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Temperature Dependent Magnetic and Structural Properties of Ni–Mn–Ga Heusler Alloy Glass-Coated Microwires

S. Shevyrtalov[a,*], K. Chichay[a], P. Ershov[a], V. Khovaylo[b,c], A. Zhukov[d,e], V. Zhukova[d] and V. Rodionova[a,b]

[a]Innovation Park and Institute of Physics and Technology, Immanuel Kant Baltic Federal University, Kaliningrad 236041, Russia

[b]National University of Science and Technology MISiS, Moscow 119049, Russia

[c]ITMO University, St. Petersburg 197101, Russia

[d]Dpto. Fisica de Materiales, Fac. Quimicas, sUPV/EHU, 20009 San Sebastian, Spain

[e]IKERBASQUE, Basque Foundation for Science, 48011 Bilbao, Spain

We report on structural and magnetic properties of glass-coated microwires of Ni–Mn–Ga-based Heusler alloys. Structural characterization of the as-prepared microwires revealed that they have a cubic structure at room temperature. It is shown that magnetic properties of microwires can be tailored by heat treatment as well as by removing the glass coating. Specifically, annealing of the microwires has a marked influence of the Curie temperature T_C which increases significatly in the heat-treated samples. Release of internal stresses in the microwires by removing the glass coating causes magnetization and the Curie temperature to decrease. This allowed us to conclude that in the studied microwires the magnetostriction constant is positive and estimate the value of internal stresses as being roughly equal to 1.5 GPa.

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1. Introduction

In last years, Ni–Mn–X Heusler alloys have attracted significant attention because of number of their functional properties such as magnetic shape memory effect, magnetic field-induced martensitic transformation, magnetocaloric effect (MCE), etc. [1].

Thin magnetic glass-coated microwires are very promising materials for magnetic applications [2–4]. One of the main advantages of microwires in comparison with bulk materials that can be used for magnetocaloric cooling is its volume to surface ratio. Another advantage of using microwires with glass-coating is their mechanical stability and a negligible influence of the glass-coating on the heat-exchange between fluid and MCE material [5].

The Taylor–Ulitovsky technique allows fabrication of a few km long glass-coated microwires with the nucleus diameter d ranging from 1 to 30 μm and the glass-coating thickness between 0.5 and 20 μm with different structures [2, 3]. Recently, this technique has been successfully used to prepare Heusler-alloy microwires with different compositions and structure [6–8], including Ni–Mn–Ga alloy [5, 9, 10].

For the present work, Ni–Mn–Ga glass-coated microwires were prepared by the Taylor–Ulitovsky technique. We have studied magnetic and structural properties of annealed and as-prepared microwires. Hysteresis loops were measured at different temperatures for annealed samples and then compared with those for microwires without glass coating. X-ray diffraction (XRD) measurements were performed at room temperature for both types of wires.

2. Experimental details

Ni–Mn–Ga glass-coated microwires were prepared by the Taylor–Ulitovsky technique [11]. The microwires have a total diameter $D = 54.6$ μm and the nucleus diameter $d = 26.7$ μm.

Pieces of the microwires were annealed at 833 K in protective argon atmosphere during 35 min. The annealed samples were slowly cooled down to room temperature to relax internal stresses. Energy dispersive spectrometer (Oxford Instr. X-Act) was used to detect composition of metallic core of as-cast and annealed microwires. XRD patterns were collected by a Bruker D8 Discover diffractometer with a Cu K_α radiation.

Magnetic measurements were performed using LakeShore 7400 vibrating magnetometer in magnetic fields up to 10 kOe.

3. Results and discussion

The EDS microanalysis, performed at 5 different points of the sample showed homogeneous chemical elements distribution. The average chemical compositions of the samples were determined as $Ni_{63.28}Mn_{11.94}Ga_{24.78}$ (at.%). After thermal treatment the chemical compositions remained basically the same.

The diffraction patterns taken at room temperature indicated that all samples have a cubic structure. Diffraction patterns of the studied microwires are shown in Fig. 1.

*corresponding author; e-mail: shevyrtalov@gmail.com

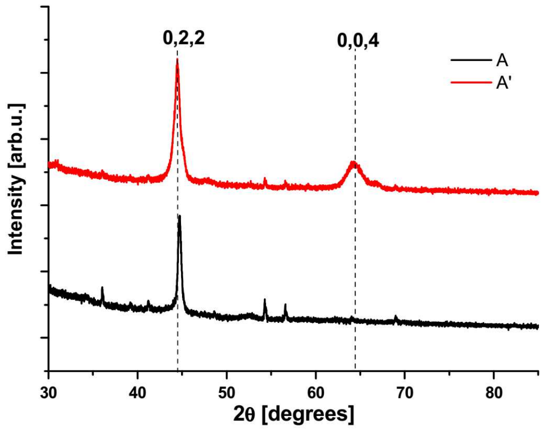

Fig. 1. X-ray diffraction patterns of as-prepared (A) and annealed (A′) $Ni_{63.28}Mn_{11.94}Ga_{24.78}$ microwires.

We assumed that the three peaks seen on $2\theta = 44.47$, 64.24 and 98.24° are diffraction from (0,2,2), (0,0,4) and (0,4,4) planes, respectively, of Ni_2MnGa cubic phase with a lattice parameter $a = 5.758$ Å. Calculated lattice parameter a of as-prepared and annealed microwire are listed in Table. Other peaks on the diffraction pattern which do not correspond to the cubic structure are due to the tape which held the sample. A more close inspection of the diffraction pattern taken for the annealed microwire (Fig. 1) points to a small splitting of the $2\theta = 45°$ and $2\theta = 67°$ peaks which probably comes from a small amount of a secondary phase in the wire. In fact, the martensitic phase is reasonably to expect at room temperature in the studied microwire due to a substantial excess of Ni atoms which leads to a marked increase of the martensitic transformation temperature in a $Ni_{50+x}Mn_{25-x}Ga_{25}$ alloy system [12].

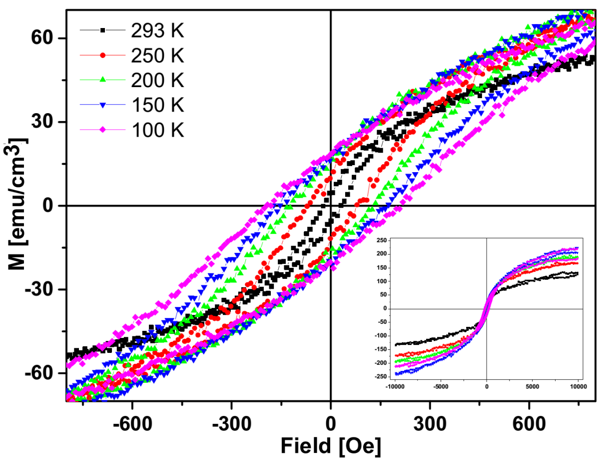

Fig. 2. Hysteresis loops measured in the annealed microwires at different temperatures. The inset shows hysteresis loops in the whole range of applied magnetic field.

The as-cast microwires did not show ferromagnetic ordering at room temperature. After annealing at 833 K magnetic properties of the sample changed dramatically. The annealed sample exhibits ferromagnetic behavior up to 300 K (Fig. 2). Coercive field value (H_c) decreases with increase of temperature, varying from $H_c = 187.8$ Oe at 100 K down to 32.7 Oe at 300 K. The coercive field decreases linearly with increasing temperature (Fig. 3). The decrease of H_c is due to a weakening of the magnetocrystalline anisotropy when approaching the Curie temperature T_C.

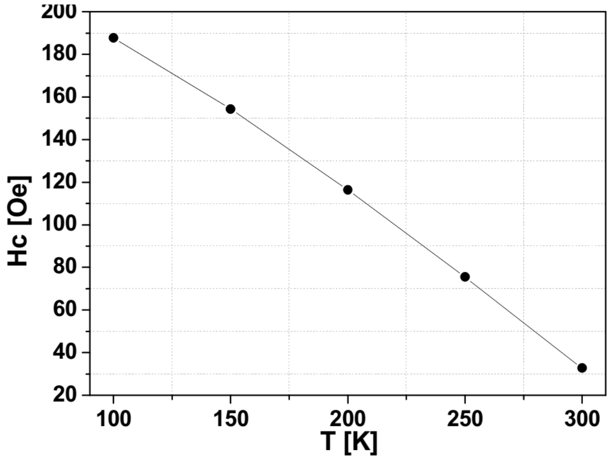

Fig. 3. Coercive field as a function of temperature for the annealed sample.

TABLE

Calculated lattice parameter a of $Ni_{63.28}Mn_{11.94}Ga_{24.78}$ microwires.

Sample	Description	(0,2,2) peak position	Lattice parameter a
A	as-prepared	44.72°	5.728 Å
A′	annealed	44.47°	5.758 Å

Hysteresis loops measured at room temperature for the sample before and after glass-coating removal are shown in Fig. 4. It is clearly seen that magnetization M of the wire without coating is lower than that in the wire with the glass coating. This observation implies that internal stresses induced by the glass coating strongly affect magnetic properties of the microwires. Specifically, since similar stress-induced change of magnetization has been reported for Fe-based amorpous microwires [13], it can be suggested by analogy with those results [13] that the magnetostriction constant is positive in the studied Ni–Mn–Ga microwires.

Magnetization vs. temperature curves measured in a temperature range from 200 to 365 K in an applied magnetic field of 1 kOe are shown in Fig. 5. (M_0 is the magnetic moment at the lowest temperature.) The Curie temperature of the microwires was determined as a minimum on the curves of partial derivative of the magnetization M with respect to temperature T, $\partial M/\partial T$. Estimated in such a way values of T_C were found to be of about 325 K for the glass-coated microwire and 310 K for the microwire after glass removal. Assuming

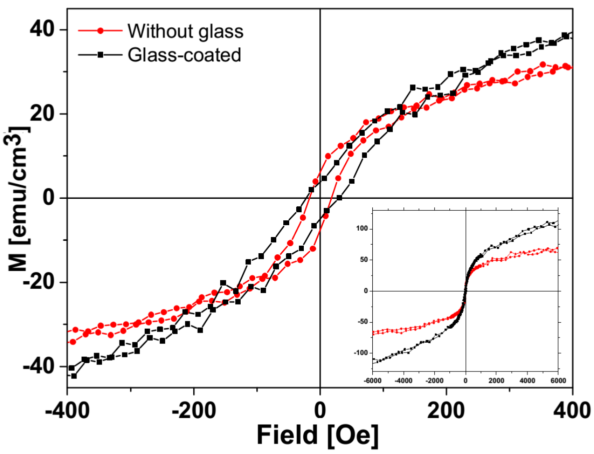

Fig. 4. Hysteresis loops measured at room temperature for the annealed sample before (squares) and after (circles) glass coating removal.

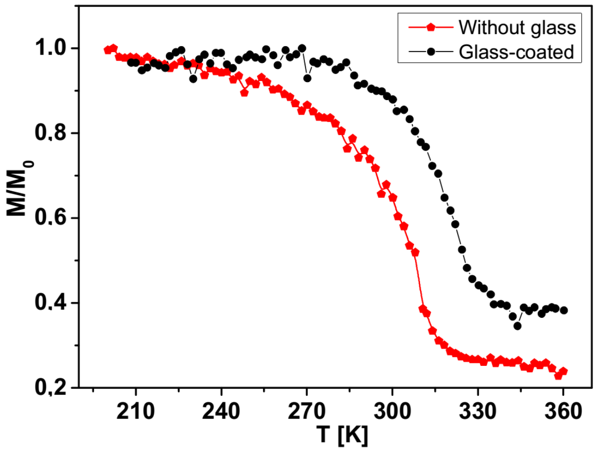

Fig. 5. Temperature dependence of the normalized magnetization for the microwires with and without glass coating in a magnetic field of 1 kOe.

that the influence of the internal stresses and an external pressure p on T_C is equivalent, one can estimate the value of the internal stresses created by the glass coating. Using reported in Ref. [14] pressure dependence of the Curie temperature for Ni_2MnGa, $dT_C/dp \approx 1$ K/kbar, the glass coating-induced internal stresses in the studied $Ni_{63.28}Mn_{11.94}Ga_{24.78}$ microwires are of about 1.5 GPa.

4. Summary

In conclusion, we have studied impact of annealing and stress on magnetic properties of Ni–Mn–Ga-based microwires. It has been shown that annealing results in an increase of the Curie temperature T_C which can be explained as due to the development of structural ordering in the as-prepared microwires. This result had clearly pointed on the role of the degree of structural disorder on magnetic properties of Ni–Mn–Ga-based materials. Alongside with the structural disorder, it has been shown that the internal stresses also affect considerably magnetic properties of the Ni–Mn–Ga-based microwires. Specifically, it has been found that removing of the glass coating which is equivalent to the removing internal stresses results in a decrease of the magnetization. We have suggested that the observed decrease of the magnetization can be explained as due to a pressure dependence of T_C in Ni–Mn–Ga alloys [14], i.e., to the dependence of exchange interactions in this alloy system on the interatomic distances.

Acknowledgments

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