Reentrant magnetism: New aspects

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Measurements are presented of the thermoremanent and the low-field magnetizations of an amorphous $(Fe_{78}Mn_{22})_{75}P_{16}B_6Al_3$ and a crystalline $Ni_{78.4}Mn_{21.6}$ reentrant alloy as a function of temperatures. The data show evidence for two different blocking mechanisms responsible for the sharp drop in the familiar zero-field-cooled susceptibility below T_f. Both mechanisms are basically connected with domain-rotation effects. Our results are reasonably explained on the basis of a phenomenological free-energy model.

A number of chemically disordered magnetic alloys exhibit conventional ferrromagneticlike behavior over a certain range of temperatures and spin-glass-like behavior at lower temperatures. The transition towards the low-temperature phase (referred to as reentrance) is marked by a sharp fall-off of the familiar zero-field-cooled (ZFC) susceptibility and a regime of strong irreversibilities. This transition is presently the object of many investigations and controversies.[1] In particular, it is not clear whether it is associated with a phase transition nor is it clear what exactly happens to the spontaneous magnetization m_s at the freezing temperature T_f. Moreover, even though the transition itself has frequently been discussed in terms of current spin-glass theories,[2-5] no attempt seems to have been made to understand the behavior of the magnetization as a function of H or T at lower temperatures. In this Brief Report we report new information on these points, deduced from systematic investigations of the magnetization of two representative reentrant [an amorphous $(Fe_{78}Mn_{22})_{75}P_{16}B_6Al_3$ and a crystalline $Ni_{78.4}Mn_{21.6}$] alloys. Their choice has been dictated by the fact that (1) they both have clearly separated and extended magnetic phases with very close transition temperatures ($T_f \simeq 40$ K, $T_C \simeq 270$ K) and (2) it seems that the spontaneous magnetization of $(Fe_{78}Mn_{22})_{75}P_{16}B_6Al_3$ and equivalent amorphous reentrant alloys collapses at T_f according to scaling laws,[3-5] whereas it stays essentially constant in the case of crystalline Ni-Mn alloy.[1,6]

Figure 1 shows the temperature variation of the external dc susceptibility of $Ni_{78.4}Mn_{21.6}$ for two different fields plotted after the sample had been prepared in various magnetic states. We first examine the familiar ZFC curve (bottom) commonly associated with the nonequilibrium susceptibility χ_n, the s-like shape of which is typical of many reentrant alloys. We want to emphasize here some of its characteristic features which do not seem to have received much attention in the past. At first, it is remarkable that below a certain threshold temperature $T^*(H)$, χ_n stays very low and nearly independent both of H and T (for weak H). We have examined this section of χ_n and found it fairly reversible upon successive cooling-heating operations, with no significant time effects on the scale of measuring times (10 min). Secondly, as T is further increased χ_n rises abruptly up to $T \simeq T_f$ and then levels off. A detailed examination of this part of χ_n ($T^* < T < T_f$) shows that it is not only characterized by the onset of strong irreversibilities and time effects, as it is known, but also by a marked dependence on the applied field. The later effect is evidenced by the dashed line below the solid curve in Fig. 1.

Consider now the first branch of the hysteresis loop (inset Fig. 1). We find that it qualitatively presents the same behavior as the ZFC curve discussed just above except that the variables T and H are exchanged. This analogy is certainly not fortuitous and could be explained by the presence of macroscopic anisotropy fields $H_a(T)$ created during cooling and having the same structure as the domain pattern in the ferromagnetic phase.[7] The idea is that as T or H is varied each magnetic domain would stay firmly stuck to the lattice along its initial direction (acquired in the ferromagnetic phase) as long as $H < H_a(T)$. It is then quite natural to assume that equation $H = H_a(T)$ defines a line in the (H,T) diagram which manifests itself either in m-vs-H or in m-vs-T curves acccording to whether it is crossed at T=const or at H=const, respectively. As in pure spin-glasses H_a is expected to decrease monotonically with increasing T, going to zero at T_f. This is also consistent with the behavior of the magnetic cycle (m-vs-H) of $Ni_{78.4}Mn_{21.6}$ (not shown here) as a function of T. In order to give more

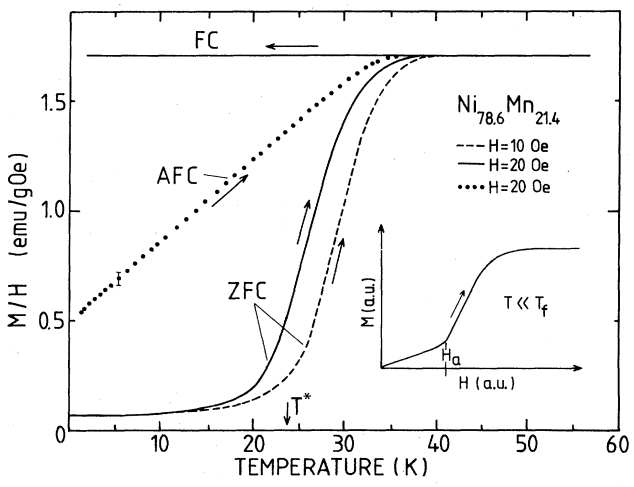

FIG. 1. dc susceptibility vs temperature for $Ni_{78.4}Mn_{21.6}$. Dashed and full lower curves traced in H=20 and 10 Oe, respectively, after cooling in H=0. Dotted line traced in 20 Oe after cooling in an alternating field [$|h(t)| \sim 2$ kOe] and demagnetizing at 1.5 K. Full upper curve shows field-cooled data at 20 Oe. In all cases, T was changed in steps of 1 K at an average rate of about 3 min. Inset symbolizes the first branch of the m-H loop (ZFC) for the two reentrant alloys considerd in this paper.

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experimental support to these analyses, we have measured the low-H susceptibility (dotted line in Fig. 1) after the sample had been cooled in an alternating field [$|h(t)|=2$ kOe] at a rate of ~ 10 cycles $K^{-1}min^{-1}$ down to 1.5 K. As shown previously[7] this method allows the supression of the alternating fields invoked just above. Clearly, the resulting anisotropy-field-cooled (AFC) curve (dotted) presents no singular behavior near T^*, varies roughly linearly with T almost up to T_f, and is nearer to the equilibrium "field-cooled" (FC) curve. It is to be stressed that this AFC curve differs from the ZFC curve in at least two important aspects: (i) it is less affected by macroscopic unidirectional anisotropy and (ii) the corresponding $m=0$ state should be more frustrated and would certainly have a very different domain pattern. In the absence of macroscopic anisotropy,[8] the drop in the AFC curve below T_f (Fig. 1) could be either due to a restraint on the rotation of the spontaneous magnetization m_s by some kind of exchange coupling with the spin-glass (SG) matrix, which coexists with the ferromagnetic (FM) phase;[5,6] or it could mean the destruction of the long-range FM order, i.e., the vanishing of the spontaneous magnetization[2-5] and a transition towards a pure SG-like state. Also shown in Fig. 1 is the so-called FC magnetization curve which is found to be rather independent of T and reversible if the sample is warmed back (from 1.5 K to T_f) in the same field H_c, immediately after cooling. It is worth noting that, contrary to the pure SG case, the FC magnetization considered here does not really correspond to true FC conditions since the field effectively applied during cooling is equal to zero as in the FM phase ($T > T_f$) for which $H_{\text{eff}}=H_c-Nm=0$ ($m < m_s$). This would suggest that the structure of the magnetic domains in the "FC" and the ZFC states are not fundamentally different in the low-H limit. Moreover, the fact that the "FC" magnetization stays constant during cooling strongly suggests that the domain structure and, as a consequence, the spontaneous magnetization, also stay unchanged during cooling.

We now consider the remanent magnetization m_r (Fig. 2) obtained just after the cooling field ($H_c=80$ Oe) had been removed at 1.5 K. The more striking feature of the temperature behavior of m_r is that it seems remarkably correlated with the ZFC magnetization curve (m) in the sense that their sum is nearly constant as a function of T and equal to the "FC" magnetization ($m+m_r \approx H_c/N$ for $H_c < Nm_s$). This relationship is found to hold regardless of the sample shape (needle- or spherelike; see inset Fig. 2). Similar results are shown in Fig. 3 for amorphous $(Fe_{78}Mn_{22})_{75}-P_{16}B_6Al_3$.

This is a further characteristic feature of the reentrant state and could be explained as follows. The effective field acting on m_r is now reduced to the demagnetizing field $H_d=-Nm_r(T)$, the role of which will depend on whether it is smaller or greater than the anisotropy field $H_a(T)$. Experimentally we find that at 2 K, $H_a \approx 300$ Oe (for the Ni-Mn alloy) whereas $H_d=Nm_r \approx H_c=80$ Oe so that $H_a \gg H_d$. As a result we expect that m_r will stay constant upon heating as long as $H_a(T) > H_d=80$ Oe.

The interpretation of our data can be made somewhat more quantitative on the basis of a simple magnetostatic free-energy model. Obviously, a rigorous treatment of the problem is out of the scope of this paper and we need the following approximations: (1) we will assume that the data can be described by a single demagnetization factor N, (2) we will use an Ising-like model, and (3) we will neglect magnetic hysteresis[9] (in the m-vs-H relationship). Therefore, as a first approximation we divide the spontaneous magnetization into two parts m^+ and m^-, respectively, parallel and antiparallel to H which is itself along the sample axis corresponding to the demagnetizing factor N.

Let $\theta^{\pm}=(\mathbf{m}^{\pm},\mathbf{H})$, the angle acquired by $m^{\pm}$ due to the variation of H at a fixed $T < T_f$. Then the simplest free energy we can imagine is

$$F=-K^+\cos\theta^+ + K^-\cos\theta^- - (m^+\cos\theta^+ + m^-\cos\theta^-)H + \tfrac{1}{2}N(m^+\cos\theta^+ + m^-\cos\theta^-)^2 \quad (1)$$

with

$$m=m^+\cos\theta^+ + m^-\cos\theta^- \ . \quad (2)$$

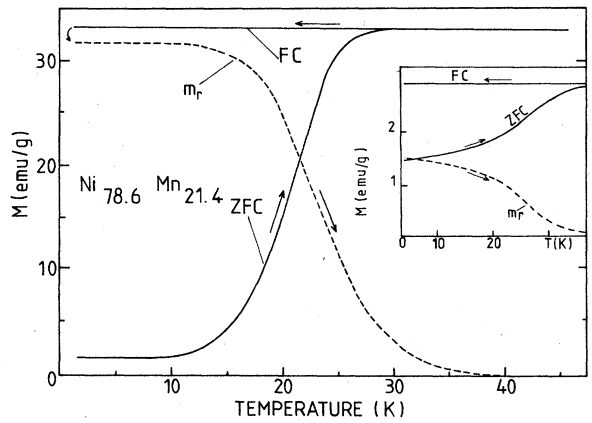

FIG. 2. Magnetization vs temperature curve of $Ni_{78.4}Mn_{21.6}$ for field values of the order of the demagnetizing field $H_d=Nm_s \sim 80$ Oe ($N \approx 0.25$, $m_s \simeq 320$ emu/cm^3, sample dimension $\approx 6\times1\times0.5$ mm^3) in the ferromagnetic phase. Full curve shows ZFC data in 80 Oe; dashed curve shows the thermoremanent magnetization (m_r) obtained after cooling in $H_c=80$ Oe and then removing the field at 1.5 K. Insert corresponds to a spherical sample. Here $H_d=1.3$ kOe, whereas the measuring field is equal to 80 Oe.

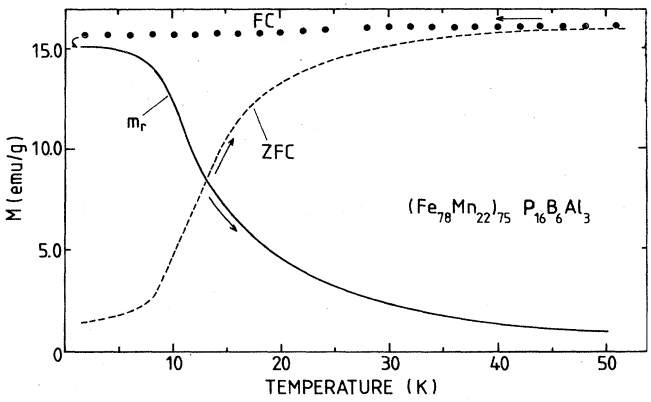

FIG. 3. M vs T curves for amorphous $(Fe_{78}Mn_{22})_{75}P_{16}B_6Al_3$ at $H=10$ Oe. Dashed curve shows ZFC data at 10 Oe, whereas full curve represents the thermoremanent magnetization (m_r) obtained after cooling in the same field (10 Oe) down to 1.5 K. Note that the measuring field (10 Oe) is much smaller here than the demagnetizing field ($H_d \sim Nm_s \approx 75$ Oe, $m_s \approx 560$ emu/cm^3 and $N=0.15$, sample dimension $\approx 6\times2\times0.08$ mm^3).

Here K^+ and K^- are related to H_a by equations

$$H_a = \frac{K^+}{m^+} = \frac{K^-}{m^-} - \frac{K}{m_s} \quad . \tag{3}$$

We note that Eq. (1) ignores any internal susceptibility term of the sort that could arise from isotropic exchange interaction with the SG matrix, for instance. The analysis of the data will prove that the role of such a possible term is negligible here. Minimizing the free energy [Eq. (1)] with respect to θ^+ and θ^-, we can easily calculate m for given values of H and T. The solutions depend on whether the sample had been cooled in a field H_c or not (i.e., $m^+ - m^- \neq 0$ or not). For the ZFC state ($m^+ = m^-$) the solutions are

$$m = 0, \quad H < H_a(T) \quad , \tag{4}$$

$$m = \frac{H - H_a(T)}{N}, \quad H > H_a(T) \quad , \tag{5}$$

$$m = m_s, \quad H > H_a(T) + N m_s \quad . \tag{6}$$

On account of the roughness of our model, we can consider that Eqs. (4) and (5) reproduce quite satisfactorily some of the most characteristic features of both the magnetic loop and the nonequilibrium susceptibilities shown in Figs. 1, 2, and 3 (lower lines): a very low value of m up to a certain field, respectively, temperature [given by $H_a(T) = H$], followed by a rapid rise of m in a manner which is governed to a large extent by N. The temperature dependence of the thermoremanent magnetization can be determined in the same way as above except that now the intial conditions are

$$M_r = m^+ - m^- \approx H_c / N \quad \text{and} \quad H = 0 \quad .$$

Indeed, we can easily check that the appropriate solutions are

$$m_r = m^+ - m^- \approx H_c / N, \quad T \lesssim T^* \quad , \tag{7}$$

$$m_r = \frac{H_a(T)}{N + \chi^{-1}}, \quad T \gtrsim T^* \quad , \tag{8}$$

with

$$H_a(T^*) = N(m^+ - m^-) \approx H_c \quad . \tag{9}$$

Once again the variation of m_r with T given by Eqs. (7), (8), and (9) above is in excellent agreement with the experimental behavior seen in Figs. 2 and 3. In all cases m_r is fairly independent of T up to a certain temperature and then decreases in a manner governed mainly by demagnetization. In addition, the close analogy with the ZFC curve is now justified since from the comparison of Eqs. (4), (5) and Eqs. (7), (8), we check that the sum $m + m_r$ is approximately independent of T and equal to H_c / N as found experimentally.

We have been able to show that the characteristic properties of crystalline as well as amorphous reentrant magnetism are essentially similar and result from an interplay between two constraints on the spontaneous magnetization. These are the demagnetizing field and the macroscopic anisotropy which appear in the reentrant phase. Moreover, we believe that the experimental techniques we used to separate these effects are interesting in their own right as far as reentrant magnetism is concerned.

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