Ferromagnetic resonance study of $Ni_{79}Mn_{21}$ alloy

Y. Öner, B. Aktaş, and F. Apaydin
Department of Physics, Hacettepe University, Beytepe, Ankara, Turkey

E. A. Harris
Department of Physics, University of Sheffield, Sheffield S3 7RH, United Kingdom
(Received 24 June 1987; revised manuscript received 18 September 1987)

In this report, we present ferromagnetic-resonance (FMR) measurements on the reentrant $Ni_{79}Mn_{21}$ alloy as a function of temperature (4.2 K $< T <$ 300 K), after cooling the sample through the spin-glass–ferromagnetic transition either in zero field (ZFC) or in high field (FC). The FMR spectra were taken with the applied magnetic field both parallel and perpendicular to the surface of the thin disklike sample. The unidirectional anisotropy has been obtained by analyzing the FMR data. The anisotropy effects appear at a much higher temperature than the spin freezing temperature, T_f ($\sim$35 K). In addition to the usual FMR signal, when the applied magnetic field is parallel to the cooling field direction, another resonance signal is observed at lower magnetic field. However, this signal disappears completely when the field is reversed. We interpreted this signal to be due to a new microscopic anisotropy created during the cooling.

The Ni-Mn system has continued to play a prototypal role in various models concerning the unusual properties of the reentrant systems. They show some characteristic properties such as displaced hysteresis loops, frustration effect, double magnetic transition, canting transition, and so on.[1-3] As it is well known, these properties arise from the competing ferromagnetic interactions between Ni-Ni or Ni-Mn atoms and the antiferromagnetic interactions between Mn-Mn atoms.

Although different techniques such as magnetization,[4] ESR,[5] neutron diffraction,[6] and magnetoresistance[7] have been applied to Ni-Mn alloys over a wide range of concentration of Mn, no ferromagnetic-resonance (FMR) study on the $Ni_{79}Mn_{21}$ alloy has yet appeared in detail. In this report, we shall give the results of the FMR measurements obtained with a conventional X-band EPR spectrometer, to gain new insights into spin-glass state (SG) of Ni-Mn alloys and their dynamical characterization at different temperatures in the range 4.2–300 K.

The samples were prepared by melting together high-purity constituents in an rf furnace. To ensure a good homogenization, each alloy was melted at least three times and was homogenized by cold working. A disklike sample of 3 mm diameter and 0.1 mm thickness was used in the FMR measurements. This sample was encapsulated in a quartz tube (in vacuum) annealed 3 h at 900 °C and water quenched.

FMR studies were performed with a Varian Associate reflection spectrometer at fixed frequency ($\sim$9.25 GHz). The sample was attached to a thin quartz plate mounted on a goniometer, so that it could be rotated relative to the applied magnetic field in the cavity in which microwave and dc fields were orthogonal. A standard field modulation (100 kHz) and phase-sensitive detector techniques were used, so that detected signal corresponded to the field derivative of the absorbed power. The temperature was controlled by a helium flow cryostat from 4.2 to 300 K. The sample was cooled down to 4.2 K either in the presence of the external magnetic field of 17 kOe (FC) or in zero field (ZFC), and then warmed up to the measurement temperature. In order to avoid the effect of the remanent field of the magnet in the ZFC case, the surface of the disklike sample was chosen parallel to the surfaces of the poles of the magnet.

The FMR spectra obtained with the applied magnetic field parallel and perpendicular to the surface of the disklike sample are shown in Figs. 1 and 2, respectively. The figures show that all spectra, except those obtained for parallel geometry in the FC case, have two main peaks in addition to the diphenylpicrylhydrazyl (DPPH) signal which we used as a g marker. One of these peaks appears at higher fields and corresponds to the expected FMR collective mode. The other one which appeared at relatively low fields is unusual and was examined further. We obtained FMR spectra with the applied field both parallel and antiparallel to the cooling field direction for perpendicular geometry. Examples of these spectra for some selected temperatures are given in Fig. 3.

Concerning the effect of the demagnetizing fields and the presence of a macroscopic unidirectional anisotropy $H_A(=K/M_s)$, one gets Kittel's well-known formulas for the resonance condition of the thin disklike ferromagnetic sample:

$$\left(\frac{\omega}{\gamma}\right)^2=(H_r+H_A)(H_r+H_A+DM) \quad (1)$$

for the parallel geometry, and

$$\frac{\omega}{\gamma}=H_r+H_A-DM \quad (2)$$

for the perpendicular geometry. Here, M is the magnetization of the sample in the internal field corresponding to the resonance field (H_r), $\gamma(=ge/2mc)$ is the gyromagnetic ratio of the processing moments, and D is the demagnetizing factor for perpendicular geometry.[8]

It is evident that the peaks at higher fields for each geometry correspond to the usual ferromagnetic absorp-

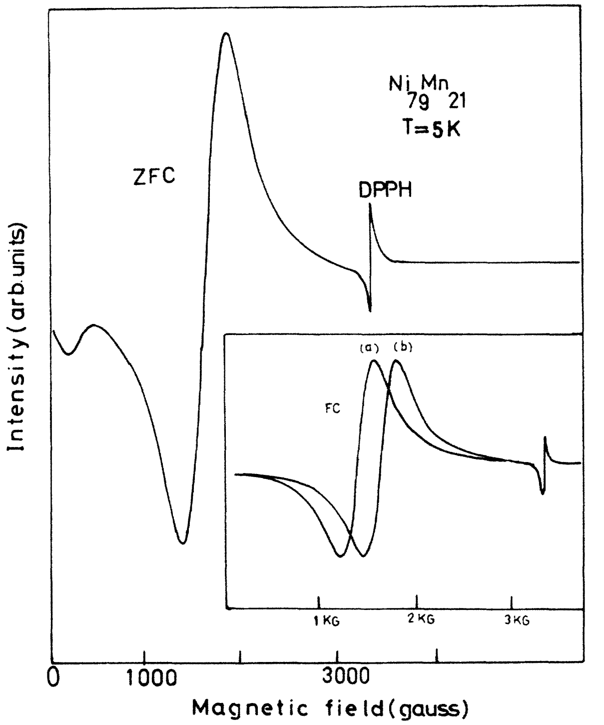

FIG. 1. Field-derivative FMR spectrum of the $Ni_{79}Mn_{21}$ alloy observed after cooling the sample in $H=0$ (ZFC) down to 5 K, for parallel geometry. Inset shows the spectra for the field cooling case (FC). The resonance field of spectrum (b) is shifted to the higher field due to the unidirectional anisotropy when the field is reversed.

tion lines predicted by Kittel's formulas [Eqs. (1) and (2)]. On the other hand, the peaks at lower fields are different from the ferromagnetic absorption lines in every respect; they are relatively smaller in amplitude and broader in shape.

In order to obtain the anisotropy field (H_A) and simultaneous magnetization as a function of temperature, we used Kittel's formulas [Eqs. (1) and (2)] with the data from FC and ZFC independently for parallel and perpendicular geometry. Results are given in Fig. 4. By using the technique of Moorjami *et al.*[9] and analyzing the data obtained above the Curie temperature (270 K), we calculated a value of 2.01 for the g value. It is, however, assumed that this g value is constant for all temperatures.

It is well known that, when some spin glasses and reentrant systems like Ni-Mn alloys are cooled in a magnetic field through the ferromagnetic spin-glass transition temperature, the M vs H curves shift towards negative field. The amount of this displacement is called the unidirectional anisotropy H_A created during the cooling whose direction is parallel to the cooling field. This effect manifests itself as a shift in resonance field in our spectra obtained for FC case as shown in Fig. 1(a).

The behavior of the anisotropy field for FC and ZFC cases may be understood with the *domain anisotropy* model.[10] According to this model, the magnetic state of the reentrant Ni-Mn at low temperature ($T<T_f$) is characterized by frozen, random domains. Each domain has a unidirectional anisotropy field directed along its initial magnetization vector. As long as the internal field (H_{int}) is smaller than the anisotropy field (H_A), the mag-

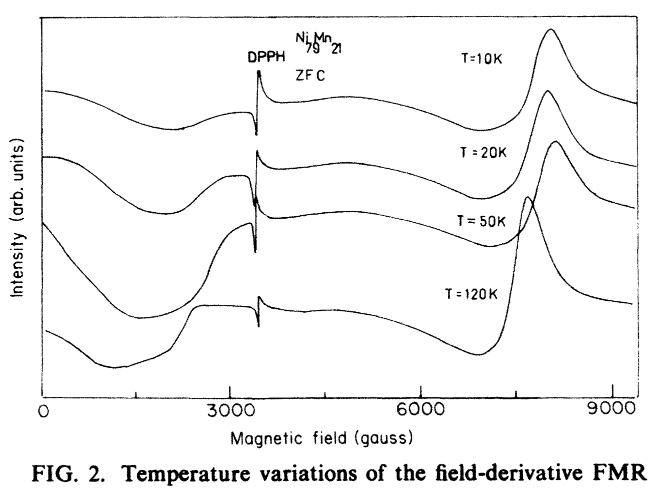

FIG. 2. Temperature variations of the field-derivative FMR spectra of the $Ni_{79}Mn_{21}$ alloy observed after cooling the sample in $H=0$, for perpendicular geometry.

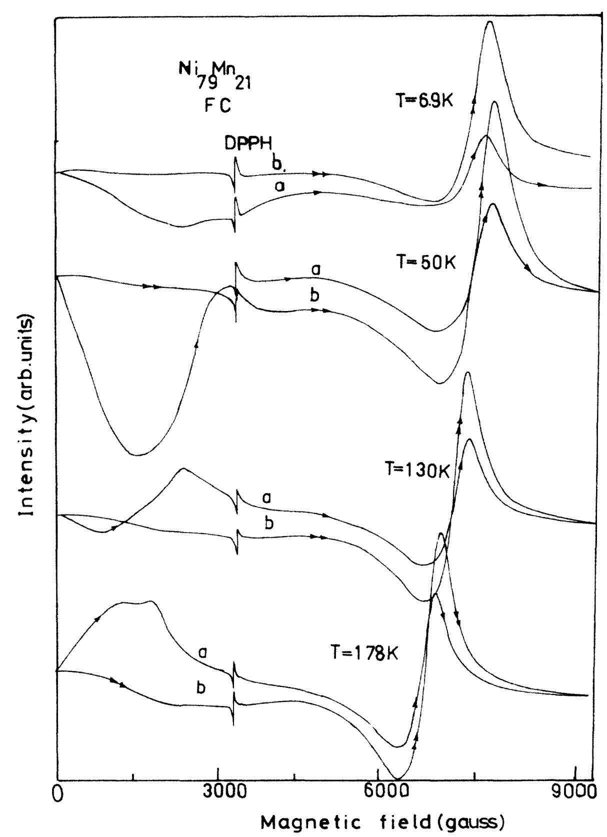

FIG. 3. Field-derivative FMR spectra of the $Ni_{79}Mn_{21}$ alloy observed at different temperatures. The sample was cooled in $H=17$ kG perpendicular to its surface. Applied magnetic field is (a) parallel to the cooling field direction and (b) antiparallel to it. Notice the change in relative intensity of the two peaks between (a) and (b).

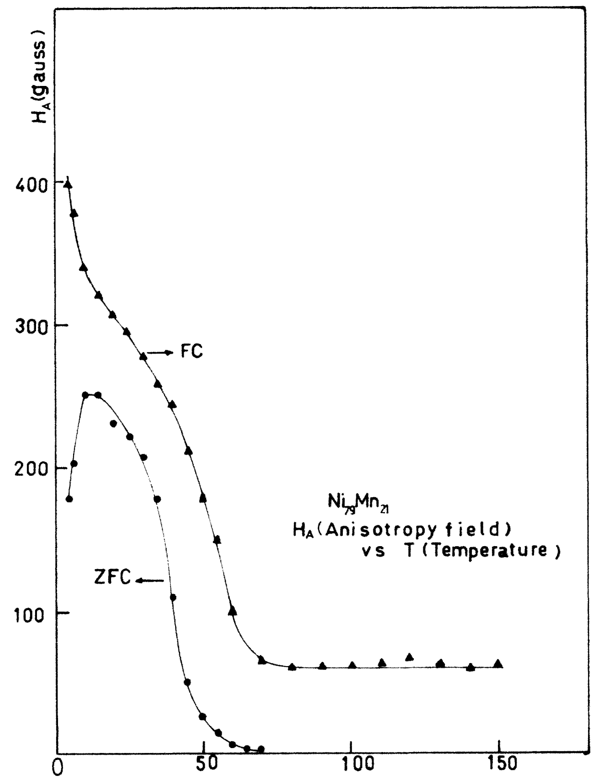

FIG. 4. Temperature dependence of the anisotropy field (H_A) calculated from Kittel's formulas for both ZFC and FC cases.

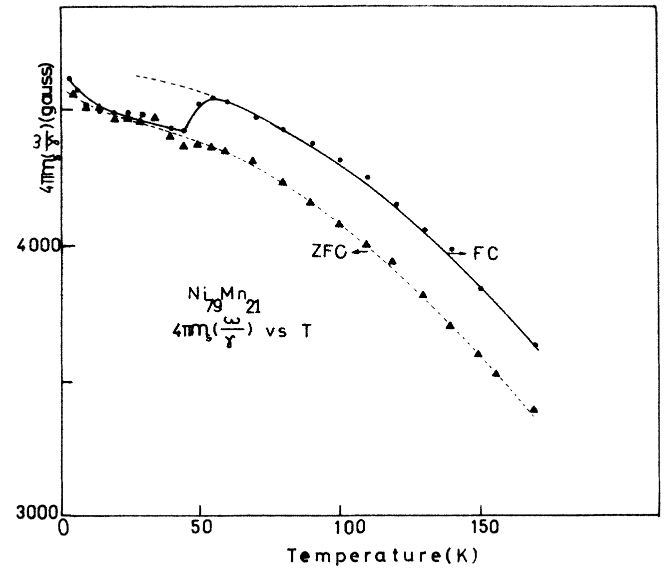

FIG. 5. Temperature dependence of the magnetization obtained from Kittel's formulas (see the text). Note that the magnetization shows a sudden increase at about 50 K for FC case.

netic domains will remain firmly fixed to the lattice.[10] However, when $H_{\rm int}$ reaches the anisotropy field by varying either H or T, the domains will no longer be locked to the lattice and will rotate. On the other hand, because of the gradually reduced coupling between the unidirectional anisotropy field and the lattice, the anisotropy field will start to follow the macroscopic magnetization with increasing temperature, and its amplitude will decrease. Behavior of the unidirectional anisotropy field (H_A) for both ZFC and FC cases are shown in Fig. 4. In the case of ZFC, the anisotropy field (H_A) increases with temperature and reaches its maximum value at 15 K, then decreases with increasing temperature, and eventually goes to zero at about 50 K. This behavior agrees well with that of the magnetic losses against temperature deduced from the magnetization measurements for the same alloy, Senoussi.[11] In the case of FC, the anisotropy field (H_A) which has been already directed along the cooling field decreases with temperature, and above 50 K remains constant at a finite value.

Figure 5 shows the variation of the magnetization obtained from Kittel's formulas [Eqs. (1) and (2)] with increasing temperature. It is clear that there is an abrupt jump of the magnetization at about 50 K, where the anisotropy field H_A goes to zero for ZFC case as shown in Fig. 4. This behavior can be explained as follows. From the measurements of the FC magnetization ($M_{\rm FC}$) in SG systems, we know that $M_{\rm FC}$ is essentially constant. This suggests that the FC state is an equilibrium state. However, Lundgren, Svedlindh, Nordblad, and Beckman[12] showed that, even though $M_{\rm FC}$ may be constant, the FC state may not be an equilibrium state. On the other hand, as Campbell pointed out,[8] when $T < T_f$ and the magnetization is rotated at ESR frequencies, the system remains within one well (in the metastable state) during the effective observation time of 10^{-10} sec. Therefore, the magnetization calculated by Kittel's formulas, will be less than its equilibrium value below T_F. Then as $T \to T_f$, there will be a sudden shift of the distribution of relaxation times $g(\tau)$ to much shorter times due to longer range, cooperative interaction corresponding to the real equilibrium state. Indeed, this is the case that we observed on the FC magnetization as $T \to T_f$. We, therefore, propose 50 K as the freezing temperature instead of 35 K suggested by Senoussi[4] for the same sample.

The unusual resonance line appears for perpendicular geometry, whether it has been cooled in magnetic field or not, because the magnetization does not reach its saturation value. However, it appears for parallel geometry in the case of ZFC only. That is to say, it does not appear for parallel geometry in the case of FC, because the magnetization has already reached its saturation value.

In order to examine the low-field resonance line in detail, we observed the spectra for perpendicular geometry as shown in Fig. 3. These spectra were recorded applying magnetic field either parallel (H_+) or antiparallel (H_-) to the cooling field (H_C) with increasing temperature (4.2 K $< T <$ 200 K). The preliminary results, we obtained, from these curves are (i) at all temperatures, there are two resonance lines for H_+ but there is only one line, the usual collective ferromagnetic mode, for H_-. (ii) For the H_- case, the normal mode (NM) is stronger, but narrower, than that for H_+ case, and its resonance field shifted to the higher field due to the unidirectional anistropy

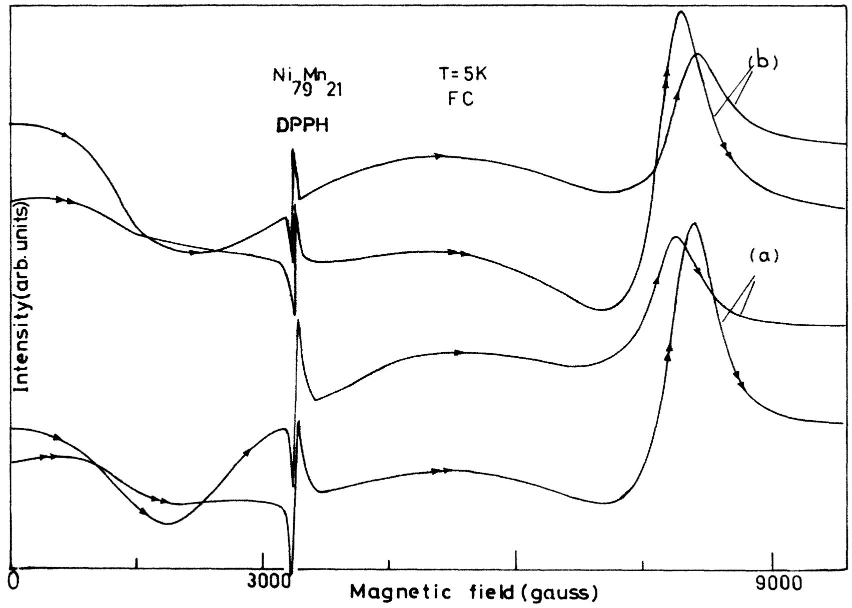

FIG. 6. Lower curves (a) show the FMR spectra of the FC sample observed at 5 K. →, →→ represent applied magnetic field is parallel (H_+) and antiparallel (H_-) to the cooling field, respectively. Upper curves (b) show the FMR spectra of the same FC sample observed at 5 K, but in this case, the sample was warmed up to 300 K and the direction of the cooling field is reversed. →, →→ have the same meaning as (a). Note that it remembers its previous state.

up to 15 K. (iii) For the H_+ case, the low-field lines increase in intensity and pass through a maximum at about 50 K, then gradually become weaker. It must be noticed that the magnetization, the anisotropy, and the absorption line show some changes at 50 K. We therefore suggest that 50 K must be the freezing temperature of $Ni_{79}Mn_{21}$ alloy.

There is another important point explored in Fig. 3. In the case of H_+, the spectra for perpendicular geometry have two resonance lines up to the Curie temperature (270 K). To understand whether this effect would follow the cooling field direction, we reversed the cooling field direction and repeated the experiment. We surprisingly observed that the spectra still remember the first cooling direction, but the resonance field of the normal mode for H parallel to the new cooling field direction shifted to lower field due to the unidirectional anisotropy as shown in Fig. 6. To be sure, FMR experiments were repeated several times choosing different samples taken from the different ingots prepared in our laboratory in one hand, and in the laboratory of Sheffield University (UK), on the other hand. All of these samples and the FMR experiments carried out by one of us (E.A.H.) in Sheffield University gave the same results.

In conclusion, we can say that $Ni_{79}Mn_{21}$ alloy has two kinds of anisotropy. One of them is the well known unidirectional anisotropy. The origin of the other has not been understood yet. In addition to the observable differences of the NM resonance line intensities, the existence of the low-field resonance line might be an important clue to the magnetic nature of the reentrant systems.

This work was supported by both NATO (RG/0789) (Y.Ö. and E.A.H.), and by the Hacettepe University Research Fund (85-10-010-24).

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

[2] S. Senoussi, Phys. Rev. Lett. **51**, 2218 (1983), and references therein.

[3] S. Senoussi and Y. Öner, J. Magn. Magn. Mater. **40**, 12 (1984).

[4] S. Senoussi, Phys. Rev. B **31**, 6086 (1985); R. G. Aitken, T. D. Cheung, and J. S. Kouvel, J. Magn. Magn. Mater. **30**, L1-L4 (1982).

[5] H. Hurdequint, J. S. Kouvel, and P. Monod, J. Appl. Phys. **53**, 2239 (1982); see also H. Hurdequint and J. S. Kouvel, J. Magn. Magn. Mater. **54–57**, 167 (1986).

[6] J. W. Cable and H. R. Child, Phys. Rev. B **10**, 4607 (1974); T. J. Hicks and D. Moze, J. Phys. F **11**, 2633 (1981).

[7] S. Senoussi and Y. Öner, J. Appl. Phys. **55**, 1472 (1984); Phys. Rev. B **28**, 455, (1983); see also Y. Öner, D. Elkhatouri, and S. Senoussi, J. Magn. Magn. Mater. **54–57**, 151 (1986).

[8] I-A. Campbell, H. Hurdequint, and F. Hippert, Phys. Rev. B **33**, 3540 (1986).

[9] K. Moorjami, T. O. Poehler, F. G. Satkiewies, M. A. Munheimer, D. J. Webb, and S. M. Bhagat, J. Appl. Phys. **57**, 3444 (1985).

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

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

[12] L. Lundgren, P. Svedlindh, P. Nordblad, and O. Beckman, Phys. Rev. Lett. **51**, 911 (1983); see also Ralph V. Chamberlin, J. Appl. Phys. **57**, 3377 (1985), and references therein.