 kG found (between 16.8% and 33% Fe), one would expect to find α-Fe (bcc phase) in all the corresponding alloys or *only* in the 33% alloy. In the latter case one would then have to admit that the Fe-rich phase is still fcc (and coherent with the Au lattice) but happens to have the same hyperfine field as the bcc α-Fe phase. The former hypothesis is, on the other hand, not compatible with the x-ray evidence.[4]

The authors state that their Mössbauer spectra are "well described" by two superimposed six-line Fe spectra. Window,[7] in very thorough work on the identical alloy series where he studied both low- and high-temperature spectra, concluded that all parameters (hyperfine field, quadrupole effect, and isomer shift) are correlated and distributed. He could not fit his spectra with two six-line patterns only. This would appear to rule out the simple analysis given by Violet and Borg. Window interpreted his results very convincingly in terms of different local Fe environments within each alloy.

Mössbauer measurements[7-11] show unequivocally that at each concentration the hyperfine fields on all the Fe sites go to zero together at one temperature (with at worst a spread of a few degrees), and this transition temperature varies strongly with Fe concentration. This is clear evidence that the alloys are magnetically homogeneous (although chemically less so): To be more specific, this means that the *magnetic correlation length* is *larger* than the *short-range-order range.* Violet and Borg also state that "the model may account for . . . the double magnetic transition." The transition referred to is the transition from canted to fully aligned Fe spin polarization with increasing temperature in 16% and 18% Fe alloys.[10,11] We fail to understand how a two-phase system could possibly reproduce this behavior.

Finally the statement that "[our model] may also apply to *Cu*Mn spin glasses where short-range order analogous to that of *Au*Fe is observed" is further adding to the confusion because the key point of the neutron work on *Cu*Mn,[12] and of the x-ray work on *Ag*Mn,[13] is that the short-range order present in both these systems is towards *anticlustering* and so is just opposite to that of AuFe.

In conclusion, chemical ordering effects in *Au*Fe alloys are certainly important, but we suggest that the oversimplified two-phase model presented by Violet and Borg is both erroneous and misleading.

P. Monod and I. A. Campbell
Physique des Solides
Université de Paris-Sud
F-91405 Orsay, France

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