The magnetic field influence on magnetostructural phase transition in Ni$_{2.19}$Mn$_{0.81}$Ga

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Magnetic properties of a polycrystalline alloy Ni$_{2.19}$Mn$_{0.81}$Ga, which undergoes a first-order magnetostructural phase transition from cubic paramagnetic to tetragonal ferromagnetic phase, are studied. Hysteretic behavior of isothermal magnetization $M(H)$ has been observed in a temperature interval of the magnetostructural transition in magnetic fields from 20 to 100 kOe. Temperature dependencies of magnetization $M$, measured in magnetic fields $H = 400$ and 60 kOe, indicate that the temperature of the magnetostructural transition increases with increasing magnetic field.

Mn-containing Heusler alloys Ni$_{2+x}$Mn$_{1-x}$Ga have been intensively studied during last years owing to their unique combination of ferromagnetism and a structural (martensitic) phase transition. Since martensitic transformation in Ni–Mn–Ga is of a thermeelastic type, the materials exhibit a well-defined one- and two-way shape memory effect [1,2]. For the stoichiometric Ni$_2$MnGa composition, martensitic transition temperature $T_m = 202$ K and Curie temperature $T_C = 376$ K [3]. A phase diagram of magnetic and structural transitions in Ni$_{2+x}$Mn$_{1-x}$Ga alloys studied, theoretically and experimentally, in Refs. [4,5]. It has been found, that a partial substitution of Mn for Ni leads to a decrease of Curie temperature $T_C$ and an increase of the martensitic transition temperature $T_m$, until they merge in a compositional interval $x = 0.18 - 0.20$ (Fig. 1). The application of an external magnetic field results in an increase of the martensitic transition temperature [6–8]. If applied magnetic field is strong enough, a reversible magnetostructural transition can be realized in the magnetic field at constant temperature and pressure [9]. This means that the structural state of a sample can be switched reversibly and that magnetic shape memory effect can be realized by means of magnetic-field-induced martensitic transition [10, 11]. Although Ni$_{2+x}$Mn$_{1-x}$Ga alloys were studied by a variety of methods, a systematic study of their magnetic properties in the vicinity of the magnetostructural transition is lacking. Magnetic properties of Ni$_{2.18}$Mn$_{0.82}$Ga were studied in Ref. [12], where hysteretic behavior of temperature dependencies of magnetization $M(T)$ has been observed. However, the anomalous field dependence of magnetization $M$, reported in Ref. [12], was found to be sample dependent and, therefore needs further investigation. The aim of this work is to investigate magnetic properties of polycrystalline Ni$_{2.19}$Mn$_{0.81}$Ga alloy in strong magnetic fields in the vicinity of the magnetostructural transition.

Polycrystalline Ni$_{2.19}$Mn$_{0.81}$Ga ingots were prepared by a conventional arc-melting method in argon atmosphere. To attain a good compositional homogeneity, the ingots were remelted three times. Since weight loss during melting was approximately 0.1%, the composition of the ingots was assumed to be the nominal one. The ingots were annealed in vacuum at 1050 K for 3 days. The sample for measurements was cut from the middle part of the ingot. Temperature and magnetic field dependencies of magnetization were measured by a magnetometer equipped with a Hall sensor. Magnetic fields up to 100 kOe were generated by a Bitter magnet. Measurements of $M(H)$ were done in the following way. Initially the sample was heated to $T = 380$ K, well above the temperature of the transition. After that the sample was cooled down to a desired temperature, which was controlled by a temperature controller. In these measurements the magnetic field was changed with a step of 10 kOe and was kept for some time in order to insure that processes of structural transformation for a given strength of the magnetic field had completely been finished.

FIG. 1: The composition dependence of Curie temperature $T_C$ and the martensitic transition temperature $T_m$ in Ni$_{2+x}$Mn$_{1-x}$Ga ($x = 0 - 0.20$).
FIG. 2: Temperature dependencies of the magnetization of Ni$_{2.19}$Mn$_{0.81}$Ga, measured in $H = 400$ Oe (a) and $H = 60$ kOe (b).

Typical temperature dependencies of magnetization $M$ in the vicinity of the magnetostructural transition are shown in Fig. 2. It is evident that as in the case of weak magnetic field $H = 400$ Oe (Fig. 2a), in a strong magnetic field $H = 60$ kOe (Fig. 2b), the magnetization has a prominent temperature hysteresis. Therefore, the magnetic transition in the Ni$_{2.19}$Mn$_{0.81}$Ga alloy possesses characteristics typical of a first-order phase transition. Under the action of the magnetic field the characteristic temperatures of the magnetostructural transition shift upward with a rate of $\approx 0.1$ K/kOe. This value agrees well with the data, published in Refs. [7,9,12].

Isothermal magnetization curves taken at various temperatures in the vicinity of the magnetostructural transition are shown in Fig. 3. The $M(H)$ curves, measured at $T = 327$ (Fig. 3a) and 345 K (Fig. 3d) have field dependencies of magnetization typical of a ferromagnet and a paramagnet, respectively. Contrary to this, the $M(H)$ curves, measured in the temperature interval of the mixed (martensite and austenite) state (Fig. 3b and 3c) demonstrate hysteretic anomalies of magnetization in magnetic field up to 100 kOe. As is seen from Fig. 3b, in the magnetic field 0–20 kOe the behavior of the magnetization is similar to the paramagnetic state. Further increase of the magnetic field results in an anomalous growth of the magnetization, which reaches, in a magnetic field of 100 kOe, 90% of the magnetization of the ferromagnetic state at $T = 327$ K (Fig. 3a). Upon subsequent removal of the magnetic field, the magnetization remains constant down to 20 kOe, where a drastic decrease, typical for technical magnetization curve of a ferromagnet, is observed. Based on the behavior of $M(H)$, measured at $T = 339$ K, it can be suggested that induced by the magnetic field, low-temperature (martensitic) ferromagnetic state of the sample remains stable upon removal of the magnetic field.

The $M(H)$ dependencies, measured during application and removal of magnetic field at $T = 341$ K are shown in Fig. 3c. At this temperature the behavior of the magnetization is similar to the paramagnetic state in magnetic fields up to 30 kOe. When the magnetic field increases from 30 to 100 kOe, the magnetization anomalously grows and reaches a value of 60% of the magnetization of the ferromagnetic state at $T = 327$ K. Upon subsequent decrease of the magnetic field from 100 to 30 kOe, the attained value of the magnetization is preserved. A rapid change of $M$ occurs at $H = 30$ kOe, where the magnetization drops to a value of 110% of the magnetization of the paramagnetic state at $T = 345$ K. During further decrease of the magnetic field from 30 to 0 kOe, the behavior of the magnetization is typical of the paramagnetic state. The anomalous $M(H)$ dependencies measured at $T = 341$ K can be explained as due to a reversible structural transformation from paramag-
magnetic austenite to ferromagnetic martensite, induced by a magnetic field at a constant temperature.

The presented results of the magnetic measurements are in accordance with experimental results, reported by Dikshtein et al. [9], where a reversible magnetic-field-induced structural phase transition was observed by an optical method. As is seen from Fig. 3, induction of a structural transition by a magnetic field is accompanied by unusual behavior of the isothermal magnetization $M(H)$ in the temperature interval of the coexistence of ferromagnetic martensite and paramagnetic austenite. Actually, similar anomalies on $M(H)$ dependencies were observed in Gd$_5$(Si$_x$Ge$_{4-x}$) [13], which is another representative of the intermetallic compounds with coupled magnetostructural transition. Since such systems, with a close relation between crystallographic structure and magnetism, are of a great technological significance, further studies of magnetostructural transitions in Ni$_{2+\delta}$Mn$_{1-x}$Ga [12], Co–Ni–Ga, and Co–Ni–Al [14] alloys are of considerable interest.

We are grateful to Professor A. N. Vasilev for helpful discussions. This work was partially supported by the Grant of RFBR–BRFBR 02–02–81030 Bel2002a, by the Grant of RFBR 02–02–16636a, and by the Grant-in-Aid for Scientific Research (B) No. 11695038 from the Japan Society for the Promotion of Science, and Izumi Science Technology Foundation.

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