Comment on "Ferromagnetism and spin-glass properties of *Pd*FeMn"

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A critique on the criterion used and a different interpretation are given for the low-field magnetization measurements of Carnegie and Claus on *Pd*FeMn. Additional evidence is presented to connect these experiments with the typical behavior of concentrated spin glasses and not that of a normal ferromagnet.

Through low-field magnetization measurements on a *Pd*FeMn sample Carnegie and Claus (CC)[1] have observed the sudden onset of hysteresis effects at a temperature $\approx 0.3 T_C$. By taking the maximum slope of a M vs H hysteresis loop, they arrive at a criterion for ferromagnetism and thus claim the alloy shows the normal behavior of a hysteretic ferromagnet. In a previous paper[2] we presented a susceptibility study for a series of $(Pd\mathrm{Fe})_{1-x}\mathrm{Mn}_x$ alloys which exhibited distinctly different magnetic properties depending upon the Mn concentration x. (The Fe concentration was kept constant at 0.35 at.% Fe). For low Mn concentrations $x < 0.03$, ferromagnetic behavior typical of a giant moment system[3] was observed, while at $x > 0.07$ the susceptibiliy was characteristic of a spin glass.[2,4] Between these two concentrations a plateau-like susceptibility occurred as a function of temperature. The high-temperature knee was indicative of ferromagnetism with a corresponding T_C, and the low-temperature drop in χ was taken as evidence for the onset of a spin-glass-like phase, i.e., the random freezing of large ferromagnetic clusters. This interpretation was consistent with the field dependence of $\chi(T)$,[5,6] the phase diagram for *Pd*FeMn and *Pd*Mn (see Fig. 1) and the good qualitative agreement[2,6] with the spin-glass theory of Sherrington and Kirkpatrick.[7] Lacking microscopic measurements on these systems, we proposed the following physical picture.[8] At low concentrations an infinite cluster of ferromagnetically coupled giant moments leads to a percolation type of inhomogeneous or nonuniform ferromagnetism. However, upon the inclusion of a sufficient amount of Mn and the corresponding nearest-neighbor antiferromagnetism[9] the infinite ferromagnetic cluster is broken at its weakest links and a series of randomly orientated ferromagnetic domains or clusters result. At large Mn concentrations only the spin-glass freezing, illustrated by a small peak in $\chi(T)$, appears. This general type of behavior is typical for a large number of alloy systems with mixed ferro- and antiferromagnetic interactions and has led us to propose a uniform magnetic phase diagram for these types of magnetic alloys.[10]

The measurements of CC are in agreement with our susceptibility results in that they characterize two distinct temperatures, $T_C = 9$ K and a lower temperature $= 3$ K which we called T_f. Yet there are two major points of departure dealing mainly with interpretation. First, the field cooling magnetization curve remains at a constant value below T_f. This, however, is a typical property of a field-cooled spin glass[11,12] resulting from its thermal remanent magnetization. Second, and more subtle is the interpretation of ferromagnetism derived from the full hysteresis loop. It should be noted here that hysteresis loops are a common property of the spin glasses[11,13] for which a ferromagnetic-like behavior may be field induced. An important difference with a normal ferromagnet is the long time relaxation or metastability of the spin glass. In fact, we do observe a long time decay of the remanent magnetization for this *Pd*FeMn alloy. No data are given for the hysteresis

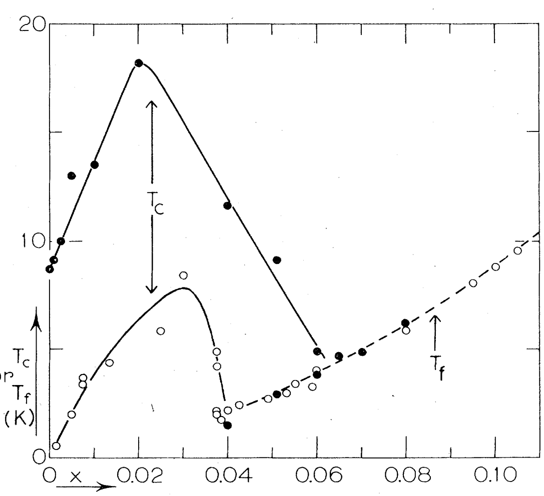

FIG. 1. Collection of experimental data for the magnetic phase diagrams of $(\mathrm{Pd}\ 0.9965\ \mathrm{Fe}\ 0.0035)_{1-x}\mathrm{Mn}_x$ (●) and $\mathrm{Pd}_{1-x}\mathrm{Mn}_x$ (○).

loop of CC and only a schematic sketch is inserted which cannot represent *Pd*FeMn since these alloys do not saturate in fields up to 15 T.[14] By taking the maximum slope of the hysteresis loop, a susceptibility is defined which remains constant just below T_f. It is stated that this is a criterion for ferromagnetism which has been applied to CuNi and VFe (see CC Ref. 4). However, in these references it was the maximum slope of the virgin curve which was used to obtain the susceptibility. For *Au*Fe these two are certainly not the same.[15] Such criteria for ferromagnetism are arbitrary since they are presented without any physical justification or model. When the virgin curve criterion is applied in the case of CuNi there is indeed a low-temperature fall off of this susceptibility[16] and furthermore the critical concentration thus obtained is significantly lower than that found by other workers.[17]

A complete discussion of the criteria used by Carnegie and Claus[1] is of minor importance here. Instead we wish to consider the underlying physics of the measurements. Certainly a hysteresis loop strongly opens up at 3 K, but what is the cause for this sudden onset of hysteresis which occurs in concentration between a nonhysteretic ferromagnet and a spin-glass phase? Hysteresis is usually associated with the formation of domains with various orientations, irreversible processes, thermal activation etc. Yet all of these descriptive terms are equally well applicable to a spin glass, mictomagnetic, or a collection of small ferromagnetic particles. The use of an external field, especially near the critical concentration of a random alloy, does not permit an unambiguous distinction. For, a ferromagnetic infinite cluster could easily be reformed with the slightest applied field. The spin-glass-like phase in the Pd-based systems is most probably a collection of ferrimagnetic clusters which are weakly frozen into random orientations. This latter concept provides at least one way for describing the onset of hysteretic effects in *Pd*FeMn.

Irrespective of the interpretation or name given to the lowest temperature state of the alloys under discussion, there is the physical meaning of our phase diagram as shown in Fig. 1. This may be summarized as follows: (a) Over a certain composition region $x < 0.04$ for *Pd*FeMn$_x$, ferromagnetism is found *without* hysteresis effects. (b) In another concentration region $x > 0.065$ for *Pd*FeMn$_x$, no long-range ferromagnetic and clearly spin-glass-like ordering appears. (c) For the intermediate concentrations, a double transition (our interpretation) or hysteretic ferromagnetism (CC interpretation) is found. Here either our T_f or the activation energy, i.e., the temperature where the coercive force rapidly increases, extrapolates to the spin-glass freezing temperatures obtained in (b) $x > 0.065$. The description of the physics connected with this phase diagram is related to the long standing and controversial question of whether the paramagnetic to spin-glass transition should be considered as a phase transition and a critical phenomenon. If one accepts this point of view then the theory of Sherrington and Kirkpatrick[7] is applicable and our results can be interpreted as a double phase transition. On the other hand one may approach the spin-glass problem from the notion of Néel superparamagnetic clusters which are blocked by an anisotropy energy barrier. Here the concepts of domains, nonequilibrium properties, irreversible processes, long relaxation times, thermal activation, etc., are intrinsic to the model. In this case a better theory for interpreting the results is that of Harris and Zobin[18] which also predicts three different phases (paramagnetic → ferromagnetic → spin glass) as a function of the anisotropy.

Additional evidence for a different low-temperature phase comes from small-angle neutron scattering experiments. In *Pd*FeMn,[19] *Au*Fe,[20] and *Cr*Fe[21] a strong increase of the small-angle scattering intensity is observed at T_f. This effect is much less pronounced for alloys with concentrations in the "normal" spin glass or ferromagnetic regime. A complete interpretation of the results and the scattering mechanisms is not yet clear, however, one very appealing description is in terms of randomly frozen ferrimagnetic clusters. In this way a consistent connection with macroscopic measurements is obtained. Furthermore, very recent neutron scattering measurements on the insulating $Eu_xSr_{1-x}S$ system[22] which is claimed to be a spin-glass ferromagnet exhibit an anomalous broadening of the magnetic Bragg peaks at low temperatures. This result was interpreted as the breakdown of long-range ferromagnetic order with the onset of a low-temperature spin-glass-like phase for x just above the ferromagnetic critical concentration.

In conclusion, we feel there is still a lack of sufficient experimental information both on the microscopic as well as the macroscopic scale to definitively interpret the observed phenomena. Nevertheless, measurements on systems which are on the borderline between ferromagnetism and spin-glass ordering will certainly add to our understanding not only of the spin-glass problem, but also of the percolative ferromagnets.

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