MAGNETIC FIELD INFLUENCE ON THE INTERMARTENSITIC TRANSFORMATION IN THE FERROMAGNETIC SHAPE-MEMORY ALLOY Ni—Mn—Ga

V.V. KOKORIN, O.M. BABIY

Institute of Magnetism, Nat. Acad. Sci. of Ukraine
(36b, Academician Vernadsky Blvd., Kyiv 03142, Ukraine; e-mail: babiy@imag.kiev.ua)

UDC 669.24′74′871:537.634

Influence of a magnetic field on the intermartensitic transformation in the Ni—Mn—Ga alloy is studied by measuring the electroresistance and the low-field magnetic susceptibility of the alloy. The effect of an abnormally large displacement of the start temperature M_s' of the intermartensitic transformation in a magnetic field with respect to the main transition temperature has been described. The displacement of the start temperature of the main intermartensitic transformation was observed to be at most 1 K in a magnetic field $H = 10$ kOe, whereas the increment of M_s' was $\Delta T = (7 \pm 1)$ K. At the same time, no displacement of the start temperature of the inverse (at heating) intermartensitic transformation was observed. The growth of the magnetic field resulted in a narrowing of the temperature hysteresis of the intermartensitic transformation.

Ferromagnetic shape-memory alloys have attracted attention due to revealing significant deformations in the martensite phase of Ni—Mn—Ga alloys caused by a magnetic field [1, 2]. The authors succeeded in discovering a magnetoplastic deformation in those alloys, which was induced by a movement of interfaces between mobile martensite twins, possessing a high magnetic anisotropy of crystals ($K_{\rm eff} \approx 10^6$ erg/cm^3), in a magnetic field $H \geq 3$ kOe. A possibility of such a deformation through twinning under the influence of an applied field was discussed earlier in work [3] in connection with an interpretation of the magnetization curves of crystals the rare-earth element Dy obtained in the fields $H \geq 100$ kOe. The suggestion to use the martensite of Ni-Mn-Ga alloy to observe the deformation induced by a magnetic field has been put forward in report [4]. The reason of such a deformation is that the set of martensite twins possessing magnetic uniaxial anisotropy includes such twins whose easy magnetic axis and, therefore, the magnetic moment **M** are close by their orientation to the direction of an external field **H**. Such twins possess a smaller magnetic energy $E = -\mathbf{MH}$ in comparison with the others. Since the constant of anisotropy is rather high in this case and the process of the magnetic moment rotation in a magnetic field is hampered, the alternative process comes into action during magnetization, namely, the growth of twins with smaller energy at the expense of less favorably oriented twins, which results in varying the dimensions of the relevant specimen, i.e. in plastic deformation. The crystal lattice of the martensite in these alloys is unstable and undergoes a sequence of martensitic transformations which were named intermartensitic transitions, under the action of mechanical stresses [5] and as a result of cooling [6]. One should bear in mind that the high-temperature phase with a cubic lattice (structure type $L2_1$) transforms into the martensite phase (hereafter, we call this transformation as "main"). This phase can possess, depending on the alloy composition, a tetragonal crystal lattice (with the degree of tetragonality $c/a < 1$) with five-layer modulation (β_1-phase), or a lattice of orthorhombic symmetry with seven-layer modulation (β_2-phase), or a tetragonal lattice (with $c/a > 1$) without modulation (β_3-phase). The study of spontaneous intermartensitic transformations is interesting both from the viewpoint of general physics, because the variation of physical properties at such transitions has not been considered sufficiently, and in a practical aspect, e.g., the use of the shape-memory effect which also manifests itself in this case [7].

In particular, it is quite important to study the influence of a magnetic field on intermartensitic transformations. To our knowledge, there are no works, where this issue has been considered. It should also be taken into account that magnetically induced martensitic transformations can be involved in the generation of deformations by an applied field [8]. Meanwhile, the temperature displacements of intermartensitic transitions can be much larger in comparison with the case where the field affects the main transition between the high-temperature and martensite phases. For example, the temperature displacement of the interphase equilibrium between two ferromagnetic martensite phases $\Delta T \sim H\Delta M/\Delta S$ [9], where ΔM and

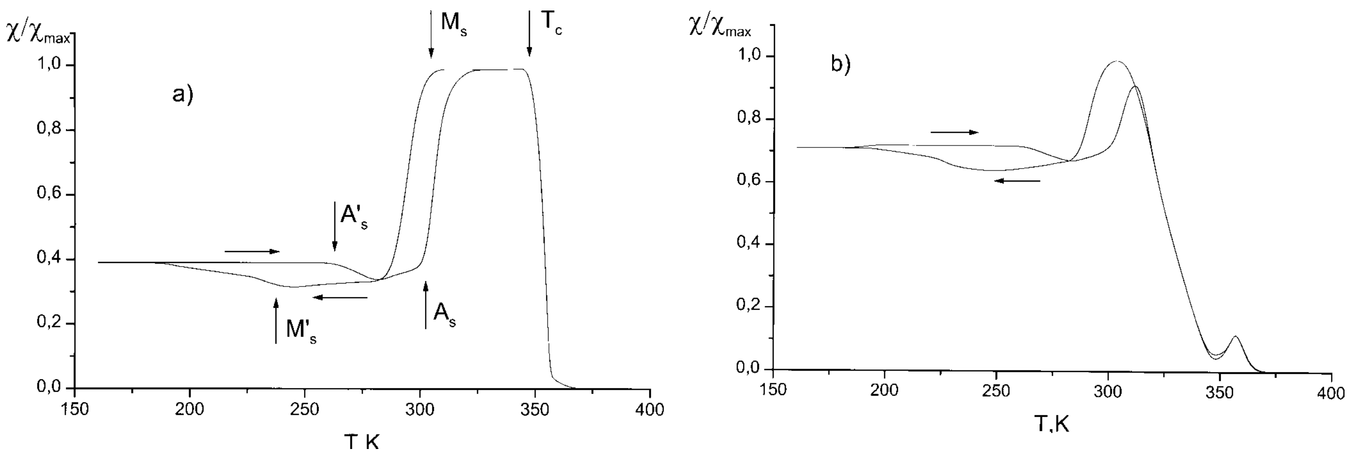

Fig. 1. Temperature dependences of the low-field magnetic susceptibility $\chi/\chi_{\max}$ of a specimen of the Ni-Mn-Ga alloy, where $\chi_{\max}$ is the maximal value of χ: (*a*) $\chi(T)$ measured at $H = 0$, where H is the external constant magnetic field; the temperature M_s corresponds to the start of the direct (at cooling) martensitic transformation and the temperature A_s does to the start of the inverse (at heating) main martensitic transformation; the temperatures M_s' and A_s' correspond to the start of the direct and inverse intermartensitic transformations, respectively; T_c is the Curie temperature of the high-temperature phase; (*b*) $\chi(T)$ measured at $H = 600$ Oe

ΔS are, respectively, the differences of the magnetic moments and entropies of these phases. In this case, ΔS is substantially smaller for intermartensitic transformations than that for the main transition. This can be judged by comparing the heats $L = T_0\Delta S$ of those transitions, T_0 being the transition temperature. The value of L for the main transition is approximately ten times as large as that for the intermartensitic one [10]. Therefore, provided that the T_0 values are close, the same is valid for a ratio between ΔS of these two types of transformations. According to the expression for ΔT given above, a reduction of ΔS can lead to a substantial increase of ΔT for the intermartensitic transition, provided that the values of ΔM are comparable.

In this article, we describe an effect of an abnormally large displacement of the start temperature of the intermartensitic transformation in a magnetic field as compared to the displacement of the main transition temperature.

Specimens composed of big crystals of the alloy (in at. %) Ni(52)-Mn(24.4)-Ga(23.6), melted in an induction-arc furnace in the argon environment, were used for researches. The obtained ingot was melted again in a furnace with a molybdenum heater. When cooling, the remelted ingot was held at 1273 K for 2 h and then slowly cooled together with the furnace. The temperature dependences of the initial magnetic susceptibility χ and the electroresistance R were measured. While measuring χ, an ac magnetic field with a frequency of 1 kHz and an amplitude of about 2 Oe was used. The electroresistance R was measured using the specimens that had been additionally annealed in a vacuum furnace at 973 K for 12 h. The electroresistance was measured using an alternating current with a frequency of 1 kHz. A rod-like specimen, cut out from the ingot, was placed into the gap of an electromagnet, with the specimen's axis being parallel to the field **H**.

As was shown earlier [5], intermartensitic transitions are accompanied by a variation of χ. The electroresistances of various martensite phases are also different [11]. The characteristic temperatures of the martensitic and intermartensitic transitions in a Ni-Mn-Ga specimen can be determined making use of its dependence $\chi(T)$ (see Fig. 1). In the case of a not annealed specimen, the start temperature of the martensitic transformation $M_s \approx 300$ K, and that of the intermartensitic transition $M_s' \approx 240$ K.

A remarkable increase of χ at M_s' corresponds to the transition of the β_2-phase with a seven-layer modulation of the lattice into the β_3-phase with a non-modulated crystal lattice, which is in agreement with the results reported in works [11–13]. When heating the specimen, the inverse transformation $\beta_3 \to \beta_2$ takes place at $A_s' = 262$ K which is accompanied by a decrease of χ. A constant magnetic field $H = 600$ Oe applied along the specimen axis changes the profile of $\chi(T)$ to some extent (Fig. 1,*b*). The temperature M_s' is shifted towards higher values by about 1 K, while the temperature A_s' retains its value. At the Curie temperature, the maximum of

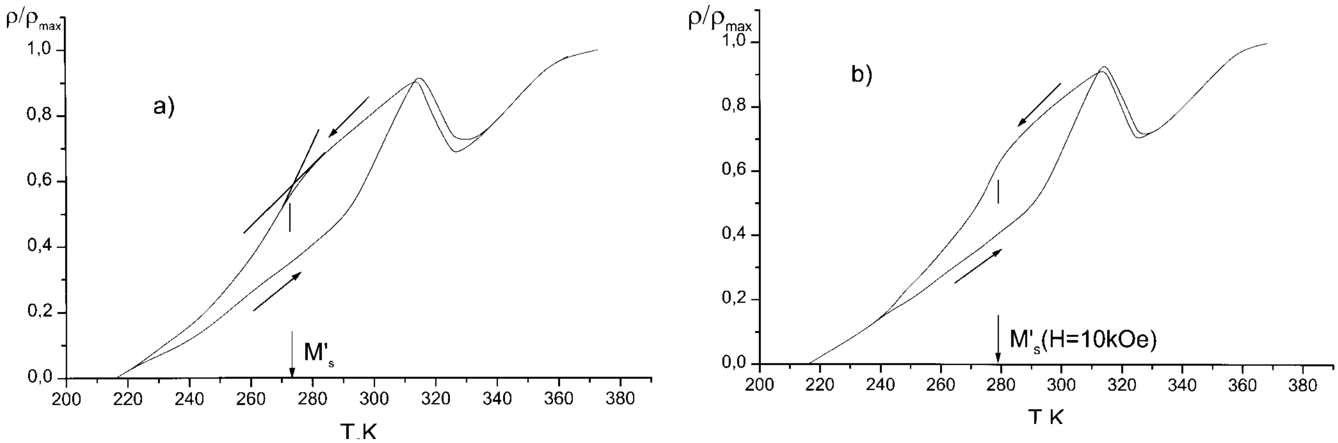

Fig. 2. Temperature dependences of the electroresistance R of a specimen made of the Ni—Mn—Ga alloy annealed at 973 K for 12 h, at (a) $H = 0$; the intersections of the straight lines shown in the figure correspond to the temperatures M'_s and A'_s; and (b) $H = 10$ kOe

the paraprocess susceptibility is observed. The constant magnetic field applied at $T \leq T_c$ results in a single-domain state, with the magnetization reversal of the specimen occurring at the expense of the paraprocess. At $H \approx 0$, the contribution of domain interfaces dominates and, owing to the paraprocess, the peak of the susceptibility is observed with difficulty.

The annealing at 973 K for 12 h changes the state of a specimen. One can see (Fig. 2,a) that the annealing leads to an increase of M_s up to 325 K, M'_s up to 273 K, and A'_s up to 293 K. The variation of martensitic transformation temperatures induced by annealing can be connected to the increase of the atomic long-range order degree in a superstructure of the $L2_1$ type, which is possessed by the high-temperature phase of the intermetallic compound Ni_2MnGa. To increase the measurement accuracy of the characteristic temperatures of the specimen in the annealed state, the curves describing the temperature dependences of the electroresistance (Fig. 2) were used. The temperature hysteresis of the main transformation reduced substantially and amounted to 2—3 K.

Fig. 2,b displays the dependence $R(T)$ for an annealed specimen which was measured under a constant magnetic field $H = 10$ kOe. Within the errors of measurements (1 K), the magnetic field does not influence the characteristic temperatures of the main transformation, which agrees with the results of work [14], where displacements of the main transformation temperatures by no more than 1 K and also in the field $H = 10$ kOe were observed. A more appreciable influence of the field is observed in the case of the intermartensitic transformation. For example, M'_s is shifted by $\Delta T = (7 \pm 1)$ K as H increases to 10 kOe

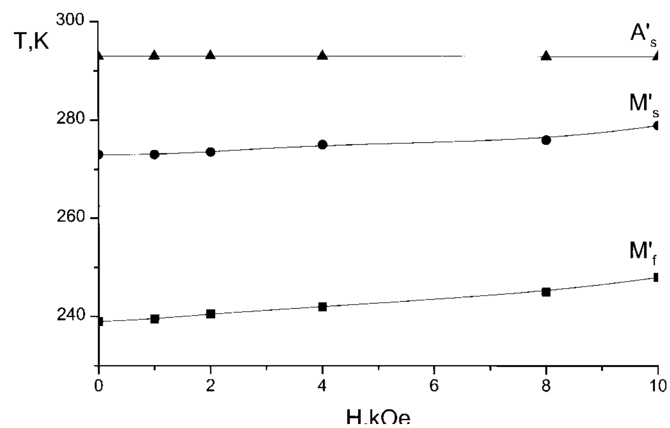

Fig. 3. Dependences of the characteristic temperatures of the intermartensitic transformation in an annealed specimen on the field value. M'_f is the final temperature (at cooling) of the intermartensitic transformation

(Fig. 2,b). The field dependences of the characteristic temperatures of intermartensitic transformations in an annealed specimen are depicted in Fig. 3. It is essential that the temperature A'_s remains constant as the field is applied, i.e. there occurs a narrowing of the temperature hysteresis of the intermartensitic transformation in a magnetic field. One can see (Fig. 3) that, as the field grows, the temperatures M'_s and A'_s approach each other. According to work [12], the specific magnetic moment of the β_3-phase is larger than that of the β_2-phase. Therefore, the magnetic (Zeeman) energy of the medium under consideration will decrease, if the β_2-martensite is replaced by the β_3-martensite. Near the equilibrium temperature of these phases, a magnetic contribution to the free energy of the system under consideration becomes appreciable, resulting in expanding the range of existence of the β_3-phase, i.e. the temperature M_s' grows. The absence of a displacement of A_s' in a magnetic field can be explained by the following. At $T = A_s'$, the difference between the magnetic moments of martensite phases becomes small in view of the proximity of A_s' to the Curie temperature. Therefore, in accordance with the above-presented formula for $\Delta T(H)$ at $\Delta M \to 0$, the transition temperature does not depend on the field.

Thus, the start temperature of the intermartensitic transformation M_s' depends noticeably on the value of the external field, in contrast to the start temperature of the main martensitic transition.

1. *Ullakko K., Huang J.K., Kantner C. et al.* // Appl. Phys. Lett. — 1996. — 69. — P. 1966.
2. *Tickle R., James R.D., Shield T. et al.* // IEEE Trans. Magn. — 1999. — 35, N 5. — P. 4301.
3. *Liebermann H.H., Graham C.D.* // Proc. Conf. held at American Institute of Physics. — 1975.— N29.— P.598.
4. *Kokorin V.V.* // Oral Communication, Intern. Conf. on Martensitic Transformations, Lausanne,1995; Proc. Intern. Conf. on Solid — Solid Phase Transformations 99, Kyoto, Japan. — 1999. — Vol.12, Pt.II. — P. 1100.
5. *Kokorin V.V., Martynov V.V.* // Fiz. Met. Metalloved. — 1991. — N 9. — P. 106.
6. *Konoplyuk S.M., Kokorin V.V.* // Ukr. Fiz. Zh. — 1998. — 43, N 10. — P. 1275.
7. *Kokorin V.V., Martynov V.V., Chernenko V.A.* // Scr. met. — 1992. — 26, Pt. 2. — P. 175.
8. *Kokorin V.V., Wuttig M.* // J.Magn. and Magn. Mater. — 2001. — 234. — P. 25.
9. *Vasil'ev A., Bozhko A.D., Khovailo V.V. et.al.* // Phys. Rev. B. — 1999. — 59, N 2. — P. 1113.
10. *Chernenko V.A., C.Segui, E.Cesari et al.* // Phys. Rev. B. — 1998. — 57, N 5. — P.2659.
11. *Wang W.H., Liu Z.H., Zhang J. et.al.* // Ibid. — 2002. — 66. — P. 052411.
12. *Kokorin V.V., Chernenko V.A., Val'kov V.I. et al.* // Fiz. Tverd. Tela. — 1995. — 37, N 12. — P. 3718.
13. *Sozinov A., Likhachev A.A., Lanska N., Ullakko K.* // Appl. Phys. Lett. — 2002. — 80, N 10. — P. 1746.
14. *Dikshtein I., Koledov V., Shavrov V. et al.* // IEEE Trans. Magn. — 1999. — 35, N 5. — P. 3811.

Received 05.04.04.
Translated from Ukrainian by O.I. Voitenko

ВПЛИВ МАГНІТНОГО ПОЛЯ НА ІНТЕРМАРТЕНСИТНЕ ПЕРЕТВОРЕННЯ У ФЕРОМАГНІТНОМУ СПЛАВІ Ni—Mn—Ga З ПАМ'ЯТТЮ ФОРМИ

В.В. Кокорін, О.М. Бабій

Р е з ю м е

Досліджено вплив магнітного поля на інтермартенситне перетворення в сплаві Ni—Mn—Ga методами вимірювання електроопору та низькопольової магнітної сприйнятливості. Описано ефект аномально великого зміщення в магнітному полі температури початку інтермартенситного перетворення M_s' порівняно з основним. Відмічено, що зміщення температури початку основного мартенситного перетворення не перевищує 1 К у полі $H = 10$ кЕ, тоді як M_s' збільшується на $\Delta T = (7 \pm 1)$ К. При цьому зміщення температури початку зворотного (в процесі нагрівання) інтермартенситного перетворення не виявлено. Збільшення поля веде до зменшення температурного гістерезису інтермартенситного перетворення.