Comment on "Magnetism in $Au_{82.5}Fe_{17.5}$"

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Low-field magnetization in parallel and perpendicular geometries, Mössbauer spectroscopy, and small-angle neutron scattering data, carried out on the same samples of amorphous reentrant spin-glass alloys $(Fe_xCr_{1-x})_{75}P_{15}C_{10}$ suggest a true phase transition with cooperative phenomena at T_c and not a simple deviation from the Langevin paramagnetism as suggested by Rakers and Beck [Phys. Rev. B **36**, 8622 (1987)] in $Au_{82.5}Fe_{17.5}$ alloy. A departure from conventional ferromagnetism is found in the small size of the domains, which could be some thousands of angstroms.

The term reentrant spin glass (RSG) denotes a magnetic phase which is observed at low temperature in random alloys which show a paramagnetic-ferromagnetic transition at a higher-temperature T_c and whose composition is close to the critical concentration for the onset of ferromagnetism. RSG behavior usually takes place in alloys in which both ferromagnetic and antiferromagnetic exchanges are present in proportion such that ferromagnetic interactions weakly dominate.[1]

Some of the classical systems known to exhibit such properties are crystalline Fe_xAu_{1-x} (Ref. 2) or Fe_xCr_{1-x} (Refs. 3 and 4) or amorphous (Fe_xNi_{1-x})PBA1 (Refs. 5 and 6), Fe_xSn_{1-x} (Ref. 7), (Fe_xCr_{1-x})PC,[8] and $(Fe_x\text{-}Mn_{1-x})$PBA1.[9,10] Mean-field calculations have likewise shown the occurrence of RSG phenomena, and predicted phase diagrams have been published by several authors.[11–13] The true nature of the RSG phase is still much debated. It is not obvious and differs perhaps from one system to another.

More surprising, the nature of the ferromagnetic phase at higher temperature and the occurrence of a real paramagnetic-ferromagnetic transition at T_c are still questioned. In a recent paper, Rakers and Beck[14] claim there is no indication for an intermediate ferromagnetic phase in a $Au_{82.5}Fe_{17.5}$ alloy which was considered[2] as a reentrant spin glass with a Curie temperature of about 140 K. Their conclusion is supported by two observations.

(1) The upper "knee" of the plateau in the susceptibility is well defined when the sample is perpendicular to the external field H but vanishes when the sample is parallel to this field. This is not an intrinsic characteristic of the alloy but is a consequence of the intrinsic susceptibility χ_i being larger than $1/N$ (N is the demagnetizing factor).

(2) The temperature T_c of the upper knee found in the perpendicular geometry differs significantly from the temperature T_m at which Mössbauer spectroscopy exhibits the onset of hyperfine splitting.

Rakers and Beck conclude there is no indication of any intermediate ferromagnetic state or even of any magnetic phase transition, but only a gradually increasing deviation from Langevin paramagnetism with a large increase of χ_i. Although all RSG alloys do not necessarily exhibit all the same properties, we present in this comment a counter example to Rakers and Beck of a RSG system $(Fe_x\text{-}Cr_{1-x})_{75}P_{15}C_{10}$ in which there is no scattering of the transition temperatures, and for which the arguments developed by the above authors are invalid.

We carried out low-field magnetization in parallel and perpendicular geometries, Mössbauer spectroscopy and small-angle neutron scattering (SANS) for several compositions x above and near the critical concentration $x \approx 0.60$. We found that the upper knee of the demagnetization plateau and the onset of the hyperfine field do occur at *the same* temperatures, even when the composition of the sample is close to that of the multicritical point. In addition, we show this temperature is that at which a peak of critical scattering occurs in SANS.

Figures 1 and 2 show the low-field magnetization with the external field parallel and perpendicular to the sample. As reported by Rakers and Beck,[14] for the $Au_{82.5}Fe_{17.5}$ alloy, a magnetization plateau with lower and upper knees is observed in perpendicular geometry (more precisely when H/N is small). In the parallel geometry (H/N large) no singularity is observed at the temperature of the upper knee observed in the perpendicular geometry. At this stage, we agree with Rakers and Beck that the question of whether or not the upper knee does correspond to a magnetic transition must be confirmed by other considerations.

Figure 3 shows very clear data obtained by Mössbauer spectroscopy and SANS. Above T_c, the Mössbauer spectrum shows a doublet due to the pure quadrupolar effect

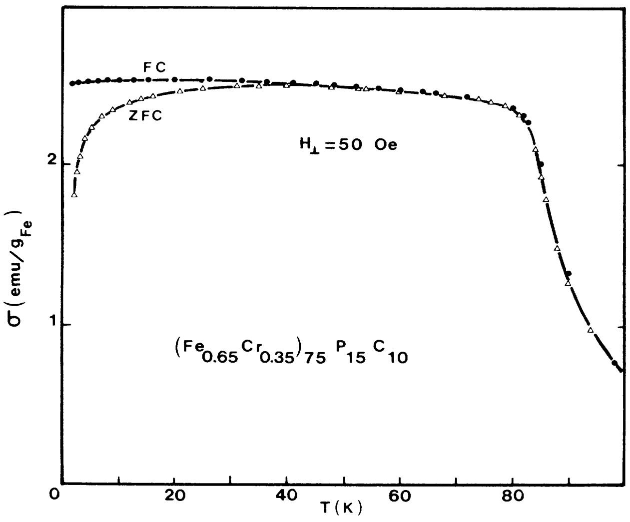

FIG. 1. Temperature dependence of the field-cooled and zero-field-cooled magnetizations measured in a 50-Oe field perpendicular to the plane of the ribbons.

in the paramagnetic phase, which is independent of the temperature. When the temperature of the sample is lowered below 84 K, the width of the two components of the quadrupole doublet increases sharply. It is obvious that this effect is not due to a change in the quadrupolar splitting but originates in the onset of the hyperfine field. The SANS measured on the same sample for scattering vector $q = 0.04$ Å^{-1} exhibits a very sharp peak at the same temperature [Fig. 3(a)]. This peak is the consequence of the near divergence of the correlation length at $T_c = 84$ K. A careful analysis of the q dependence of the intensity[8] shows that above T_c the correlation function follows an Ornstein-Zernike law (Lorentzian line shape),

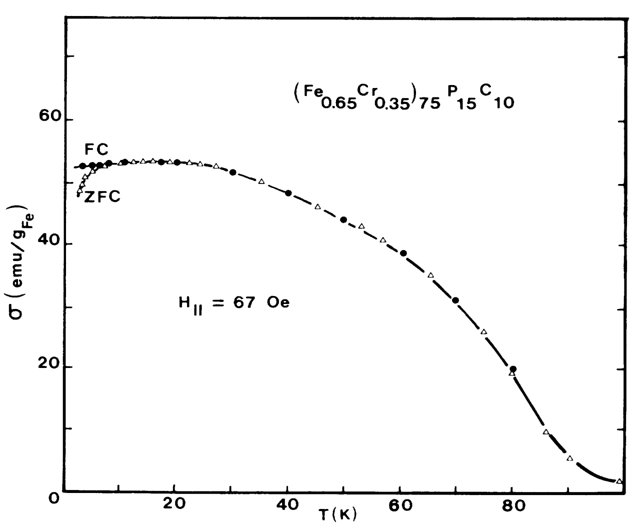

FIG. 2. Temperature dependence of the field-cooled and zero-field-cooled magnetizations measured in a 67-Oe field parallel to the plane of the ribbons.

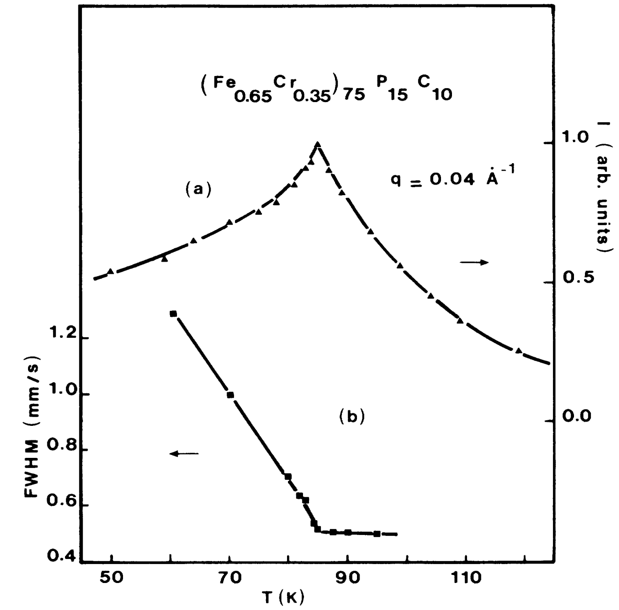

FIG. 3. (a) Temperature dependence of the small angle neutron scattering intensity at $q = 0.04$ Å^{-1} and (b) of the full width at half maximum (FWHM) of the components of the absorption doublet obtained by transmission Mössbauer spectroscopy.

but below T_c, the sum of two Lorentzians reflecting transverse and longitudinal fluctuations have to be introduced.

The correlation length of the transverse Lorentzians remains infinite (within the instrumental resolution) while the longitudinal correlation length becomes finite and decreases with the temperature,[15] as expected from mean-field theories.[16] The occurrence of these two contributions below T_c shows the presence of the symmetry breaking at the paramagnetic-ferromagnetic phase transition (at least at the length scale of the instrumental resolution $\simeq 500$ Å).

The identical temperatures found (1) for the upper knee of the demagnetization plateau, (2) the onset of the hyperfine splitting, and (3) of the critical divergence in SANS at $T_c = 84$ K strongly suggests a true phase transition with cooperative phenomena at T_c, and not a simple deviation from the Langevin paramagnetism. The same behavior has been observed on the $(Fe_{0.7}Cr_{0.3})_{75}P_{15}C_{10}$ alloys where $T_c = 134$ K. As T_c is higher, the result is less significant but in addition to the above measurement, we could measure spin waves in the ferromagnetic phase,[8] with negligible anomalous lifetime broadening. Similar measurements carried out in $(Fe_{0.62}Cr_{0.38})_{75}P_{15}C_{10}$ also gave identical temperatures for T_c from both magnetization and Mössbauer-spectroscopy techniques.

In these alloys one departure from conventional ferromagnetism is found in the size of the domains. As shown previously in $(Fe_{0.65}Cr_{0.35})_{75}P_{15}C_{10}$, the size of the domains could be some thousands of angstroms,[17] at least one order-of-magnitude smaller than "conventional" ferromagnets. However, this has little relationship to the magnetic "viscosity" described by Rakers and Beck.[14]

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