# Effect of low dc magnetic field on the premartensitic phase transition temperature of ferromagnetic $Ni_2MnGa$ single crystals



2001 J. Phys.: Condens. Matter 13 2607

(http://iopscience.iop.org/0953-8984/13/11/316)

INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER

J. Phys.: Condens. Matter **13** (2001) 2607–2613 www.iop.org/Journals/cm PII: S0953-8984(01)19408-7

# Effect of low dc magnetic field on the premartensitic phase transition temperature of ferromagnetic $Ni_2MnGa$ single crystals

**W H Wang[1], J L Chen[1], S X Gao[1], G H Wu[1], Z Wang[2], Y F Zheng[2], L C Zhao[2] and W S Zhan[1]**

[1] State Key Laboratory for Magnetism, Institute of Physics and Centre for Condensed Matter Physics, Chinese Academy of Science, PO Box 603, Beijing 100080, People's Republic of China
[2] School of Materials Science and Engineering, Harbin Institute of Technology, Harbin 150001, People's Republic of China

Received 27 November 2000, in final form 15 January 2001

**Abstract**

The temperature and field dependence of alternating-current (ac) susceptibility in a $Ni_{50}Mn_{25}Ga_{25}$ single crystal was measured using an ac susceptometer with a dc magnetic field oriented in the [100] and [110] directions, respectively. It is found that even at very low fields the premartensitic transition temperature decreases monotonically with the increase of the applied field, and it decreases more rapidly with field applied along the [110] direction than with field along the [100] direction. In accord with a previous model, present results confirm that the magnetoelastic interaction is responsible for the premartensitic transition, and the magnitude of the magnetoelastic interaction in the [110] direction is stronger than that in the [100] direction.

## 1. Introduction

Ni–Mn–Ga alloys with composition close to the stoichiometric Heusler $Ni_2MnGa$ have been attracting investigation due to their some unique precursor phenomena at temperatures above the martensitic transformation. X-ray [1] and neutron [2] studies were the first to report the existence of the premartensitic phase before the martensitic transition. Subsequently, ultrasound [3, 4], thermal and electron microscopy [5, 6] studies on this alloy showed that a phonon anomaly exists in the slow transverse acoustic branch (TA2) well above the premartensitic transition temperature $T_I$, and the soft mode in the TA2 branch occurs at the wave vector $\xi = 0.33$. Furthermore, Planes *et al* [7] proposed a new Landau-type model for the premartensitic transitions by studying the magnetic field dependence of the structural transitions in $Ni_2MnGa$. It was suggested that the premartensitic transition is a consequence of the magnetoelastic interaction between the phonon and the spin. More recently, based on the degenerate Blume–Emery–Griffiths model, Castan *et al* [8], by means of mean-field theory and Monte Carlo simulation, indicated that a variety of premartensitic effects may appear due to the magnetoelastic interaction, when such an interaction is strong enough to freeze the relevant phonon. One recent interesting finding by Zuo *et al* [9] in a polycrystalline $Ni_2MnGa$ alloy is that $T_I$ remained constant for fields below 800 Oe but was found to decrease for higher fields suggesting a large magnetoelastic effect occurred in premartensitic phase. As well known, the magnetoelastic interaction is anisotropic, and strongly depends on the structure of the material. Therefore, it is very necessary to investigate the magnetoelastic properties in the different crystallographic directions on a single crystalline sample, especially at the lower magnetic fields in order to gain profound understanding of the origin of the phase transition in $Ni_2MnGa$. In this paper, we report the influence of low external dc magnetic field on the premartensitic phase transition in a stoichiometric $Ni_{50}Mn_{25}Ga_{25}$ single crystal. The field dependence of premartensitic transition temperature demonstrates the magnitude of the magnetoelastic interaction depends upon the direction of the applied magnetic field.

## 2. Experiment

The starting materials of the stoichiometric $Ni_{50}Mn_{25}Ga_{25}$ single crystal were prepared from metal elements Ni, Mn and Ga with the purity of 99.95%. A [001] oriented single crystal bar was used as a seed. The single crystal was grown by an MCGS-3 CZ (Czochralski) instrument with a cold crucible system [10]. The growth rates of 12–15 mm $h^{-1}$ and the rotation rate of 30 rpm were adopted. The grown single crystal was annealed at 1123 K for 4 days, and then quenched into ice water [11]. Considering the demagnetization effects, a thin disclike sample 4 mm in diameter and 1 mm in thickness was cut from the treated single crystal. The sample was oriented so that the plane of the disc is perpendicular to the [001] direction. The ac magnetic susceptibility measurements were performed by a susceptometer with an ac magnetic field of 5 Oe and a frequency of 77 Hz. The sample could be rotated in the sample holder between the poles of an electromagnet, and magnetic fields up to 10 kOe were applied along the different crystallographic directions of the sample. Furthermore, this system enabled us to perform ac magnetic susceptibility measurements over a wide temperature range, from 400 K to 77 K. The temperature alternating cooling and heating was varied at about 0.5 K $min^{-1}$.

## 3. Results and discussion

Figure 1 shows the temperature dependence of ac susceptibility $\chi$ measured in the cooling and heating processes, with the ac fields applied along the [001] direction of the sample. Since the magnetic ordering stringently follows the crystal lattice deformation, the martensitic phase transition from the cubic $L2_1$ to the tetragonal structure is marked by the drastic drop of $\chi$ on cooling because the lower symmetry of the martensitic phase enhances the magnetocrystalline anisotropy. Therefore, one can easily see that the martensitic transformation start temperature $M_s$ and reverse transition start temperature $A_s$ and the Curie temperature $T_c$ marked in figure 1 are 195, 200 and 378 K, respectively. Moreover, as marked by the arrows, two relative minimum-like anomaly kinks are clearly observed, at 241 K in the cooling run and 251 K in the heating run, respectively, which correspond to $T_I$. The inset in figure 1 presents the electrical resistance as a function of temperature with current along the [001] direction in the heating run. The $R$–$T$ curve shows two kinks at 200 and 251 K, which are in agreement with the corresponding branch in the $\chi$–$T$ curves.

Fixing the ac magnetic field along the [001] direction, we measured the magnetic field dependence of $T_I$ by measuring the ac magnetic susceptibility across the premartensitic transition with various low external dc magnetic fields applied along the [100] and [110] directions, respectively. In figure 2, we show the relative orientation of the sample, ac magnetic

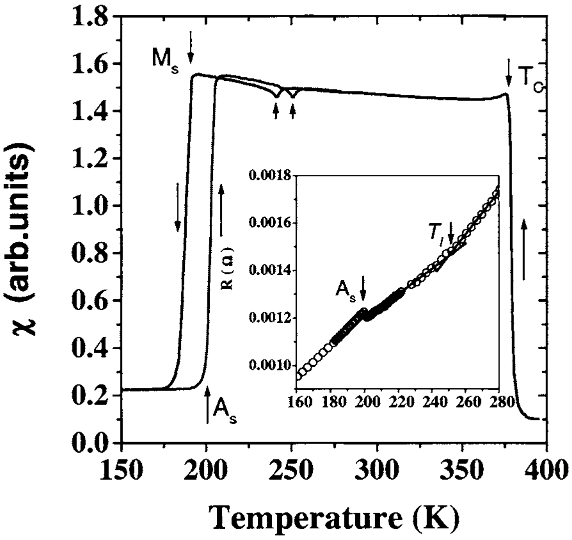


**Figure 1.** Ac magnetic susceptibility $\chi$ as a function of temperature during cooling and heating processes, for the ac magnetic field applied along the [001] directions. The inset is the electrical resistance as a function of temperature on heating. The arrows are guides to the eye.

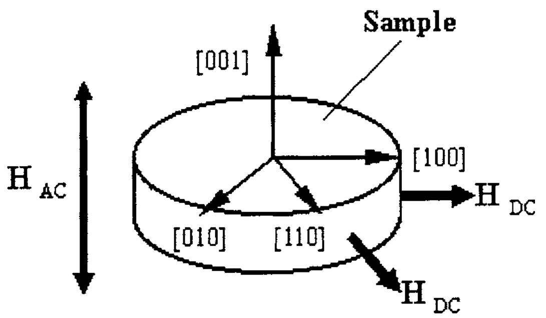


**Figure 2.** Schematic of relative orientation of sample, ac magnetic field ($H_{AC}$), and applied magnetic field ($H_{DC}$).

field ($H_{AC}$), and applied magnetic field ($H_{DC}$). Figure 3 shows an example of the results found during heating runs at magnetic fields of 0 Oe (squares), 400 Oe (circles) and 800 Oe (triangles), applied along the [110] (a) and [100] (b) directions, respectively. One can easily see that an unambiguous decrease in $T_I$ with the increase of the magnetic field has been found. If we plot $T_I$ as a function of applied magnetic field, as shown in figure 4, it is clear that $T_I$ decreases monotonically with the increase of the applied field. It is noteworthy that the relative decrease of $T_I$ for fields applied along the [110] direction is more pronounced than that of the [100] direction. In the [100] direction, the $T_I$ value at 800 Oe is about 248 K, a decrease of 3 K from the $T_I$ value at zero field. However, in the [110] direction, the $T_I$ value at 800 Oe is about 246.7 K, a decrease of 4.3 K from the $T_I$ value at zero field.

From the point of view of the thermodynamics, the external magnetic field, like the temperature, is an independent variable that changes the free energy of the $Ni_2MnGa$ alloy. Usually, the free energy of the ferromagnetic premartensitic phase can be expressed as

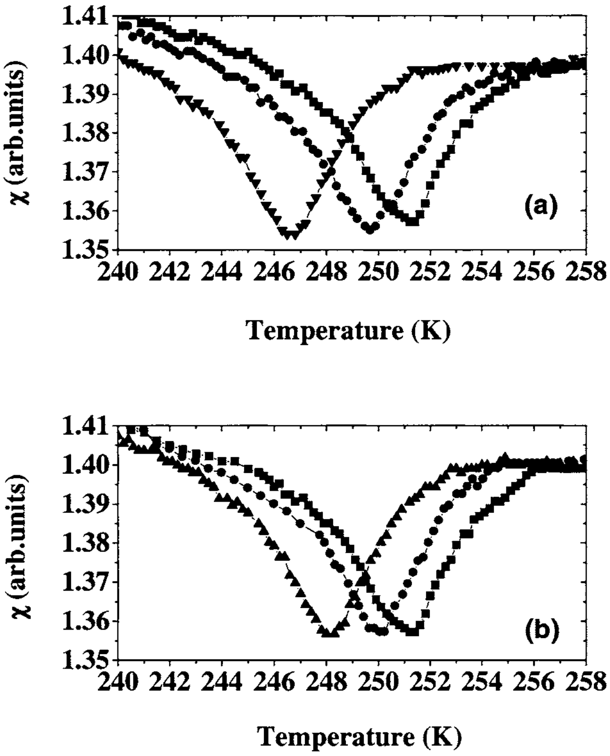


**Figure 3.** Ac magnetic susceptibility $\chi$ as a function of temperature across the premartensitic transition during heating process for dc magnetic fields (0 Oe (squares), 400 Oe (circles) and 800 Oe (triangles)) applied along the [110] (a) and [100] (b) directions, respectively.

$F = F_e + F_m + F_{em}$, where $F_e$ is the elastic energy term, $F_m$ is the magnetic energy term and $F_{em}$ is the magnetoelastic energy term. In general, the magnetic energy can be written as follows:

$$F_m = K(m_z^2 m_y^2 + m_z^2 m_x^2 + m_x^2 m_y^2) - MH \tag{1}$$

where $m = M/|M|$, $M$ is the magnetization of the crystal, $H$ is the external magnetic field and $K$ is the magnetic anisotropy constant. For simplicity, the change in free energy of the parent phase was neglected here. Therefore, when the external magnetic field is applied, the free energy $F$ of the premartensitic phase decreases mainly due to the increase of the Zeeman energy ($-MH$). Therefore, the applied magnetic field will increase $T_I$. The effect of the external magnetic field is analogous to the effect of hydrostatic pressure and external stress on the premartensitic transition in the $Ni_2MnGa$ single crystal [12, 13]. On the other hand, according to the Landau-type model [7], the premartensitic transition is a consequence of the magnetoelastic interaction between the phonon and the spin, and the magnetoelastic energy $F_{em}$ is quasilinear with $M^2$ [9]. In this case, application of the magnetic field will increase the alignment of the magnetic domains in the direction of the external magnetic field, which in turn will promote $F_{em}$, and increase the magnetoelastic energy $F_{em}$ contribution. It is obvious that the increase of the magnetoelastic interaction will lead to a premartensitic transition temperature $T_I$ decrease due to the increase of free energy of the premartensitic phase. Therefore, the total effect of the magnetic field on the free energy $F$ depends on the competition between the magnetic energy $F_m$ and the magnetoelastic energy $F_{em}$. In fact,

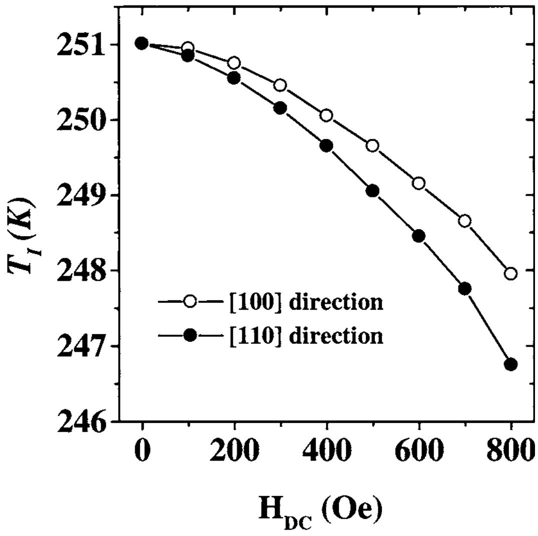


**Figure 4.** Premartensitic transition temperature $T_I$ as a function of magnetic field during the heating process, for various dc magnetic fields applied along the [100] (in open symbols) and [110] (in solid symbols) directions, respectively.

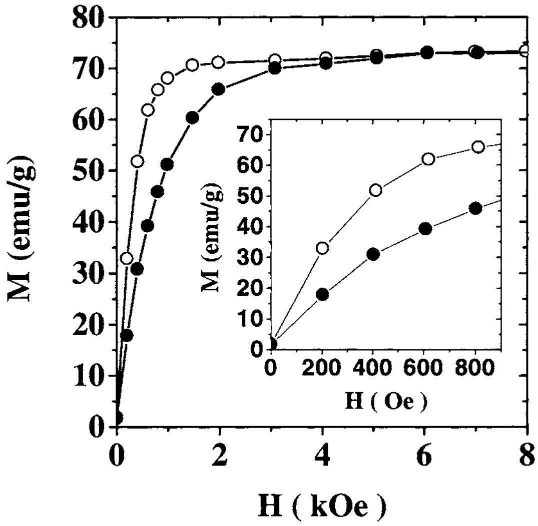


**Figure 5.** Magnetization as a function of magnetic field measured at 240 K (premartensitic phase), for dc magnetic field applied along the [100] (in open symbols) and [110] (in solid symbols) directions, respectively. The inset is a replot in the field range of interest.

in the present study, $T_I$ decreases with increasing field, suggesting that even at low field the magnetoelastic energy contribution to the free energy is the crucial term.

On the other hand, we also found that $T_I$ decreases more rapidly with field applied along the [110] direction than with field along the [100] direction. Recently, Comas *et al* [13] performed an experimental investigation of the premartensitic transition in a $Ni_2MnGa$ single crystal by the use of ultrasonic techniques. They have found that at room temperature (ferromagnetic state) all elastic constants ($C'$, $C_L$ and $C_{44}$), with increasing magnetic fields applied along

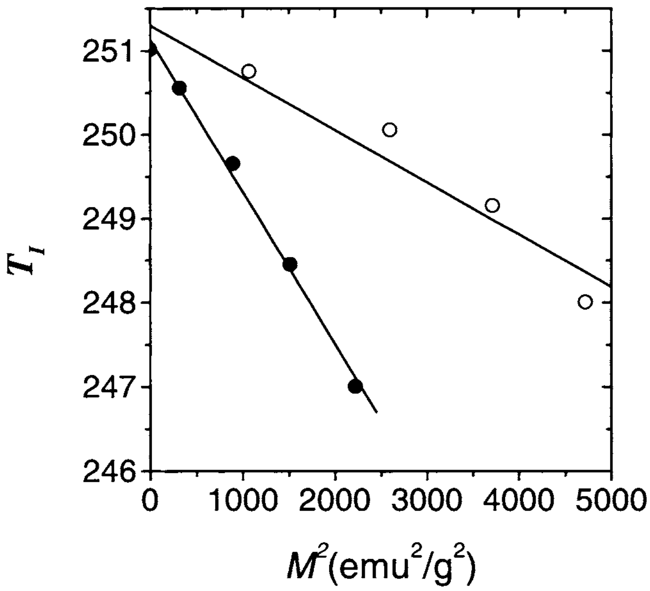


**Figure 6.** Premartensitic transition temperature $T_I$ as a function of $M^2$, for various dc magnetic fields applied along the [100] (in open symbols) and [110] (in solid symbols) directions, respectively. The line is a linear fit to the data.

the [100] and [110] directions, increase up to the same saturation value at about 1 kOe and 3 kOe, respectively. This further indicates that the magnetoelastic interaction depends on the direction of the applied field. Figure 5 shows the magnetization curves taken for magnetic field applied along the [100] and [110] directions in the premartensitic phase. They indicate that the saturation magnetization is $\sigma_s = 73$ emu g$^{-1}$, and the strength of the uniaxial magnetic anisotropy is $K_1 = 0.95 \times 10^5$ J m$^{-3}$. Moreover, before reaching saturation, the Zeeman energy ($-MH$) for field applied along the [100] direction is larger than that for field applied along the [110] direction (see the inset of figure 5). As a result, the value of $T_I$ is larger for field applied along the [100] direction than along the [110] direction. However, if we plot $T_I$ versus $M^2$ curve, as shown in figure 6, one can easily see that $T_I$ decreases significantly faster with field along the [110] direction than along the [100] direction, although a linear dependence of $T_I$ on $M^2$ is not obeyed. This suggests that the magnetoelastic interaction in the [110] direction is stronger than that in the [100] direction. Therefore, the origin of the different shape of the magnetization curves should be attributed to the different magnitude of magnetoelastic interaction in [100] and [110] directions, and, due to the magnetoelastic interaction, the elastic energy stored by the premartensitic phase gives rise to the magnetic anisotropy, which dominates the magnetization processes. As a result, the magnetization of the [110] direction is harder to saturate than that of the [100] direction in the premartensitic phase. Since the premartensitic transition strongly depends upon the orientation of the applied field, different behaviour is expected for polycrystalline and single crystal samples, especially in the low field region.

## 4. Conclusion

We have investigated the effect of low dc magnetic fields (less than 800 Oe) on the premartensitic transition temperature of a $Ni_{50}Mn_{25}Ga_{25}$ single crystal by means of ac-susceptibility measurements. It is found that even at very low fields the premartensitic transition temperature decreases monotonically with the increase of the applied field, and the relative decrease for the field oriented along the [110] direction is more pronounced than that along the [100] orientation. Present results undoubtably state a magnetoelastic interaction is responsible for the premartensitic transition, and the magnitude of the magnetoelastic interaction in the [110] direction is stronger than that in the [100] direction.

## Acknowledgment

This work has been supported by State Key Project of Fundamental Research and National Natural Science Foundation of China grant 59871062.

## References

[1] Fritsch G, Kokorin V V and Kempf A 1994 *J. Phys.: Condens. Matter* **6** L107
[2] Zheludev A, Shapiro S M, Wochner P, Schwarz A, Wall M and Tanner L E 1995 *Phys. Rev.* B **51** 11 319
[3] Worgull J, Petti E and Trivisonno J 1996 *Phys. Rev.* B **54** 15 695
[4] Manosa L, Gonzalez-comas A, Obrado E, Planes A, Chernenko V A, Kokorin V V and Cesari E 1997 *Phys. Rev.* B **55** 11 068
[5] Cesari E, Chernenko V A, Kokorin V V, Pons J and Segui C 1997 *Acta Mater.* **45** 999
[6] Kokorin V V, Chernenko V A, Cesari E, Pons J and Segui C 1996 *J. Phys.: Condens. Matter* **8** 6457
[7] Planes A, Obrado E, Gonzalez-comas A and Manosa L 1997 *Phys. Rev. Lett.* **79** 3926
[8] Castan T, Vives E and Lindgard P A 1999 *Phys. Rev.* B **60** 7071
[9] Zuo F, Su X and Wu K H 1998 *Phys. Rev.* B **58** 11 127
[10] Clark A E, Verhoeven J D, Gibson O D and McMasters O D 1986 *IEEE. Trans. Magn.* **22** 973
[11] Vasil'ev A N, Bozhko A D, Khovailo V V, Dikshtein I E, Shavrov V G, Buchelnikov V D, Matsumoto M, Suzuki S, Takagi T and Tani J 1999 *Phys. Rev.* B **59** 1113
[12] Chernenko V A and L'vov V A 1996 *Phil. Mag.* A **73** 999
[13] Comas A G, Obrado E, Mansosa L, Planes A and Chernenko V A 1999 *Phys. Rev.* B **60** 7058