Rotation of anisotropy in a $Ni₇₆Mn₂₄$ spin-glass alloy

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Measurements of the longitudinal and transverse magnetization components (M_L, M_T) were performed on a field-cooled thin-disk-like sample of disordered $Ni₆Mn₂₄$ as it was rotated in various fixed fields. Our experimental data were presented at low temperatures which are well below the spin-glass freezing temperature (\approx 116 K). The observed rotational behavior of the anisotropy is consistent with neither a purely directional character nor a unidirectional and uniaxial character in the assumption of a rigid rotation. We also take into account the possible incoherent domain rotations. Such a possibility also does not seem solely to explain this rotational behavior. However, the data are well described by the fact that the unidirectional anisotropy is not rigidly linked to the lattice along its initial direction, but it can be elastically rotated by the applied field. These experimental results are also in good agreement with a "domain anisotropy model" suggesting a multidomain configuration even in a field-cooled state, which affects the rotation of the anisotropy.

INTRODUCTION

From the time that $Kouvel^{1,2}$ first observed that at low temperature [well below the spin-glass (SG) freezing temperature T_f] the field-cooled disordered alloys of Mn in Ag, Cu, or Ni exhibit a predominantly unidirectional anisotropy, as manifested in narrow displaced M -versus- H hysteresis loop, there has been considerable interest in the intrinsic magnetic properties of such a system in which the unidirectional anisotropy is created during cooling the sample to low temperatures. The existence of the induced unidirectional macroscopic anisotropy in a fieldcooled SG system has also been demonstrated by several experimental techniques including hysteresis (Refs. 1, 3, and 4}, transverse susceptibility (Refs. 5 and 6), torque (Refs. 7 and 8), NMR (Ref. 9), and ESR (Refs. 10, 11, and 12) measurements. The vector model was introduced by many authors^{$1-12$} to interpret the observed ESR and dc magnetization for the SG system under the condition when the applied field H was parallel to the cooling field direction. This model has been based on the rigid rotation of the anisotropy, i.e., the unidirectional anisotropy H_A is considered to be rigidly linked to the lattice. But, the new alternative model of the so-called triad model, as opposed to a rigid rotation, was developed by Saslow, 13 Henley, Sompolinisky, and Halperin, ¹⁴ and Halperin and Saslow¹⁵ to describe the dynamic behavior of the anisotropy. This model implies that the unidirectional anisotropy is to be changed in orientation during rotation of the field-cooled sample in an applied field, corresponding to what has been called a "redefinition" of the anisotropy. Recently, we have first shown¹⁶ that the unidirectional anisotropy is not rigidly linked to the lattice along its initial direction but it can be elastically rotated by the applied field. This result is consistent with the idea of the plied field. This result is consistent with the idea of the domain-anisotropy model. ^{17, 18} This claim has been supported by Sato¹⁹ performing transverse ac susceptibility measurements on the $Ni_{77}Mn_{23}$ alloy. However, the Chicago group has shown^{20–22} that the anisotropy rotate rigidly with the sample for all the applied field at lower

temperatures, but, at sufficiently high field, becomes irreversibly (dissipatively} relative to the sample toward the direction of the applied field H by means of rotational magnetization-vector measurements (RVM). By this technique the magnetization parallel, as well as perpendicular, to H has been measured simultaneously. These measurements, therefore, enable us to describe an angular behavior of the unidirectional anisotropy. This technique is discussed in several reports in detail. $20 - 22$ Using the same technique, we have seen that the anisotropy rotates elastically at lower temperatures (provided that the temperature is much less than the spin-glass temperature T_f), as opposed to the claim of some authors. $20 - 22$

We have also discussed the intrinsic nature of the anisotropy created during cooling the sample, considering that it has only either the pure unidirectional character or both the unidirectional and uniaxial character, in comparison to the theoretical results for each case with the experimental data.

EXPERIMENTAL TECHNIQUE

The measurements were performed on a polycrystalline $Ni₇₆Mn₂₄$ disordered alloy. The sample was prepared as reported by our earlier studies¹⁶ whose shape was disklike, 5 mm in diameter and 0.5 mm thick. It was annealed at 850'C for several hours and quenched in water. Our ac susceptibility measurements indicate a freezing temperature of about 116 K and a Curie temperature of about 240 K. Magnetization measurements were done with the modified vibrating-sample magnetometer whose sensitivity is about 10^{-5} emu. For such a study we have mounted two identical sets of pickup coils in geometric quadrature about the cryostat tail. Two coil sets allow us to measure simultaneously the component parallel (M_L) , and perpendicular (M_T) to the applied field H in the sample disk plane. For this geometry, the demagnetization effects are very small. A magnetic field was provided by Electromagnet (Varian-Model 3800) up to 23 kG. The temperature was controlled by a helium flow cryostat

FIG. 1. Magnetic vector diagram for rotational measurements on the sample in a high field, which saturates thermoremanence and then rotates by angle θ relative to fixed external field.

(Oxford Instruments) from 4.2 to 300 K. The experimental situation is represented schematically in the vector diagram of Fig. 1, which also shows M_L and M_T , the components of M parallel and perpendicular to H , respectively. We assume that the unidirectional anisotropy rotates and its final direction relative to H is given by θ_K . When the sample is rotated by angle θ relative to H , the vector magnetization will be oriented along the direction of the effective field H_{eff} , where ϕ is the orientational angle of M_S relative to H.

RESULTS AND DISCUSSION

Figure 2 shows the magnetization of this sample as a function of the applied field after cooling the sample to 5 K in a magnetic field of about 10 kOe. Then the sample was warmed up to 9 K; at this temperature the plot of M versus H is also shown in Fig. 2. The total magnetization M in the cooled field state can be represented by the linear equation $M = M_S + X(T)H$, where M_S is the thermoremanent magnetization and $X(T)$ is the reversible susceptibility determined from outside the hysteresis loop. The values of X (0.2 emu/g kOe and 0.4 emu/g kOe for $T = 5$ and 9 K, respectively) and the anisotropy fields are needed for the analysis of the rotational magnetization data. The anisotropy fields can be deduced from the hysteresis loop where the center of the loop is calculated, which gives the unidirectional anisotropy H_A (550 Oe and 375 Oe for $T = 5$ K and $T = 9$ K, respectively). The width of the hysteresis loop is determined twice by the uniaxial anisotropy H_K (80 Oe and 190 Oe for $T=5$ K and $T=9$ K, respectively). In fact, it is still open to discussion whether the anisotropy for such a system is purely unidirectional or not. We therefore assume that either the anisotropy is purely directional, whose value will be taken to be the sum of H_A and H_K , or consisting of both the unidirectional component H_A and the uniaxial component H_K . There is another possible case in which the hysteresis effect is only governed by a possible incoherent domain (or cluster) rotation. This case will be discussed below.

In RMV measurements, the sample was initially cooled

FIG. 2. Magnetization-field hysteresis loop for $Ni_{76}Mn_{24}$ in a field-cooled state (a) at 4.2 K and (b) at 9 K.

to 5 K in 10 kOe. The field was then reduced to a measuring field of H , in which the sample was rotated with the 10 steps starting from $\theta = 0^{\circ}$ up to 180° and was brought back to its original position, using the same steps. At each step, the longitudinal (M_L) and transverse magnetization (M_T) were measured simultaneously, each of the M_L values were corrected for a small isotropic contribution linear in H (corrected M_L =measured M_L $-XH$). The measuring field H was indicated in the figures (see Fig. 3). Here we have deliberately selected the values of the measuring fields H such that one of them is less than, the other one is higher than, and the third one is close to the value of the unidirectional anisotropy value for each temperature ($T = 5$ and 9 K). The values of M_T and the corrected M_L were then combined to give its orientational angle ϕ relative to H (see Figs. 4 and 5). We note that the rotational hysteresis is relatively large for the measuring field being close to the unidirectional anisotropy. Our results at 9 K are clearly similar to those of $T = 5$ K, except the former generally exhibits relatively large hysteresis.

Regarding the variation of ϕ for the lowest measuring field, ϕ rises almost linearly up to 150° for both temperatures. The experimental curves for $H \ge H_A$ starts out linearly at low θ and then drops from a maximum to a low but nonzero ϕ at θ = 180° and then returns with little hysteresis as shown in Figs. 4 and 5. As can be seen from these figures the curve for the highest field starts with relatively lower values of slope. It should be noted that these nonzero ϕ values for 9 K are always higher than those of ⁵ K. Both are similar except hysteresis for the

FIG. 3. For $Ni_{76}Mn_{24}$ at $T=5$ and 9 K, longitudinal and transverse magnetization (M_L, M_T) vs sample rotation angle (θ) for different applied fields, where closed (open) symbols are for increasing (decreasing) θ .

highest field is smaller.

So far, we have exhibited the experimental data. Now, we want to give the theoretical results. We consider that the unidirectional anisotropy field rotates rigidly with the sample. When the sample is rotated by an angle θ relative to H external field, H_A and H_K exert the torques on
the vector magnetization M_S . Thus the balancing torque equation is given by

$$
H_A \sin(\theta - \phi) + H_K \cos(\theta - \phi) \sin(\theta - \phi) = H \sin \phi \tag{1}
$$

This equation is equivalent to the condition that M_S lies parallel to the total effective field H_{eff} as shown in Fig. 1. Substituting our measured value of H_A and H_K , into Eq. (1) we have computed the values of ϕ for different H and θ . We also consider two different cases; one of them corresponds to the case in which the anisotropy field is assumed purely directional, i.e., the total unidirectional anisotropy field is the sum of H_A and H_K , the other case in which the sample has both the unidirectional and uniaxial components. On the other hand, one might expect small incoherent domain (or cluster) rotation effects to occur in our sample during the sample rotation in a field.

The rotational hysteresis (though small) may also be attributed to possible incoherent domain rotations, which could cause the hysteresis loop to be much wider. Therefore, it seems quite natural to invoke this effect. Such a possibility should be included in the calculation, considering that the width of a displaced hysteresis loop does not necessarily evidence a uniaxial component of the spin-glass anisotropy. For simplicity, we assume that the hysteresis loop is only governed by the unidirectional anisotropy H_A and the incoherent domain rotations $(H_K=0)$. The calculated results of each case for $T=5$ and 9 K are given in Figs. 4 and 5. (Each case is indicated in the figures.) As can be seen in the figures, for the lowest field, the experimental results are slightly below the corresponding computed result. The deviation from each other is much higher at around $\theta = 90^{\circ}$ than that of lower and higher values of θ . For the fields $H \geq H_A$, the separation from each other is more remarkable. The upper data points belong to the pure-directional case. The experimental data lie markedly below the calculated data for all cases. Similar disagreement was also observed in the earlier experiments made on $Ni_{75}Mn_{25}$ alloy.²⁰ This discrepancy leads us to suggest that the uni-

directional anisotropy rotates elastically as opposed to a claim of a rigid rotation by some authors. 20^{-2} In order to be sure about the elastic anisotropy rotation, we were forced to repeat the following experiment introduced first by the same group. In the experiment, the field-cooled sample was rotated by 180 \degree relative to different H (in our experiment, the maximum H was about 23 kOe) and then H was removed. Subsequently the parallel (M_L) and perpendicular (M_T) components of M were measured simultaneously. Our experiments for both $T=5$ and 9 K showed that the perpendicular components M_T were almost zero up to 23 kOe. It means that the unidirectional

FIG. 4. ϕ (orientational angle of M_S) versus θ (sample rotation angle) of field-cooled $Ni_{76}Mn_{24}$ in various H at 5 K. Dashed curves of ϕ vs θ are for decreasing θ and show some rotational hysteresis. Symbols show calculated rotational behavior from Eq. (1) for unidirectional anisotropy (UD) and both unidirectional and uniaxial anisotropies (UA). Broken line (IR) represents the case in which domains rotate incoherently.

anisotropy field is again directed toward its initial cooled direction. This experimental fact can be attributed to one of the two possible cases as follows: The anisotropy was rotated rigidly with the sample as proposed by the same researcher or the anisotropy first rotates elastically in the field H and then comes to its initial direction after removing the external field. However, this research group has overlooked the latter case. According to our experiments, the latter case seems to be more appropriate to interpret the experimental behavior of ϕ with respect to θ . If the former case had occurred, the values of ϕ for different H and θ obtained from the experiments and the balancing torque equation would have been in agreement with each other. It is obvious that if the anisotropy field H_A rotates elastically from its initial direction, the values of ϕ will be less than those deduced from the balancing equation based on the rigid anisotropy rotation. This behavior will be discussed below in detail.

The observed hysteresis in the magnetization components (M_L, M_T) may be interpreted in terms of the anisotropy rotations. We believe that the multidomain structure still persists even in the field-cooled state. Suppose that a number of domains are distributed radially along the directions limited to the area around the fieldcooled direction. Each domain has a unidirectional anisotropy field initially directed along its magnetization vector M_S . When the sample is rotated by θ , each magnetization vector will restabilize due to counterbalancing torques exerted on it by the anisotropy field H_A and the

FIG. 5. ϕ (orientational angle of M_S) vs θ (sample rotation angle) at 9 K. Symbols are represented in Fig. 4. Note that symbols used in Figs. 4 and 5 represent the same cases.

ature T_f .

applied field H, while the anisotropy fields H_A tend to rotate toward the field direction as long as the Dzyaloshinsky-Moriya torque, exerted on M_S by the anisotropy field H_A , is balanced by the torque due to spinlattice coupling. The total change in the anisotropy field directions takes place gradually over N steps $(N \text{ is sup-}$ posed to be a very large number) during the sample rotations. If one assumes that the directional change in each step is proportional to the product $M_S H_A$ sin α or β sin α , where α is the angle between H_A and M_S , β is proportionally constant as a characteristic of the sample, one should observe a difference in the domain structure for the forward and reverse direction of the rotation as it passes from the same θ angle. This difference becomes more remarkable for $\alpha=90^\circ$ occurring at angles of θ greater than 90'. This is exactly what we observe in our experiment. This picture of the anisotropy field distribution has been discussed in detail by means of ESR measurements on the NiMn alloy in our recent work.¹⁶

To conclude the discussion, in view of the overall

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agreement, the domain-anisotropy model^{17,18} involvin the elastic anisotropy rotation appears to account for the observed rotational behavior at sufficiently low temperatures. The unidirectional anisotropy rotates elastically as opposed to the claimed rigid rotation²⁰⁻²² as long as the measuring temperature is well below the freezing temper-

In order to gain further insight on the dynamic behavior of the unidirectional anisotropy field H_A , it is necessary to determine the time effect of the magnetization components (M_L, M_T) separately. We intend to study them further in this alloy and other SG alloy sys-

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tems under the conditions describe above.

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