Rotational magnetic properties of Ni-Mn and Au-Fe spin-glass alloys

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Magnetization-vector measurements were performed on field-cooled polycrystalline disks of $Ni_{75}Mn_{25}$ and $Au_{85}Fe_{15}$ rotated in a stationary field **H**. For $Ni_{75}Mn_{25}$ at 4.2 and 10 K, it is found that the induced anisotropy, which rotates rigidly with the sample for all H, is predominantly unidirectional but with uniaxial and higher-order components that are not negligible. Moreover, the rotational magnetization $\mathbf{M}_{rot}$ is seen to be a unique function of the total effective field $\mathbf{H}_{eff}$ composed vectorially of **H** plus the anisotropy field $\mathbf{H}_K$, where $\mathbf{M}_{rot}$ extends from its saturation value down to zero at $\mathbf{H}_{eff}=0$. For $Ni_{75}Mn_{25}$ at higher temperatures, it is found that for H above a threshold value the direction of $\mathbf{H}_K$ changes relative to the rotating sample. This phenomenon is also observed in $Au_{85}Fe_{15}$ at 4.2 K, and the directional changes of $\mathbf{H}_K$ are seen to be a dissipative process, which is macroscopically frictional. Qualitative comparisons are made with the available theory.

INTRODUCTION

Although it is generally agreed that a spin-glass (SG) state requires that a material have both atomic disorder and spin frustration,[1] these necessary (and sufficient) conditions do not specify the SG ground state uniquely, in that under identical circumstances there are many different possible spin configurations with essentially the same magnetic interaction energy.[2] Nevertheless, despite the large degeneracy of states, most of the characteristic magnetic properties of a SG are experimentally reproducible if measured with care.[3] An important example is the saturation value of the thermoremanent magnetization (TRM) achieved by cooling slowly from above the freezing temperature T_g in a sufficiently strong external field. Typically, the saturation TRM decays with time, but the decay is usually slow enough that subsequent measurements with a cycled field yield a hysteresis loop that is virtually closed. However, the nature of the hysteresis loops measured after field cooling to low temperatures is qualitatively different for different spin glasses.

In the much-studied case of the SG alloys of Mn in Cu, Ag, or Ni, it was discovered long ago[4] that at low temperatures (well below T_g) the M versus H hysteresis loops measured after field cooling are narrow and displaced from the origin in the $-H$ direction, i.e., opposite to the direction of the field applied during cooling $\mathbf{H}_{cool}$. In such a loop, when a negative field large enough to reverse the magnetization M is removed, M returns to the positive TRM value. Thus, the anisotropy produced by field cooling is unidirectional, the single easy direction of magnetization being that of $\mathbf{H}_{cool}$. Contrastingly, in most other SG materials, including prototypically the alloys of Au-Fe, the low-temperature hysteresis loops measured after field cooling are broad and symmetric about the origin. From the latter behavior, it has been tempting to consider that the $\mathbf{H}_{cool}$-induced anisotropy of Au-Fe is uniaxial—for which, however, there has been no independent evidence; whereas the unidirectional anisotropy of field-cooled Ni-Mn and Cu-Mn has been clearly confirmed by torque (Ref. 5), ESR (Ref. 6), and NMR (Ref. 7) experiments.

Moreover, the unidirectional anisotropy manifested in certain spin glasses has found firm theoretical justification.[8] Specifically, when spin-orbit interactions of Dzyaloshinsky-Moriya (DM) form are taken into account but considered weak relative to the isotropic spin-spin interactions, it was shown that a rigid rotation of all the randomly oriented spins of a SG about a single axis produces an energy change of unidirectional symmetry. Ideally, this process is perfectly elastic, which may explain why displaced loops are often seen to be narrow, involving very little hysteresis energy loss. However, if the DM interactions are relatively strong, they will cause some spins or spin clusters to flip discontinuously as the net magnetization induced by $\mathbf{H}_{cool}$ responds to a changing external field. Not only is this a dissipative process, but the unidirectional anisotropy will have changed in orientation, corresponding to what has been called a "redefinition" of the anisotropy.[9]

Experimentally, such processes can be probed to best advantage with rotating external fields of fixed size rather than with the cycled fields used in hysteresis-loop measurements. Indeed, magnetic torque measurements have been carried out on a dilute CuMn alloy below but not far below T_g, and it was found that sample rotations in various fields resulted in substantial orientational changes of the anisotropy produced originally by field cooling.[10] A similar phenomenon was revealed by torque measurements on Au-Fe at fields above a threshold value; at lower fields the measured torque was reversible, showing that the anisotropy turns rigidly with the rotating alloy sample.[11]

However, revealing as they may be, magnetic torque experiments sense only the component of the sample magnetization transverse to the applied field **H**. For a more complete characterization of rotational magnetic processes in a SG, the magnetization components parallel as well as perpendicular to **H** need to be measured at the same time. We have performed such magnetization-

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vector measurements on field-cooled samples of Ni-Mn and Au-Fe. Our preliminary results at 4.2 K, reported earlier in brief,[12] show that in Ni-Mn the anisotropy is basically unidirectional and rotates rigidly with the sample, while the magnetization undergoes some anomalous changes in magnitude, and that in Au-Fe the anisotropy turns dissipatively relative to the rotating sample when the external field exceeds a threshold value. In the present paper, our more complete results are reported in full, including the changes in Ni-Mn with increasing temperature, which are such that its rotational magnetic behavior becomes similar to that of Au-Fe at low temperatures.

RESULTS AND DISCUSSION

For our experiments, two identical sets of pickup coils were mounted in geometric quadrature about the cryostat tail in which a vibrating-sample magnetometer assembly was suspended within the gap of an electromagnet. One coil set detects the sample magnetization parallel to the fixed external field **H** and the other coil set the magnetization perpendicular to **H**. These longitudinal and transverse magnetization components (M_L,M_T) both lie in the horizontal plane of the circular-disk sample, which is rotated with the magnetometer assembly about the vertical axis of vibration. Thus, the total magnetization of the sample is measured as a vector of determined magnitude and orientation for different angles of sample rotation in a fixed external magnetic field.

Our samples of study were polycrystalline alloy disks of nominal compositions, $Ni_{75}Mn_{25}$ and $Au_{85}Fe_{15}$, both of which had been cut from arc-melted ingots. The $Ni_{75}Mn_{25}$ disk (5-mm diameter, 0.6-mm thick) was annealed for 3 days at 900 °C and water quenched. In this disordered state, its low-field longitudinal magnetization M_L was seen to increase to a peak at 135 K with rising temperature T after zero-field cooling to 4.2 K and then to increase again and level out as T is lowered from above 135 K. This manner of irreversibility identifies a SG state with a freezing point T_g of 135 K. Moreover, the high ratio (~ 11) of the peak M_L value at T_g to the initial M_L value at 4.2 K (both measured at low H) is consistent with the sample composition being close to but on the nonferromagnetic (high-Mn) side of the multicritical point in the magnetic phase diagram of disordered Ni-Mn.[13] In the case of the $Au_{85}Fe_{15}$ disk (5-mm diameter, 0.4-mm thick), which was annealed for 3 h at 600 °C and water quenched, a similar irreversibility observed in M_L versus T indicates a SG state with $T_g = 52$ K, consistent with previous work.[14] In all our experiments, the samples were initially cooled from above T_g to the temperature of measurement in $H_{cool} = 10$ kOe, which was sufficiently strong to saturate the TRM of either alloy.

For $Ni_{75}Mn_{25}$ in the saturated-TRM state at 4.2 K, the M versus H hysteresis loop measured conventionally is displayed in Fig. 1. The loop is very narrow and is displaced in the $-H$ direction even though M reaches equal and opposite values at the cycled-field extremes. This kind of displaced loop is typical of field-cooled Ni-Mn at $T \ll T_g$ and has been attributed to a H_{cool}-induced unidirectional anisotropy.[4] However, from such an anisotropy, one might expect an abrupt reversal of M at $H = -H_K$, where H_K is the anisotropy field, whereas the measured loop, even when corrected for demagnetization (dashed curve labeled D), exhibits a very gradual change. Indeed, similar measurements on a series of disordered Ni-Mn alloys in the saturated-TRM state at 4.2 K have previously shown that as the Mn concentration rises through 25 at. % the reversal of M on the displaced loop becomes increasingly more gradual, starting from an almost discontinuous change in $Ni_{77}Mn_{23}$ (which is a reentrant SG).[15]

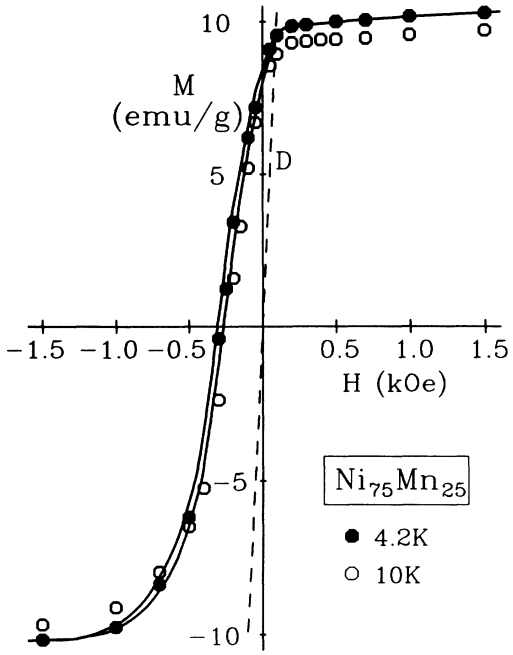

FIG. 1. Magnetization-field hysteresis loop for $Ni_{75}Mn_{25}$ in a saturated-TRM state at 4.2 K. Solid and open circles are from rotational measurements at 4.2 and 10 K, respectively, of longitudinal magnetization at $\theta = 0$ ($H > 0$) and $\theta = 180°$ ($H < 0$). Demagnetization D curve shown as dashed line.

In Fig. 1, we have also plotted (as solid circles) the values of the longitudinal magnetization (M_L) for different H at 4.2 K measured before the field-cooled $Ni_{75}Mn_{25}$ sample was rotated and after the sample was rotated by 180° relative to **H**. In the latter case, for appropriate comparison with the hysteresis loop, $-M_L$ is plotted versus $-H$. Clearly, these results trace a curve that runs closely along the narrow displaced loop, thus confirming the gradual reversal of M as a real and peculiar property. Moreover, since no transverse magnetization M_T was observed in this experiment, the measured M_L corresponds to the total magnetization, which is seen to vanish at the average coercive field of the displaced loop. This additional anomaly will be discussed further with the complete results of our rotational experiments.

In these experiments (with reference to the vector diagram of Fig. 2), the $Ni_{75}Mn_{25}$ sample was rotated slowly such that the angle θ between the $\mathbf{H}_{cool}$ direction in the

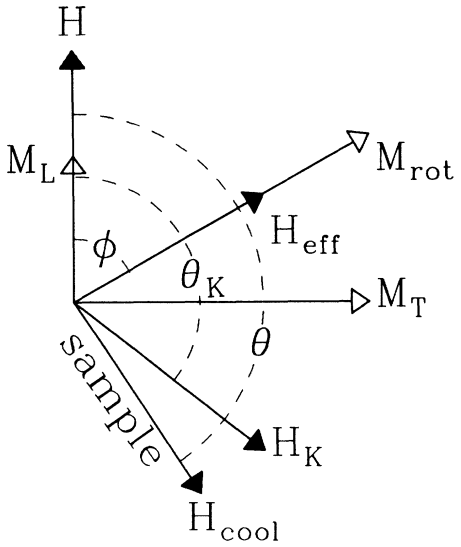

FIG. 2. Magnetic vector diagram for rotational measurements on sample cooled in $\mathbf{H}_{cool}$ and then turned by angle θ relative to fixed external field $\mathbf{H}$, as discussed in text.

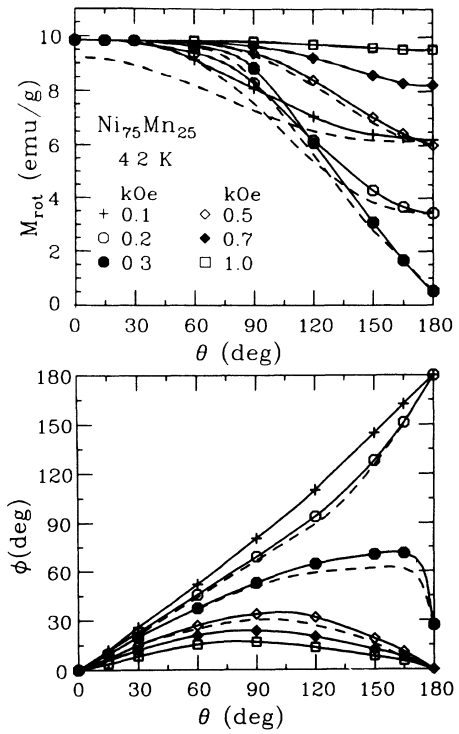

FIG. 3. $Ni_{75}Mn_{25}$ in a saturated-TRM state at 4.2 K. Magnitude of rotational magnetization $\mathbf{M}_{rot}$ and its orientational angle ϕ relative to external field $\mathbf{H}$ for increasing and decreasing sample-rotation angle θ (solid and dashed curves, respectively) in various H.

sample and the fixed external field $\mathbf{H}$ increased from 0° to 180° and then decreased back to 0°. During this rotational cycle, the longitudinal and transverse magnetizations were measured simultaneously. The M_L values were each corrected for a small isotropic contribution linear in H, which was evaluated from the residual slope of the hysteresis loop at positive H (Fig. 1). The values of M_T and the corrected M_L were then combined in quadrature to give the magnitude of the total rotational magnetization $\mathbf{M}_{rot}$ and its orientational angle ϕ relative to $\mathbf{H}$.

Our results for $Ni_{75}Mn_{25}$ at 4.2 K are displayed in Fig. 3, where M_{rot} and ϕ are plotted against the sample-rotation angle θ for different values of H. In all cases, the increasing-θ branches (solid curves through plotted points) and the decreasing-θ branches (dashed curves, points omitted for clarity) are in fairly close proximity, indicating little rotational hysteresis. Regarding ϕ versus θ, we note that all the curves start out linearly at low θ with a decreasing slope for increasing H but that at higher θ they split into two groups. The curves for $H<0.3$ kOe continue to rise until ϕ reaches 180° at $\theta=180°$, while those for $H>0.3$ kOe pass through a maximum and descend to $\phi=0$ at $\theta=180°$. The curves for $H=0.3$ kOe appear exceptional in dropping from a maximum to a low but nonzero ϕ at $\theta=180°$. However, the latter ϕ value was seen to creep steadily downward with time (the time dependence being negligibly small elsewhere) and possibly reaches 0°. Allowing for this possibility, the overall behavior of ϕ versus θ is qualitatively consistent with a unidirectional anisotropy that rotates rigidly with the sample and with the anisotropy field being slightly below 0.3 kOe. However, although the directional changes of $\mathbf{M}_{rot}$ are easy to understand, the variations of its magnitude seem anomalous. But we note that the decrease of M_{rot} as θ approaches 180° becomes especially large for H close to 0.3 kOe, which is the approximate size of the anisotropy field. This interesting correlation will be discussed further later.

In a simple macroscopic analysis of the ϕ versus θ results in Fig. 3, we consider that the anisotropy field $\mathbf{H}_K$ rotates rigidly with the sample, so that the angle θ_K between $\mathbf{H}_K$ and $\mathbf{H}$ equals the sample-rotation angle θ. Since the orientation of $\mathbf{M}_{rot}$ is determined by the balancing of the torques exerted on it by $\mathbf{H}_K$ and $\mathbf{H}$, it follows that

$$H_K \sin(\theta-\phi)=H\sin\phi \ , \tag{1}$$

which is equivalent to the condition that $\mathbf{M}_{rot}$ lies parallel to the total effective field $\mathbf{H}_{eff}=\mathbf{H}_K+\mathbf{H}$ as shown in Fig. 2. Substituting our measured values of ϕ for different H and θ into Eq. (1), we obtain the values of H_K that are plotted in Fig. 4(a) as a function of $\theta-\phi$, the angle between $\mathbf{M}_{rot}$ and $\mathbf{H}_K$. Despite the scatter of points, we see that H_K is not constant but varies fairly systematically. Thus, in $Ni_{75}Mn_{25}$ at 4.2 K the anisotropy produced by field cooling is not purely unidirectional but contains some higher-order components. Analytically, this result is independent of any changes in the magnitude of $\mathbf{M}_{rot}$.

For a formal rationale of the variations of H_K with $\theta-\phi$, the macroscopic magnetic energy is expressed as

$$E_m=-\sum_n K_n \cos n(\theta-\phi)-HM_{rot}\cos\phi \ , \tag{2}$$

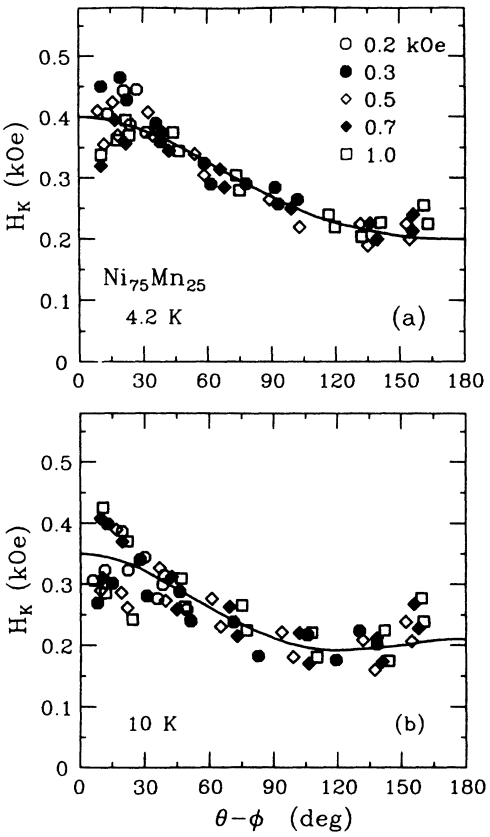

FIG. 4. Anisotropy field H_K versus angle $\theta-\phi$ for $Ni_{75}Mn_{25}$ at (a) 4.2 K and (b) 10 K in various external fields. Curves represent best fits to Eq. (4).

where the anisotropy energy is Fourier expanded ($n=1,2,\ldots,$) with the primary easy direction at $\theta=0$. For equilibrium at fixed H and θ, $\partial E_m/\partial\phi=0$, which together with Eq. (2) gives

$$\sum_n nH_n \sin n(\theta-\phi)=H\sin\phi\ , \tag{3}$$

where $H_n=K_n/M_{\rm rot}$, the nth-order component of the anisotropy field. Comparison of Eqs. (1) and (3) yields

$$H_K=\sum_n nH_n \sin n(\theta-\phi)/\sin(\theta-\phi)\ ,$$

which, truncated at $n=3$, becomes

$$H_K=H_A+H_B\cos(\theta-\phi)+H_C\cos 2(\theta-\phi)\ , \tag{4}$$

with

$$H_A=H_1+3H_3,\ H_B=4H_2,\ H_C=6H_3\ .$$

The best fit of Eq. (4) to the points in Fig. 4(a) is represented by the curve drawn in the figure, for which the values of H_A, H_B, H_C, and of H_1, H_2, H_3 are listed in Table I. We see that the predominant unidirectional component of the anisotropy (H_1) is somewhat less than 0.3 kOe, as expected, and that the uniaxial and third-order components (H_2 and H_3) are 1 and 2 orders of magnitude smaller, respectively, but cannot quite be neglected.

These results at 4.2 K appear to disagree with the theory involving Dzyaloshinsky-Moriya interactions that predicts a purely unidirectional anisotropy for rigid spin rotations in a SG, as described earlier. Hence, we repeated our rotational measurements on $Ni_{75}Mn_{25}$ at a somewhat higher temperature (10 K) to find if there is any significant growth of the uniaxial anisotropy component, which would indicate that its appearance is not intrinsic but thermally activated. Our results at 10 K for sample rotations of 0° and 180° are included in Fig. 1 (open circles), where they clearly resemble what was measured at 4.2 K. This close correspondence was found to exist at all sample-rotation angles, showing that the anisotropy continues to rotate rigidly with the sample. The values deduced for H_K versus $\theta-\phi$ at 10 K are plotted in Fig. 4(b), where again the curve represents the best fit with Eq. (4). The corresponding components of the anisotropy field are listed in Table I, and we see that in going from 4.2 to 10 K the uniaxial component H_2 decreases by about the same relative amount as the unidirectional component H_1; the higher-order H_3 increases but remains small compared to H_1. We can therefore conclude that the appearance of H_2 is an intrinsic effect, possibly deriving from anisotropic interactions other than those of Dzyaloshinsky-Moriya form.[16]

From each value of H_K obtained via Eq. (1) from our rotational data on $Ni_{75}Mn_{25}$, we also determined the magnitude of the total effective field $\mathbf{H}_{\rm eff}$. We proceeded to plot each measured value of $M_{\rm rot}$ against the corresponding $H_{\rm eff}$, and the plots for both 4.2 and 10 K are displayed in Fig. 5. For each temperature, the $M_{\rm rot}$ versus $H_{\rm eff}$ points for different H and θ describe essentially a universal curve that rises linearly from the origin and rapidly levels out as $M_{\rm rot}$ approaches the saturated TRM value. The initial slopes of these curves, though smaller than the slope of the demagnetization D curve, are much larger than the slopes of the magnetization curves for zero-field cooling (shown as ZFC). Thus, $M_{\rm rot}$ versus $H_{\rm eff}$ represents a strong but gradual magnetization process with $M_{\rm rot}$ vanishing at zero $H_{\rm eff}$. Since zero $H_{\rm eff}$ occurs when $\mathbf{H}=-\mathbf{H}_K$, the vanishing of $M_{\rm rot}$ is directly related to its disappearance at the coercive field of the displaced loop in Fig. 1. In fact, the $M_{\rm rot}$ versus $H_{\rm eff}$ curves in

TABLE I. Components of the anisotropy field H_K (in kOe) for $Ni_{75}Mn_{25}$ at 4.2 and 10 K, as determined from the curves of fit in Fig. 4.

T (K)	H_A	H_B	H_C	H_1	H_2	H_3
4.2	0.285	0.100	0.015	0.2775	0.0250	0.0025
10	0.245	0.070	0.035	0.2275	0.0175	0.0058

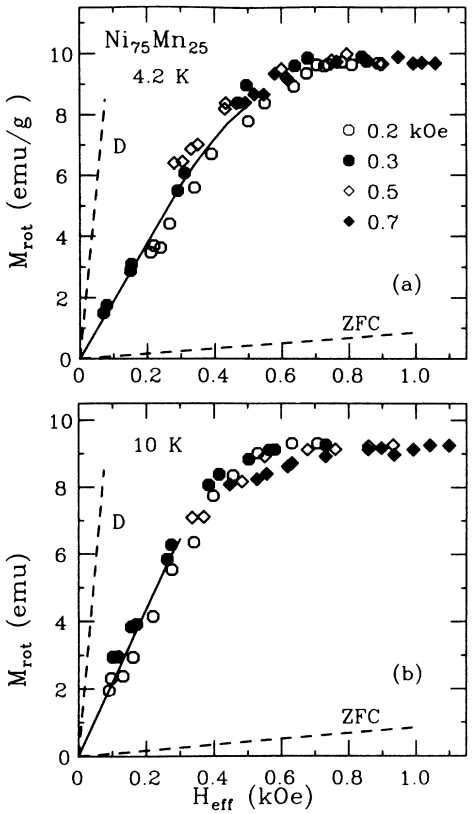

FIG. 5. Rotational magnetization M_{rot} versus total effective field H_{eff} for $Ni_{75}Mn_{25}$ at (a) 4.2 K and (b) 10 K in various external fields. Demagnetization D and zero-field-cooled (ZFC) curves shown as dashed lines.

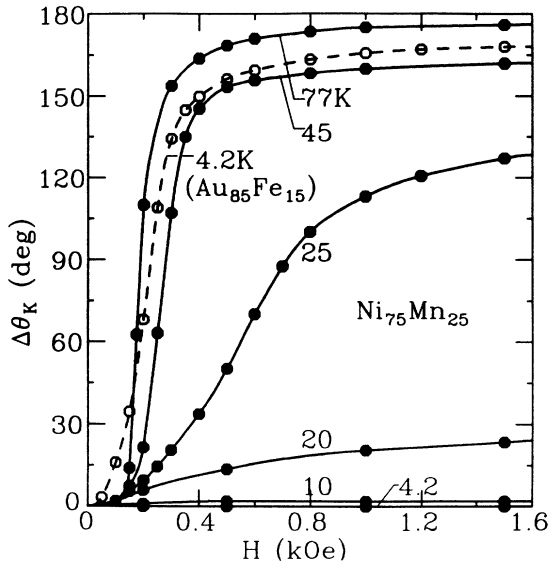

FIG. 6. Change in orientational angle ($\Delta\theta_K$) of anisotropy field relative to $\mathbf{H}_{cool}$ direction in $Ni_{75}Mn_{25}$ sample after rotation of 180° in various fields (abscissa axis) at different temperatures (solid curves). Same for $Au_{85}Fe_{15}$ at 4.2 K (dashed curve).

their entirety encompass the anomalously gradual magnetization changes associated with the hysteresis loops and place them in a more general context. The nature of these curves suggests to us that the sample in its saturated-TRM state consists of weakly coupled regions with a broad distribution of H_K values ranging down to zero, whose magnetization rotations occur over a correspondingly broad range of external fields. Although our preliminary model calculations appear to be bearing out this conjecture, other inhomogeneous modes of magnetization change (possibly involving nonrigid rotations) should also be considered.

When the temperature of $Ni_{75}Mn_{25}$ is raised above 10 K, the rotational magnetic behavior is seen to change radically. Since the behavior becomes very similar to that of $Au_{85}Fe_{15}$, which will be reported in detail later, it is more expeditious to follow the evolution with temperature in terms of the results of a simpler but related experiment. In this experiment, the sample in the saturated-TRM state is rotated by 180° in a given fixed **H**, which is then removed and the direction of the remanent magnetization ($\mathbf{M}_R$) is determined. Since the direction of $\mathbf{M}_R$ is the direction of the anisotropy field $\mathbf{H}_K$, as denoted by angle θ_K in Fig. 2, any observed difference between θ_K and 180° represents a rotation of $\mathbf{H}_K$ relative to the sample that arises from the 180° sample rotation in the given **H**. Such an experiment has previously been used in studying the SG alloy YTb.[17]

The results of this "remanence" experiment on $Ni_{75}Mn_{25}$ are displayed in Fig. 6, where $\Delta\theta_K$ ($=180°-\theta_K$, the directional shift of $\mathbf{H}_K$ in the sample) is plotted against H for different temperatures. At 4.2 K and nearly so at 10 K, $\Delta\theta_K$ remains at 0° for all H, as expected. However, at higher temperatures, $\Delta\theta_K$ rises when H exceeds a threshold level (~0.1 kOe) and approaches a limiting value at higher H. The high-H value of $\Delta\theta_K$ increases monotonically with increasing temperature (most rapidly near 25 K) and at 77 K it almost reaches the maximum possible level of 180°. This dramatic evolution signifies that at the lowest temperatures the torque exerted on $\mathbf{H}_K$ by $\mathbf{M}_{rot}$ (before **H** is removed) does not exceed the torque holding $\mathbf{H}_K$ to the sample; at higher temperatures the latter torque weakens and $\mathbf{H}_K$ turns towards $\mathbf{M}_{rot}$ until the two torques come into balance. At 77 K, which is still well below T_g (135 K), the evolution is almost complete; the torque that the sample can exert on $\mathbf{H}_K$ has become so weak that $\mathbf{H}_K$ turns close to the direction of $\mathbf{M}_{rot}$, which now is nearly parallel to **H**. Implicit in this description is that the directional changes of $\mathbf{H}_K$ in $Ni_{75}Mn_{25}$ are macroscopically frictional, which agrees with what was deduced earlier from analogous results on YTb.[17]

For comparison, we have also plotted in Fig. 6 our results for the "remanence" experiment on the $Au_{85}Fe_{15}$ sample at 4.2 K. The behavior of $Au_{85}Fe_{15}$ at this low temperature is obviously similar to that of $Ni_{75}Mn_{25}$ at a temperature between 45 and 77 K. The only difference is that in $Au_{85}Fe_{15}$ the rise of $\Delta\theta_K$ starts at a much lower H and is nearly linear, thus resembling more closely the

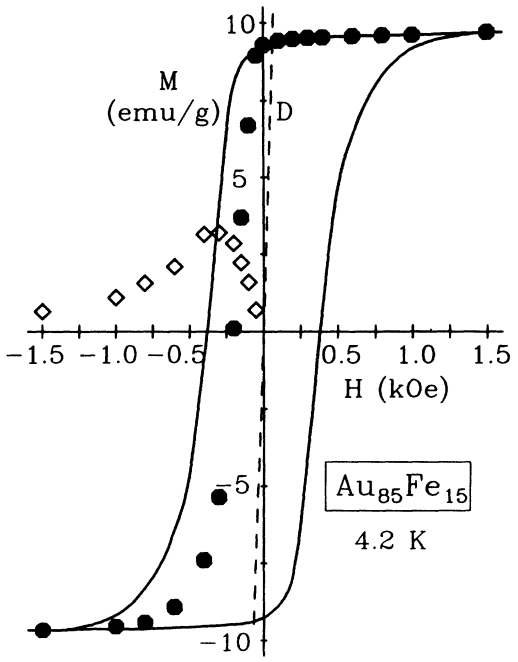

FIG. 7. Magnetization-field hysteresis loop for $Au_{85}Fe_{15}$ in a saturated-TRM state at 4.2 K. Solid circles are from rotational measurements at 4.2 K of longitudinal magnetization at $\theta=0$ ($H>0$) and $\theta=180°$ ($H<0$). Open diamonds are corresponding values of transverse magnetization at $\theta=180°$. Demagnetization D curve shown as dashed line.

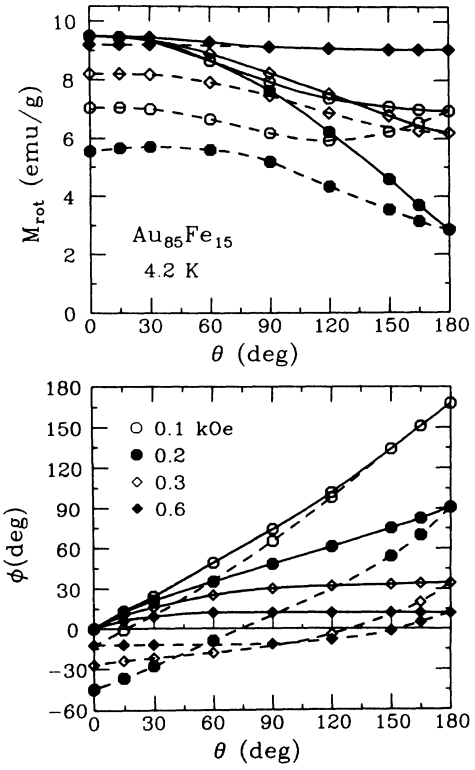

FIG. 8. $Au_{85}Fe_{15}$ in a saturated-TRM state at 4.2 K. Magnitude of rotational magnetization $\mathbf{M}_{rot}$ and its orientational angle ϕ relative to external field $\mathbf{H}$ for increasing and decreasing sample-rotation angle θ (solid and dashed curves, respectively) in various H.

linear behavior seen in YTb.[17] Hence, even at 4.2 K, the forces that hold the anisotropy field $\mathbf{H}_K$ in the $Au_{85}Fe_{15}$ sample are so weak that they allow $\mathbf{H}_K$ to turn frictionally in small external fields. This situation, as shown later, is borne out by our standard rotational measurements on this sample.

The conventionally measured hysteresis loop of our $Au_{85}Fe_{15}$ sample in the saturated-TRM state at 4.2 K is displayed in Fig. 7. Consistent with previous studies of field-cooled Au-Fe, the loop we obtained after repeated field cycling is broad and symmetric about the origin, in contrast to the narrow displaced loop for $Ni_{75}Mn_{25}$ obtained under the same conditions (Fig. 1). (Note that $Ni_{75}Mn_{25}$ exhibits broad undisplaced loops at higher temperatures, similar to the loop shown for $Au_{85}Fe_{15}$ at 4.2 K.) As in the case of $Ni_{75}Mn_{25}$, the longitudinal magnetization (M_L) of $Au_{85}Fe_{15}$ at 4.2 K was measured after sample rotations of 0° and 180° in various H. The results are represented by the closed circles in Fig. 7 (analogous to Fig. 1), and they do not quite follow the descending-H branch of the loop, showing that in $Au_{85}Fe_{15}$ the two processes are different. This difference is also manifested in a transverse magnetization M_T that was measured after the 180° sample rotation and is plotted versus $-H$ (open diamonds) in Fig. 7. Thus, unlike $Ni_{75}Mn_{25}$, for which no M_T was observed analogously at 4.2 K, the total magnetization vector of $Au_{85}Fe_{15}$ does not remain collinear with the field in which the sample is rotated. This behavior certainly implies that the anisotropy field $\mathbf{H}_K$ has shifted from its original direction in the field-cooled sample.

The full results of our rotational measurements on $Au_{85}Fe_{15}$ at 4.2 K are displayed in Fig. 8, where M_{rot} and ϕ are plotted versus θ for various H (as in Fig. 3). Again, to obtain $\mathbf{M}_{rot}$, the measured M_L values were corrected for a small isotropic contribution. Clearly, $Au_{85}Fe_{15}$ differs from $Ni_{75}Mn_{25}$ at 4.2 K in that M_{rot} and ϕ show considerable rotational hysteresis as θ is cycled between 0° and 180°. However, for $H=0.1$ kOe, ϕ changes almost reversibly and nearly reaches 180° at $\theta=180°$, indicating that the anisotropy field $\mathbf{H}_K$ turns almost rigidly with the sample—which is consistent with Fig. 6, where $\Delta\theta_K$ for $Au_{85}Fe_{15}$ is fairly small at this low field. At higher fields, ϕ versus θ gradually evolves into another simple but totally different behavior, such that for $H=0.6$ kOe ϕ rises to, and stays at, a small plateau value ($\sim 12°$) with increasing θ and then descends to a negative plateau value of equal size with decreasing θ.

The latter behavior is totally consistent with the following scenario (as described with reference to Fig. 2): the torque exerted on $\mathbf{H}_K$ by $\mathbf{M}_{rot}$ (which equals the torque on $\mathbf{M}_{rot}$ by $\mathbf{H}$ at equilibrium) grows with increasing θ until its strength equals that of the opposing torque holding $\mathbf{H}_K$ in the sample, at which point $\mathbf{H}_k$ and $\mathbf{M}_{rot}$ stay fixed in direction relative to $\mathbf{H}$ as the sample continues to turn—and this process repeats with equal and opposite angles relative to $\mathbf{H}$ when the sample rotation is reversed. This whole process is macroscopically frictional,

where on the atomic scale the rotational hysteresis losses arise from the spin flips required for $\mathbf{H}_K$ to change its direction in the sample.[9]

The changes in the direction of $\mathbf{H}_K$ in the $Au_{85}Fe_{15}$ sample at 4.2 K—and in the $Ni_{75}Mn_{25}$ sample at higher temperatures—are such that $\mathbf{H}_K$ always lies fairly close to the direction of $\mathbf{M}_{rot}$. This close proximity makes it virtually impossible to determine the symmetry components of $\mathbf{H}_K$, as we have done for $Ni_{75}Mn_{25}$ at 4.2 K, where $\mathbf{H}_K$ is rigidly fixed in the sample. We can only speculate that the $\mathbf{H}_K$ in these two SG alloys is predominantly unidirectional at all temperatures below T_g. In any case, the broad undisplaced hysteresis loop for $Au_{85}Fe_{15}$ in Fig. 7 can be readily attributed to the dissipative directional changes of $\mathbf{H}_K$ (whatever its symmetry) that accompany the changes of magnetization.

For very small rotations of the $Au_{85}Fe_{15}$ sample at 4.2 K, the changes of $\mathbf{M}_{rot}$ are found to be reversible in all fields. Since it can therefore be presumed that $\mathbf{H}_K$ turns rigidly with the sample at small θ, its magnitude can be determined from the expression

$$(d\phi/d\theta)_0^{-1}=1+H/H_K \ , \tag{5}$$

which, as derived from Eq. (1), shows that the inverse initial slope of ϕ versus θ should rise from unity as a linear function of H and that the slope of this rise depends inversely on H_K. Our experimental values for $(d\phi/d\theta)_0^{-1}$ are plotted against H in Fig. 9. From the linearity of this plot, we find via Eq. (5) that $H_K=0.52$ kOe. However, the symmetry of the anisotropy represented by this H_K value remains undefined, and it is uncertain that $\mathbf{H}_K$ retains this magnitude at higher θ when its direction in the sample changes.

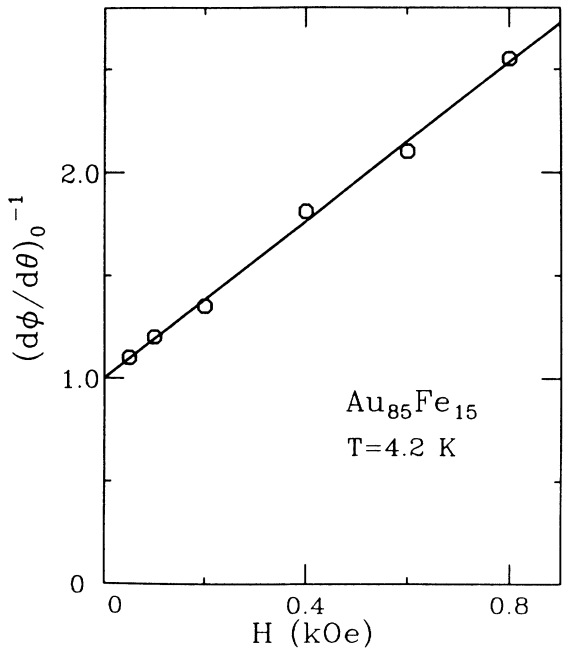

FIG. 9. Inverse initial slope of ϕ versus θ as function of external field for $Au_{85}Fe_{15}$ at 4.2 K. Straight line from Eq. (5) is for $H_K=0.52$ kOe.

The fact that $\mathbf{H}_K$ changes its direction in the $Au_{85}Fe_{15}$ sample plus the distinct possibility that its magnitude also may change with sample rotation make it very difficult to interpret the large variations of M_{rot} with θ evident in Fig. 8. Although the decreases of M_{rot} at high θ vary systematically with H in a like manner to that of $Ni_{75}Mn_{25}$ (Fig. 3), they cannot be converted analogously into a functional connection between M_{rot} and the total effective field H_{eff}, since H_{eff} is directly affected by any ambiguity in $\mathbf{H}_K$. Nevertheless, qualitatively, the variations of M_{rot} in $Au_{85}Fe_{15}$ do suggest that the sample acts inhomogeneously, similar to what was described earlier for $Ni_{75}Mn_{25}$ at 4.2 K—but with the additional complication that the $\mathbf{H}_K$ in each weakly coupled region turns in the sample (with a frictional torque that probably differs for different regions).

SUMMARY

The results of our rotational magnetic measurements have clarified macroscopically some of the basic SG properties of Ni-Mn and Au-Fe that relate to the anisotropy produced by field cooling. In the case of $Ni_{75}Mn_{25}$ at 4.2 and 10 K, where the anisotropy is seen to rotate rigidly with the sample, detailed analysis of our data reveals that the anisotropy is predominantly unidirectional but with uniaxial and higher-order components that are not negligible. The same data analysis also shows that the magnitude of the rotational magnetization ($\mathbf{M}_{rot}$) depends uniquely on the magnitude of the total effective field ($\mathbf{H}_{eff}$), which is the vector sum of the external field $\mathbf{H}$ and the anisotropy field $\mathbf{H}_K$. Unexpectedly, M_{rot} versus H_{eff} extends from the saturated TRM gradually down to the origin, suggesting that the sample subdivides into SG regions of different H_K. At higher temperatures, the direction of $\mathbf{H}_K$ in the $Ni_{75}Mn_{25}$ sample is seen to change when the external field in which the sample is rotated exceeds a threshold value.

Directional changes of $\mathbf{H}_K$ relative to the rotated sample are also observed in $Au_{85}Fe_{15}$ at 4.2 K, and our detailed measurements show that the redefinition of $\mathbf{H}_K$ is a dissipative (macroscopically frictional) process, as manifested in rotational hysteresis. The broad undisplaced hysteresis loop exhibited by field-cooled $Au_{85}Fe_{15}$ at 4.2 K is seen to be a direct outcome of the dissipative directional changes of $\mathbf{H}_K$ and not to reflect on the symmetry of the anisotropy.

Most of our findings come out of the unique capability of magnetization-vector measurements. Such measurements have recently been used in studying random-anisotropy systems[18] and ferromagnetic crystals[19]—but, to our knowledge, the work reported here (and briefly elsewhere[12]) represents the first application of magnetization-vector measurements to SG problems.

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