Induced anisotropy in reentrant $Ni_{77}Mn_{23}$ studied by transverse ac susceptibility

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The transverse ac susceptibility of $Ni_{77}Mn_{23}$ was measured in a dc field to analyze the field-dependent anisotropy in the mixed spin-glass–ferromagnetic state. The results support a picture that the field-induced Dzyaloshinsky-Moriya-type anisotropy field can be elastically rotated by an applied dc field. The uniaxial field and the exchange field of coupling between adjacent domains are also estimated at 4.2 K. The procedure of rotating a dc field in a plane perpendicular to the ac field gave evidence indicating that there is multidomain configuration even in a dc field almost sufficient to saturate magnetization, which affects the rotation of the anisotropy. A plausible picture for motion of the anisotropy is proposed in connection with the domain configuration.

Field-induced anisotropy in a low-temperature spin-glass–ferromagnetic (SG-F) mixed state of reentrant Ni-Mn alloys has been intensively investigated through measurements of magnetization,[1–5] magnetoresistance,[6] and electron-spin resonance (ESR).[7–9] Aspects in the magnetization reversal have been consistent with a macroscopic unidirectional nature in the anisotropy.[1] Recent ESR studies proposed some evidence indicating 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.[8,9] This result is consistent with the idea of the domain-anisotropy model.[3,6,10] However, information has been scarce about rotation of the induced anisotropy in the SG-F mixed state with applied field except for ESR studies.

One of the most effective and direct experimental methods for characterizing the magnetic anisotropy is to measure transverse ac susceptibility in a dc field. The unidirectional and uniaxial terms in the anisotropy field in pure spin-glass CuMn have been evaluated by measuring the transverse ac susceptibility.[11–14] The former term agreed well with that determined from nuclear magnetic resonance (NMR), but there is significant discrepancy between the latter term and that determined from magnetization data, which can be attributed to inadequate application of the single domain picture of anisotropy energy to practical multidomain configuration in the sample.[11,13] On the other hand, both terms of anisotropy in the SG-F mixed state must be well evaluated from the transverse ac susceptibility since the magnetization is easily saturated using conventional apparatus. There is further merit in characterizing the anisotropy using ac susceptibility measurements, namely, the anisotropy field can be determined as a function of applied field. However, there has been no reported transverse ac susceptibility in the SG-F mixed state of reentrant Ni-Mn alloys. In the present work, the ac susceptibility of reentrant $Ni_{77}Mn_{23}$ was measured at 4.2 K in the dc field perpendicular to the ac field and the induced anisotropy field was obtained as a function of the dc field. I will also discuss the uniaxial term and the exchange field of coupling between adjacent domains, and finally propose a plausible picture for motion of the anisotropy in connection with domain configuration in the mixed SG-F state.

The $Ni_{77}Mn_{23}$ alloy was prepared as reported by Abdul-Razzaq and Kouvel.[15] The resultant alloy sample was shaped as a sphere 1.3 mm in diameter. The spin-freezing temperature T_f and the Curie temperature T_C agreed well with those in Ref. 15. The ac susceptibility measurement (frequency 210 Hz and field 4.5 Oe) was performed as described in the previous paper.[16] The accuracy of absolute value of the measured ac susceptibility was about 5%, limited by deviation of the sample position. The dc field is rotated by an angle θ_H in the y-z plane perpendicular to the ac field as described in Fig. 1, where the field-cooled (FC) procedure was performed at $H_{cool}=3400$ Oe with $\theta_H=0$.

The ac susceptibility in the FC state was obtained at 4.2 K as a function of the internal dc field H_{in} with $\theta_H=0$ (Fig. 2). There are some points of similarity between the ac susceptibility and magnetization curve (see inset in Fig. 2), i.e., an irreversible behavior accompanied with magnetization reversal and a perfect reversibility at high field.

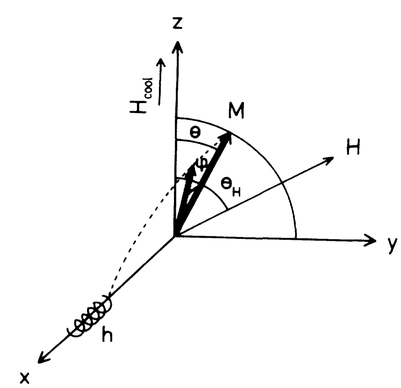

FIG. 1. Geometry of transverse ac susceptibility measurements. When the dc field H rotates away from the direction of field cooling (z direction) by an angle θ_H in the y-z plane perpendicular to the ac field h, the magnetization rotates by an angle θ in the same plane. The angle ϕ is defined in the x-M plane.

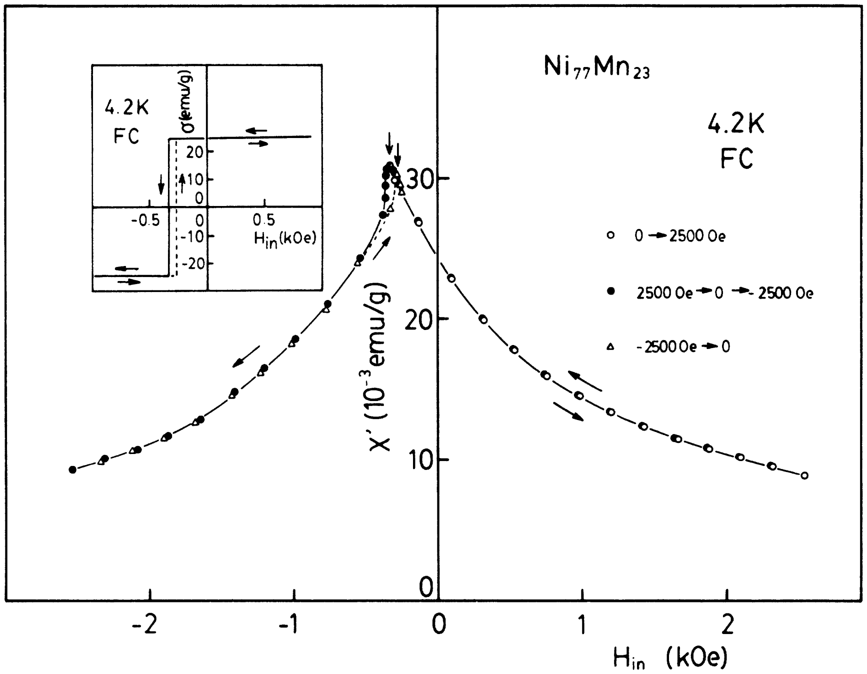

FIG. 2. ac susceptibility measured at 4.2 K in the field-cooled state of $Ni_{77}Mn_{23}$ as a function of the internal field H_{in}. Data were obtained at fields between -2.5 and 2.5 kOe. The inset shows the magnetization curve of the same sample where an irreversibility centered at 300 Oe is observed.

This similarity has also been observed in pure spin-glass Cu-Mn.[13] Then, the following assumptions are made to give an expression for transverse ac susceptibility in the mixed SG-F state: (1) The saturation magnetization M_s takes the place of the induced magnetization in the FC state in pure spin glass.[11] (2) The isotropic ac susceptibility independent of macroscopic magnetization is negligibly small. (3) There is the exchange energy E_{ex} of coupling between adjacent domains, resulting in the effective exchange field H_{ex} which should depend on the angle θ through the domain configuration. (4) The field-induced anisotropy has triad symmetry and the θ dependence of the anisotropy field can be calculated based on the Dzyaloshinsky-Moriya (DM) Hamiltonian.[17]

The total energy for rigid rotation of the spin system would be written as

$$E = \tfrac{1}{2}K_d(3-\cos\theta-\cos\phi-\cos\theta\cos\phi) + \tfrac{1}{2}K_a(\sin^2\theta\cos\phi-\cos\phi) - M_sH_{in}\cos(\theta_H-\theta)\cos\phi+E_{ex}(\theta,\phi)\ , \qquad (1)$$

where ϕ is the angle in the x-M plane as shown in Fig. 1, and the first and second terms correspond to the DM-type and uniaxial terms, respectively. The ac susceptibility with $\theta_H=\theta=0$ for positive and negative internal field enough to saturate almost magnetization would be given separately based on Eq. (1) as follows:

$$\chi'_+ = M_s/[H_d+H_a+H_{in}+H_{ex}(\theta=0)]\ , \qquad (2a)$$

$$\chi'_- = M_s/[-H_d+H_a+H_{in}+H_{ex}(\theta=0)]\ , \qquad (2b)$$

where H_d and H_a correspond to the unidirectional and uniaxial anisotropy fields, respectively. Figure 3 shows reciprocals of χ'_+ and χ'_- as a function of H_{in}, where the data in the region with irreversibility are left out. The reciprocal of χ'_+ has linear dependence on H_{in} except for several low-field data, and the slope corresponds to $M_s=36.7$ emu/g which is larger than the measured magnetization. If one notes the frustrated nature of spins originated from spin-glass interaction and the difficulty of

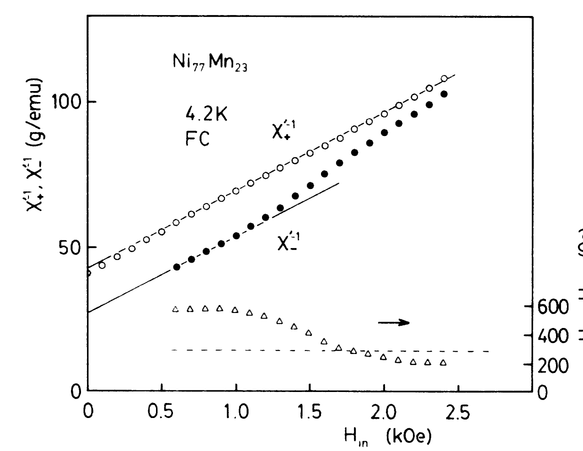

FIG. 3. The reciprocals of ac susceptibilities of $Ni_{77}Mn_{23}$ at angle $\theta_H=0$ for positive and negative fields $\chi_+'^{-1}$ and $\chi_-'^{-1}$ as a function of the absolute value of internal field. The sum of the DM-type anisotropy field $H_{d+}+H_{d-}$ is also shown, which is estimated from the ac susceptibility data at internal fields below 1 kOe where $\chi_-'^{-1}$ shows linear relation with H_{in}. Sign of H_{d-} reverses at the field $H_{in}=1.7$ kOe.

uniting domains into a single one even in a quenched ferromagnetic sample,[18] however, the value of M_s is not inconsistent with the magnetization data. The reciprocal of χ'_- also shows the linear relation with equal slope to that of χ'_+ at fields below 1 kOe, while χ'^{-1}_- starts to approach χ'^{-1}_+ with increasing H_{in} for $H_{in} > 1$ kOe. The lower field data on χ'^{-1}_+ and χ'^{-1}_- yield $H_d = 290$ Oe based on Eqs. (2a) and (2b), which agrees well with the unidirectional anisotropy field estimated from the magnetization reversal (see inset in Fig. 2). On the other hand, the high-field behavior in χ'^{-1}_- must correspond to decrease in the effective component of the DM-type anisotropy field due to the rotation of the anisotropy with high negative field. The sum of the DM-type anisotropy fields H_{d+} and H_{d-} determined from χ'_+ and χ'_- is also shown in Fig. 3. The field dependence is perfectly reversible at fields with $H_{in} > 0.5$ kOe when the field is applied up to $H_{in} = 2.5$ kOe and then reversed, and the value of H_{d-} becomes negative at fields above 1.7 kOe. These results are consistent with the picture for the unidirectional anisotropy estimated from ESR studies.[8,9] Then, I claim that the detailed behavior of the anisotropy with applied field and the reversible motion consistent with the assumption of the elastic rotation are well characterized by the transverse ac susceptibility measurements.

An evaluation of the contribution from the other terms in ac susceptibility is attempted. On the basis of Eqs. (2a) and (2b), $H_a + H_{ex}(\theta = 0) = 1250$ Oe is obtained. The irreversible feature in ac susceptibility reflecting the uniaxial term gives $H_a = 40$ Oe as well as the magnetization curve. The exchange field at $\theta = 0$ is finally given as $H_{ex}(\theta = 0) = 1210$ Oe. On the other hand, H_{ex} for zero-field-cooled state can be also estimated based on the SG-F domain model[19] as follows:

$$\chi'_{ZFC}(H_{in} = 0) = (2M_s/3)/(H_k + 2H_{ex}) , \quad (3)$$

where $\chi'_{ZFC}(H_{in} = 0)$ is the zero-field-cooled (ZFC) ac susceptibility at $H_{in} = 0$, and $H_k = H_d + H_a$. The measured $\chi'_{ZFC}(H_{in} = 0)$ is 13.6×10^{-3} emu/g at 4.2 K, which yields $H_{ex} = 740$ Oe. This difference between both the values of H_{ex} suggests that a preferential domain configuration is arranged in the FC process so that the average exchange field becomes larger than that of random configuration in the ZFC state.

The ac susceptibility was measured as a function of the angle θ_H to evaluate these estimated values. Figure 4 shows the experimental results after cooling down to 4.2 K in the external field and using a field of $H_{ex} = 1.6$ kOe to rotate magnetization. Assuming that E_{ex} is independent of the angle θ, the relation between θ and θ_H can be given by the condition $\partial E/\partial \theta = 0$ based on Eq. (1) as follows:

$$K_d \sin\theta + K_a \sin\theta \cos\theta - M_s H_{in} \sin(\theta_H - \theta) = 0 . \quad (4)$$

Using equilibrium values of θ, the transverse ac susceptibility would be written as

$$\chi' = \frac{M_s}{H_d(1+\cos\theta)/2 + H_a \cos^2\theta + H_{in}\cos(\theta_H - \theta) + H_{ex}} , \quad (5)$$

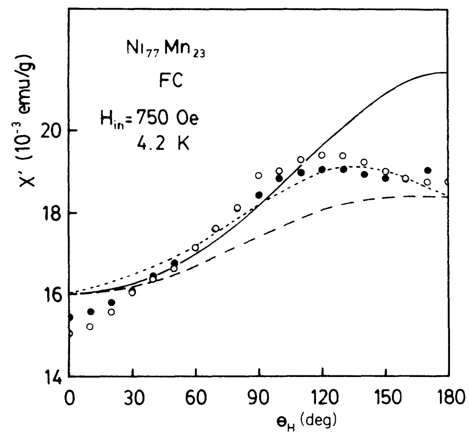

FIG. 4. ac susceptibility in the field-cooled (FC) state of $Ni_{77}Mn_{23}$ in the field $H_{in} = 750$ Oe at 4.2 K as a function of the angle θ_H. The calculated curves are shown based on Eq. (4) in the following case: (1) with the conventional unidirectional anisotropy and the constant H_{ex} (solid line), (2) with the DM-type anisotropy and the constant H_{ex} (dashed line), and (3) with the DM-type anisotropy and $H_{ex}(\theta)$ using Eq. (5) (dotted line). Open circle shows rotation from 0° to 180° and closed circle shows subsequent rotation from 180° and 0°.

where the θ dependence is obtained by calculating the effective field along the magnetization M based on the ϕ dependence in Eq. (1). The calculated curve based on Eq. (5) shows better quantitative agreement with the measured values in comparison with that assuming a conventional unidirectional anisotropy (see Fig. 4). This feature is also reflected in difference between χ'_- and χ' at $\theta_H = 180°$ in the same internal field. There is still significant disagreement between the measured data and the calculated curve with triad symmetry, however, which must be attributed to the inadequate assumption for $H_{ex}(\theta)$. It is difficult to estimate θ dependence of H_{ex}, since there is little information about the domain configuration in the FC state. Then, I can tentatively obtain a calculated curve which gives better agreement with the measured data as shown in Fig. 4, using the following expression:

$$H_{ex}(\theta) = 1110 + 100(\theta/90 - 1)^2 \quad (6)$$

in Oe. These results suggest a significant role of domain configuration even in a field almost sufficient to saturate magnetization, i.e., there are several domains with magnetization practically parallel to the applied field under the present experimental condition.

Based on above discussion, I can propose a plausible picture of the mixed SG-F state of reentrant $Ni_{77}Mn_{23}$ as follows: (1) There are randomly distributed domains in the ZFC state, in which the anisotropy field lies along the domain magnetization. (2) At zero internal field multidomain configuration with a preferential orientation of the anisotropy field is arranged in the FC state. (3) When the internal field, which is almost sufficient to saturate magnetization, is applied with an angle of θ_H there are still several domains in which the magnetization are practically rotated to the direction of applied field, and then the anisotropy field rotates away from the direction of field cooling to that of the applied field.

In conclusion, my experimental results of the transverse ac susceptibility of $Ni_{77}Mn_{23}$ support the picture that the field induced DM-type anisotropy can be rotated elastically by applied field. Further, I obtained evidence that domain configuration plays a significant role in motion of the anisotropy even in a field which is almost enough to saturate magnetization. A plausible picture for the motion in anisotropy is proposed in connection with the domain configuration.

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[1]J. S. Kouvel and C. D. Graham, J. Phys. Chem. Solids **11**, 220 (1959).

[2]W. Abdul-Razzaq and J. S. Kouvel, J. Appl. Phys. **55**, 1623 (1984).

[3]J. S. Kouvel, W. Abdul-Razzaq, and Kh. Zip, Phys. Rev. B **35**, 1768 (1987).

[4]S. Senoussi, Phys. Rev. Lett. **51**, 2218 (1983).

[5]S. Senoussi, J. Phys. (Paris) **45**, 315 (1984).

[6]S. Senoussi, Phys. Rev. Lett. **56**, 2314 (1986).

[7]S. Schultz, E. M. Gullikson, D. R. Fredkin, and M. Tovar, Phys. Rev. Lett. **45**, 1508 (1980).

[8]Y. Oner, B. Aktas, F. Apaydin, and E. A. Harris, Phys. Rev. B **37**, 5866 (1988).

[9]B. Aktas, Y. Oner, and E. A. Harris, Phys. Rev. B **39**, 528 (1989).

[10]S. Senoussi and Y. Oner, J. Phys. (Paris) **46**, 1435 (1985).

[11]H. Alloul and F. Hippert, J. Phys. (Paris) Lett. **41**, L201 (1980).

[12]F. Hippert, H. Alloul, and F. Fert, J. Appl. Phys. **53**, 7702 (1982).

[13]F. Hippert and H. Alloul, J. Phys. (Paris) **43**, 691 (1982).

[14]H. Alloul and F. Hippert, J. Magn. Magn. Mater. **31-34**, 1321 (1983).

[15]W. Abdul-Razzaq and J. S. Kouvel, Phys. Rev. B **35**, 1764 (1987).

[16]T. Nemoto, T. Sato, E. Ohta, and M. Sakata, J. Magn. Magn. Mater. **78**, 43 (1989).

[17]A. Fert, D. Arvanitis, and F. Hippert, J. Appl. Phys. **55**, 1640 (1984).

[18]S. Mitsuda and Y. Endoh, J. Phys. Soc. Jpn. **54**, 1570 (1985).

[19]S. Kouvel and W. Abdul-Razzaq, J. Magn. Magn. Mater. **53**, 139 (1985).