Thermomagnetic hysteresis effects in NiMn and NiMnPd thin films

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dc magnetization measurements, for zero-field cooled (M_{ZFC}) and field-cooled (M_{FC}) cases, have been carried out for flash-evaporated Pd-doped NiMn thin films. These included reentrant phases $(Ni_{76-x}Pd_x)Mn_{24}$, for $0 \leqslant x \leqslant 5$, and $Ni_{75}Mn_{23}Pd_2$, a pure spinglass phase. The studies were performed over the temperature range 3–300 K. Low-field magnetization measurements show the irreversibility effect (M_{ZFC} and M_{FC} diverge) at temperatures below the Curie temperature T_c. In $Ni_{75}Mn_{23}Pd_2$, M_{ZFC} falls below M_{FC}, as usually observed. However, in reentrant compositions, M_{ZFC} crosses M_{FC} upon warming into the ferromagnetic regime, where it stays above M_{FC} at temperatures below T_c. This unusual behavior is attributed to a model of Imry and Ma in which, in a ferromagnet with antiferromagnetic impurities, the impurities can couple to the host ferromagnetic alignment and force the system to break into domains antiferromagnetically coupled to each other. Field-cooled hysteresis measurements indicate the uniaxial anisotropy in these samples to be small, in contrast with the rigid uniaxial anisotropy reported for the corresponding polycrystalline bulk samples. Since the lattice-orbit coupling is weak in the amorphous phase, this clearly demonstrates that the physical origin of the unidirectional anisotropy is associated with the spin-orbit coupling.
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We have recently observed[1] that the addition of nonmagnetic Pt and Pd impurities significantly modifies the magnetic properties of bulk NiMn alloys. For instance, the hysteresis loops are broadened, and the shift of these loops induced by a cooling field is increased. The latter corresponds to an increase in $-H_a$ (rigid component of the unidirectional anisotropy) induced by the impurities. But, the Curie temperature T_c, the freezing temperature T_f, and the saturated magnetization M_s remain practically unchanged. Very recently, we prepared some NiMn and NiMnPt amorphous thin films from the corresponding bulk samples and pointed out that the amorphous condition causes a notable change in the magnetic ordering within the range of the mean free path of conduction electrons, and also Pt impurities give some additional peculiarities in the resistivity and magnetization.[2] The main purpose of this article is to present low-field magnetic susceptibility measurements in Pd doped amorphous NiMn thin films and to suggest a possible magnetic order to account for the thermomagnetic hysteresis observed in ferromagnetic regime.

$Ni_{76}Mn_{24}$, $Ni_{75}Mn_{23}Pd_2$, $Ni_{71}Mn_{24}Pd_5$, and $Ni_{74}Mn_{24}Pd_2$ amorphous films were prepared by flash evaporation techniques, as previously discussed.[3] Their amorphous nature was verified by x-ray diffraction. The film thickness was determined by means of a DEKTAL profilometer. The thicknesses of the films used in this experiment were between 100 and 150 nm. All the magnetization measurements were made with a Quantum Design superconducting quantum interference device magnetometer with a dc field parallel to the film surfaces.

The zero field cooled (ZFC) and field cooled (FC) magnetic hysteresis loops were obtained after cooling the samples from above T_c, in zero field and in $H_{\mathrm{cool}}=5$ kOe, respectively. H_{cool} was made large enough to saturate the thermoremanence. We note that the measured loops for both cases display either a smooth and gradual, or a sharp and boxlike shape. These forms are often associated with the relative size of the macroscopic anisotropies (unidirectional H_a and uniaxial H_u), M_s, the rigidity of the unidirectional anisotropy, and the degree of coupling between these anisotropies and M_s. It is also interesting to note that the ZFC and FC loops are similar in most respects, but with the difference that the FC magnetization changes much more abruptly near the coercive field compared to the corresponding ZFC case. This behavior may be attributed to the fact that the FC samples behave more like a single spin-glass domain with unidirectional anisotropy. The hysteresis loops are asymmetric with respect to the origin, indicating the rotations of some individual spins or spin clusters, oriented in different directions within the single domain, against the unidirectional anisotropy along the applied field. Figure 1 shows a typical ZFC and FC loop for the $Ni_{75}Mn_{23}Pd_2$ thin film. Each loop gives the basic parameters M_s, the coercive field H_c^+ for the initial magnetic field direction, and H_c^- for the reversed field direction. These parameters were obtained quantitatively from the measured loops, as summarized in Table I. T_c and T_f are also given in the table, as estimated from the M versus T curves.

The dc magnetic susceptibilities were measured first by ZFC to 3 K followed by heating in the measuring field of 20 Oe. FC data were then measured by cooling through T_c in the measuring field. These data are shown in Fig. 2. It is

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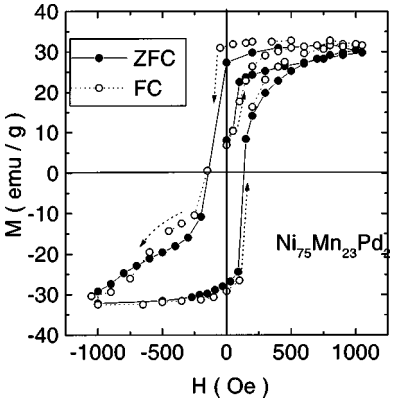

FIG. 1. Magnetic hysteresis loop for the $Ni_{75}Mn_{23}Pd_2$ film. Open and closed symbols are for FC and ZFC cases, respectively.

clear from the figures that irreversibility effects persist up to T_c, as evidenced by the splitting of FC and ZFC curves. In spinglass phases, M_{ZFC} falls below M_{FC}, as usually observed. However, in reentrant compositions, M_{ZFC} crosses the M_{FC} curve upon further warming into the ferromagnetic phases.

Unlike the bulk samples mentioned above, not only the unidirectional anisotropy, but also the other parameters are affected by Pd impurities. The unidirectional anisotropy completely loses its rigidity for $Ni_{71}Mn_{24}Pd_5$ and $Ni_{75}Mn_{23}Pd_2$ thin films while, for the $Ni_{74}Mn_{24}Pd_2$ and $Ni_{76}Mn_{24}$ thin films, the measured loop for the FC case is slightly displaced from the origin in the $-H$ direction, i.e., opposite to the cooling field direction. The amount of this displacement (the negative value of the average coercive field) for the former film is found to be about 100 Oe, whereas this amount is about 500 Oe for the corresponding bulk sample.[1] Similar behavior has been reported on NiMn films (whether amorphous or not) by Hauser and Bernardini.[4,5] They suggested that *a displaced hysteresis loop is the result of a specific crystal structure and is not necessarily linked to the spin glass state*. However, recent torque,[6] ESR,[7] and susceptibility[8] experiments clearly demonstrated that the anisotropy observed in these systems is purely unidirectional and rotates elastically at lower temperatures, but dissipatively at higher temperatures, resulting in larger undisplaced hysteresis. Therefore, one can claim that in the case of breaking long range crystal order, owing to

TABLE I. Parameters obtained from hysteresis loops at $T=3$ K after cooling in zero (ZFC) and 5 kOe (FC), and the Curie and freezing temperatures pertaining to these samples. H_a is calculated from H_c^+ and H_c^-. Upper and lower coercive field values shown correspond to the FC and ZFC cases, respectively.

Sample	M_s (emu/g)	H_c^+ (Oe)	H_c^- (Oe)	H_a (Oe)	T_c (K)	T_f (K)
$Ni_{76}Mn_{24}$	40	200	−360	80	270	35
		180	−280	50		
$Ni_{75}Mn_{23}Pd_2$	35	135	−145	5	240	50
		140	−140	–		
$Ni_{74}Mn_{24}Pd_2$	~2	725	−925	100	180	60
		725	−725	–		
$Ni_{71}Mn_{24}Pd_5$	24	280	−340	30	230	50
		280	−280	–		

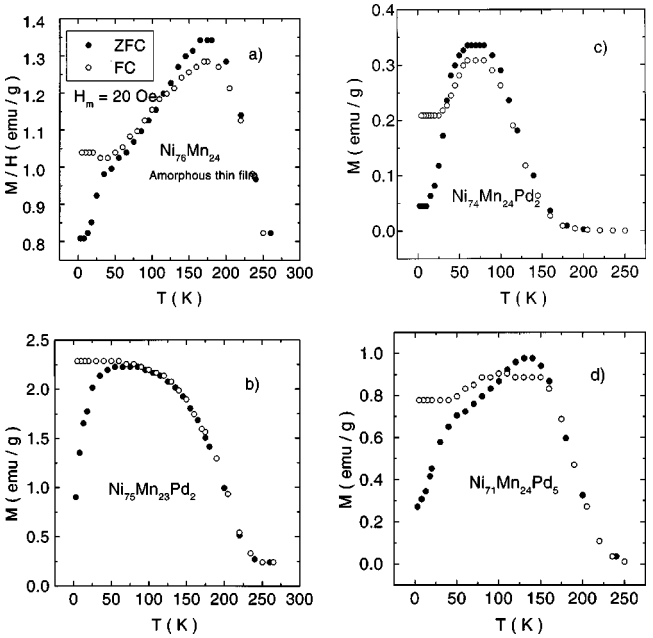

FIG. 2. M vs T of (a) $Ni_{76}Mn_{24}$, (b) $Ni_{75}Mn_{23}Pd_2$, (c) $Ni_{74}Mn_{24}Pd_2$, and (d) $Ni_{71}Mn_{24}Pd_5$. Note that the onset of irreversibility occurred at temperatures just above the Curie point T_c. Open symbols show data obtained while cooling in 20 Oe, while closed symbols correspond to the ZFC case.

weak lattice-orbit coupling averaged over a whole sample, the unidirectional field becomes much more free to rotate along the applied field direction. Thus, we believe that the dynamic character of the unidirectional anisotropy depends on the local crystal structural order as being responsible for the hysteresis. On the other hand, and in contrast with previous observations on bulk NiMnPd alloys, a considerable reduction in M_s is clearly seen in these data, with increasing Pd substitution up to 2 at.%. As the Pd content is further increased, Pd impurities favor magnetic order; T_c and M_s increase (see Table I). Since a broadening of the magnetic hysteresis observed in these films has been attributed to the loss of the unidirectional anisotropy, few percent Pd impurities definitely enhance this anisotropy, whether or not the sample is polycrystalline.

As for the irreversibility effect observed in M versus T, it is remarkable to note that this irreversibility appears at temperatures just below T_c. The other, most striking feature of the M versus T curves is that, in the ferromagnetic phase, at temperatures below T_c, M_{FC} stays below M_{ZFC}. Although many authors examined the weak and strong irreversibility effects observed at temperatures below T_f, based on the models relying upon the freezing of the transverse spin components[9] and the longitudinal spin components,[10] respectively, most of them overlook the irreversibility observed at much higher temperatures than T_f. At first sight, this irreversible effect seems to be a puzzle, however a model proposed by Imry and Ma[11] may account for this experimental behavior. According to this model, in a classical ferromagnet with antiferromagnetic site impurities, a random component of the field due to such impurities can couple to the host ferromagnetic alignment and force the system to break into domains antiferromagnetically coupled to each other. As the sample passes through T_c, either in FC or ZFC cases, the

onset of long-range ferromagnetic ordering accompanied by antiparallel alignment of some spins at antiferromagnetic impurity sites may yield a decrease in the average moment per atom. Since the degree of antiferromagnetic alignment along the applied field for the FC case will be much higher than that for the ZFC case, it is reasonable to expect that M_{FC} stays below M_{ZFC}. As a conclusion, our experiment seems to favor the Imry–Ma model to account for this unusual irreversibility effect observed in the NiMn system.

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