Anisotropy in the Zero-Field-Cooled States of $Ni_{79}Mn_{21}$ and $Au_{81}Fe_{19}$: Irreversibility Effects

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The relationship m vs H for the alloys $Ni_{79}Mn_{21}$ and $Au_{81}Fe_{19}$ has been investigated at 1.5 and 4.2 K. Below a certain field H_a, it is linear, reversible, and governed by anisotropy fields even in the zero-field–cooled state. These fields are correlated with the spatial distribution of the spontaneous magnetization during cooling. A procedure allowing their suppression is presented. It leads to a radical change of the hysteresis loops without affecting significantly the degree of irreversibility.

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Despite the recent progress concerning the low-temperature magnetic properties of alloys in which coexist both spin-glass and ferromagnetic orders many of their aspects are still unclear. A typical example is that of the alloy $Ni_{79}Mn_{21}$, the magnetization of which presents some strange variations, as a function of the applied field, which are hardly understood on the basis of the present theories. According to the theoretical study of the infinite-range isotropic models,[1,2] the mixed spin-glass–ferromagnetic state would be characterized by strong irreversibilities which set in during cooling and which are connected with the freezing of the longitudinal magnetization. These predictions have been confirmed by experimental results.[3] However, recent numerical calculations seem to show that irreversibilities cannot exist in the spin-glass-like alloys, without the presence of some kind of anisotropic interactions.[4]

One of the purposes of this Letter is to demonstrate that at low enough temperature the relationship between the magnetization and the field (m vs H), for two very typical alloys $Au_{81}Fe_{19}$ and $Ni_{79}Mn_{21}$, is strikingly influenced by macroscopic anisotropy fields even in the zero-field-cooled (ZFC) state. However, these fields have no marked influence on the degree of irreversibility.

The working hypothesis is that during cooling through the de Almeida–Thouless transitional region each quasiferromagnetic domain freezes in a Weiss-like molecular field. The latter behaves like a strong cooling field having the direction that the associated magnetic domain had during cooling. The analogy with pure spin-glasses is that in this case the anisotropy constant at a given temperature is found to be the same whether the sample is cooled in zero field or in a cooling field.[5,6] The latter ensures the creation of a remanent magnetization, i.e., one which is analogous to a spontaneous magnetization.

In order to ensure a good homogenization, the alloys were melted three times and further homogenized by cold working. Each specimen was than annealed ~4 h at 900 °C and water quenched. The samples were then kept in liquid nitrogen until transferred into the cryostat.

The hysteresis loop (m vs H) of $Ni_{79}Mn_{21}$ cooled in zero field down to 1.5 K is given in Fig. 1(a). As reported by earlier investigators,[7] the curve has some very unusual features. First, it is remarkably linear and reversible up to a certain field $H_a \simeq 150$ G. Secondly, when the field is further increased a sudden rise of the magnetization

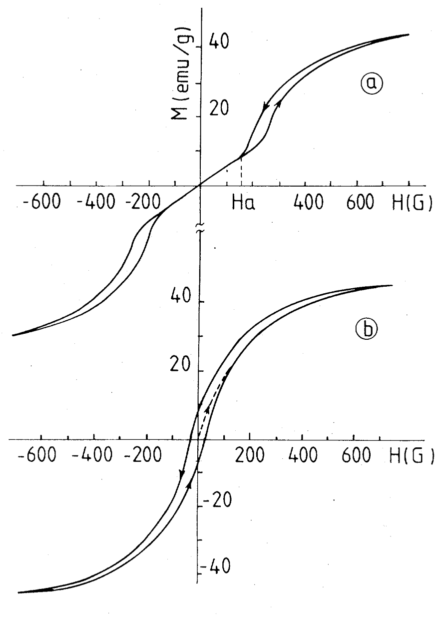

FIG. 1. Two complete hysteresis loops for $Ni_{79}Mn_{21}$ at 1.5 K: (a) after zero-field cooling; (b) after slow cooling in an alternating field (1 kG peak to peak) and demagnetizing the sample at 1.5 K. H_a represents a field value below which the ZFC curve is linear and reversible.

takes place. A third result (not reported in Ref. 7) concerns the appearance of irreversibilities above $H = H_a$. The first two effects just mentioned have been ascribed[7] to a field-induced spin-glass–ferromagnetic transition. However, it has been shown later,[8] by transport and magnetic measurements, that the degree of magnetic order hardly changes at any field value near the field H_a. I believe that the curve in Fig. 1(a) can be explained by the presence of anisotropy fields of the kind introduced before. In order to prove this I have performed a series of different experiments, under new experimental conditions. The experiment I am going to describe now was suggested to me by the fact that if the anisotropy really exists then it could be removed by cooling, very slowly, in an alternating field ($h \exp i\omega t$) high enough to align the spontaneous magnetization. In this way no preferred direction would be created during cooling. A somewhat equivalent method was applied recently[9] to CuMn and was shown to be successful in removing the anisotropy from these alloys.

The curve in Fig. 1(b) was recorded after the $Ni_{79}Mn_{21}$ alloy had been cooled down to 1.5 K in an alternating field ($h \exp i\omega t$) of about 1 kG and at a rate of about 1 cycle/degree. Moreover, to obtain the initial magnetization curve (dashed) the sample was demagnetized at 1.5 K according to the usual technique, i.e., by reducing gradually the amplitude h to zero. Obviously, the new loop has lost the most characteristic features of the ZFC curve (i.e., the linearity, the reversibility, and the double-step-like shape). The only link with the ZFC curve is that the areas (or the energy losses) are found to be approximately the same in the two cases. More generally we observe that the losses are roughly independent of the cooling field also. Such a result suggests that, to a first approximation, the degree of irreversibility in $Ni_{79}Mn_{21}$ is independent of the macroscopic anisotropy and is more consistent with the predictions of Refs. 1 and 2 than with Ref. 4.

Now, I would like to present further experimental evidence showing that the anisotropy fields are principally determined by the spatial distribution of the spontaneous magnetization during cooling rather than by the cooling field eventually present: In a first experiment I have cooled the sample in $H = 0$ down to a temperature of 25 K, slightly lower than the de Almeida–Thouless temperature, $T_{AT} \simeq 30$ K (this choice will be justified below). A steady field of $\simeq 100$ G was then applied (at 25 K) and the sample was subsequently cooled in this field down to 1.5 K. After that the dashed curve shown in Fig. 2 was recorded.

In a second experiment I also cooled the alloy in $H = 0$ down to 25 K. But this time, arriving at this temperature, I cycled the field up to 100 G and back to $H = 0$. By doing so I have prepared the sample with a remanent magnetization the value of which is comparable to the magnetization that the alloy had during the preceding cooling in $H = 100$ G. The partially magnetized sample was then cooled in $H = 0$ down to 1.5 K and the full curve shown in Fig. 2 was subsequently plotted. It is clear that the two curves in Fig. 2 are very similar. The small difference which is seen near the origin can be accounted for naturally by the difference of the values of the two magnetizations during cooling. The temperature of 25 K mentioned above was chosen because it minimizes this difference while satisfying the condition $(T_{AT} - 25\ \text{K})/T_{AT} \ll 1$.

Thus, the above results show that the relevant parameter in determining the relationship (m vs H) is not the cooling field itself but the state of m during cooling. Further support for this claim comes from the observation that the hysteresis loop (of $Ni_{79}Mn_{21}$) becomes independent (both in shape and position) of the strength of the cooling field as soon as the latter exceeds the technical saturation limit (a few hundred gauss).

It is to be noted that (1) the same kind of results as in Figs. 1 and 2 were obtained at 4.2 K except that the hysteresis losses were found to be

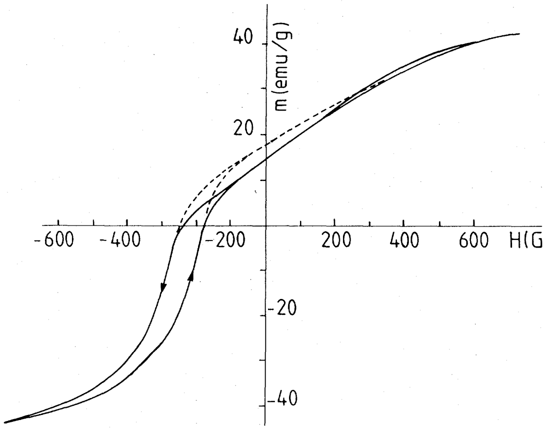

FIG. 2. Two hysteresis loops for $Ni_{79}Mn_{21}$ at 1.5 K: dashed curve, recorded after cooling in a steady field of 100 G from 25 to 1.5 K; full curve, recorded after cooling in $H = 0$ but in the presence of a remanent magnetization prepared at 25 K.

larger at this temperature; (2) each of the curves in Figs. 1 and 2 was found to be quite unchanged upon cycling the field between ± 30 kG at 1.5 K.

Let us now examine very briefly the Au$_{81}$Fe$_{19}$ sample. The associated hysteresis loop has a more conventional form than in the case of Ni$_{79}$Mn$_{21}$. However, a detailed study reveals some very interesting similarities between the two alloys. The loop displayed in Fig. 3(a) was plotted after cooling in $H = 0$ down to 1.5 K. The most abnormal features of this loop are connected with the initial magnetization curve (dashed branch) which I found to be remarkably linear and reversible (as in the NiMn case) as long as the applied field is lower than a certain limit H_a near which it intersects the cyclic loop (full branch). Very strong irreversibilities appear abruptly above $H = H_a$. This low-field behavior of the virgin curve is not usual and I believe it to be associated with anisotropy fields also. In order to prove this I have made the same experiment as in Fig. 1(b). More precisely, the curve in Fig. 3(b) was recorded after cooling in an alternating field of about 2 kG and demagnetizing the Au$_{81}$Fe$_{19}$ sample by reducing the amplitude of the alternating field to zero at 1.5 K. The new virgin curve (dashed) is found to be highly irreversible and has a more conventional form. As in Fig. 1(b) we observe a net increase in the initial slope dm/dH. This would be due to the suppression of the constraints exerted by the anisotropy forces on the spontaneous magnetization. It is to be stressed that whereas the ZFC loops associated with the two alloys [Fig. 1(a) and Fig. 3(a)] are markedly different, the loops obtained after cooling in an alternating field have very similar shapes [Fig. 1(b) and Fig. 3(b)].

Other spin-glass-like alloys exhibit low-temperature behaviors similar to those reported here.[10] By analogy with pure spin-glasses I propose that the alloys in which both spin-glass and ferromagnetic orders coexist can be classified into two main groups according to the properties of their hysteresis loops at $T \ll T_{AT}$: those for which the anisotropy fields are relatively low, as in CuMn, and those for which the anisotropy fields are relatively high, as in dilute AuFe. I suggest that Ni$_{79}$Mn$_{21}$ belongs to the former group whereas Au$_{89}$Fe$_{19}$ belongs to the latter one. The analysis of the data is obviously oversimplified, but it is a necessary first step in view of the complexity of the problem. A more detailed analysis together with an extended study of the irreversibilities in Au$_{81}$Fe$_{19}$ and Ni$_{79}$Mn$_{21}$, up to 40 K, will be reported elsewhere.

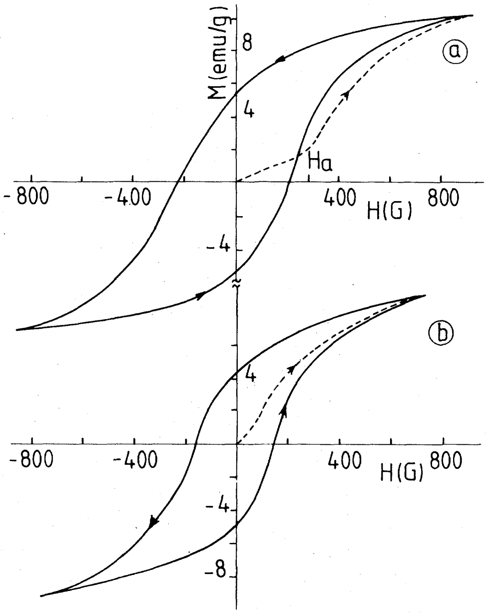

FIG. 3. Two complete loops for Au$_{81}$Fe$_{19}$ at 1.5 K recorded in the same conditions as in Fig. 1.

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