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Temperature dependent magnetostrains in polycrystalline magnetic shape memory Heusler alloys

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ABSTRACT

The transformation-induced strains with and without applied magnetic field and magnetostriction loops at different temperatures have been measured for polycrystalline ferromagnetic and metamagnetic shape memory Heusler alloys such as Ni—Mn—Ga, Ni—Fe(Co)—Ga and Ni—Mn—Sn, respectively. The results evidence a magnetic field oriented growth of martensitic variants during the martensitic transformation in Ni—Mn—Ga sample, whereas this effect is not evident in the other two alloys. In the Ni—Mn—Sn alloy, the isotropic volume magnetostriction dominates the anisotropic domain magnetostriction.

1. Introduction

Ferromagnetic shape memory alloys (FSMAs), e.g., the off-stoichiometric Ni_2MnGa Heusler compounds, are well-known for their ability to generate a large magnetic field-induced strain, MFIS. This effect is due to a magnetic field induced reorientation of the martensitic domains. Other Heusler-type materials, typically Mn-rich Ni—Mn—X compounds (X = In,Sn,Sb), known as metamagnetic shape memory alloys (MMSMAs), exhibit a large MFIS due to the reverse magnetostructural transformation occurring under magnetic field. According to a magnetoelastic model, the first mechanism is triggered by a magnetostress and related magnetoelastic energy. The second one is mainly controlled by a Zeeman term ([1–3] and references therein).

Magnetoelastic coupling plays a crucial role in the magnetomechanical behaviour of FSMAs whereas its role in MMSMAs has not been clarified yet. In typical FSMA, such as Ni—Mn—Ga, the MT is accompanied by a cubic-to-tetragonal unit cell distortion with $c/a < 1$. The magneto-elastic coupling is described by a negative magnetostriction constant and responsible for the appearance of an easy-magnetization direction along the short c-axis and the large uniaxial magnetic anisotropy of martensite. It is also responsible for the occurrence of magnetostress [4]. As an analogue of Hook's law, this stress is linearly related to the magnetostriction. The magnetostriction magnitude can be comparable with the value of mechanical elastic deformation needed to trigger a twin boundary motion. So, an enhanced magnetostriction, alongside with low elastic shear modulus and low twinning stress, is an important prerequisite for large MFIS [1].

Despite such an importance, not much published work about the magnetostriction is available, probably, because of difficulties of its separation from MFIS measured in the martensitic phase (see, e.g., [5]). Incidentally, the magnetoelastic model of ferromagnetic martensite [6] and ab initio calculations [7] interrelate the magnetoelastic coupling in the martensitic and austenitic phases so, the measurements of magnetostriction in the cubic phase are necessary also in terms of MFIS of martensite. The experimentally determined magnetostriction values of an austenite phase in the single crystalline [5,6,8–11] and polycrystalline Ni—Mn—Ga alloys [12] vary from 40 to 600 ppm, the largest values are obtained in the vicinity of MT and the premartensitic transition [11].

In this work, a comparative study of the influence of the magnetic field on the sample deformation accompanying the martensitic transformations (MTs) is carried out in two polycrystalline FSMAs [Ni—Mn—Ga, Ni—Fe(Co)—Ga] and one MMSMA (Ni—Mn—Sn). We also present cyclic domain magnetostriction measurements in the cubic and martensitic phases of these alloys at different temperatures. A significant magnetic field-induced orientation effect during MT was observed in Ni—Mn—Ga FSMA only. The magnetostriction is analyzed in terms of anisotropic (domain) and isotropic (volume) contributions.

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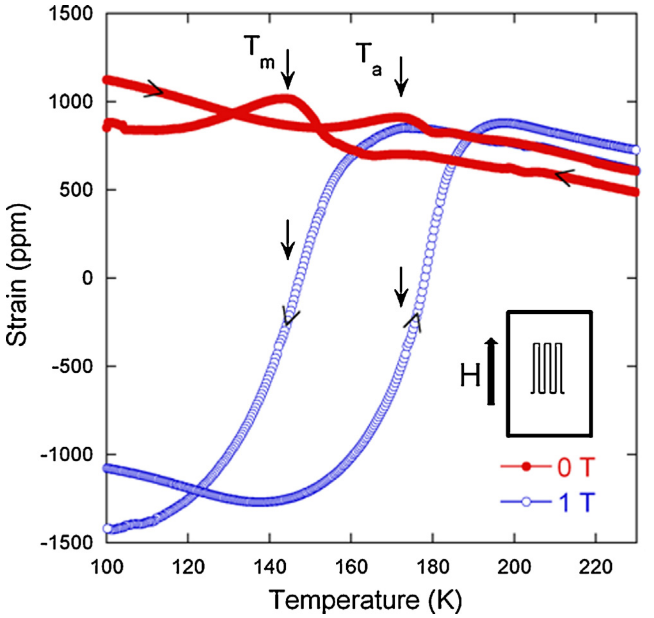

Fig. 1. Strain of Ni—Mn—Ga (alloy A1) on cooling and heating through the MT under zero field and 1 T parallel to the measuring direction. The field has a large influence in the strain at MT evidencing a preferential orientation of the variants with the short crystallographic axis in a field direction.

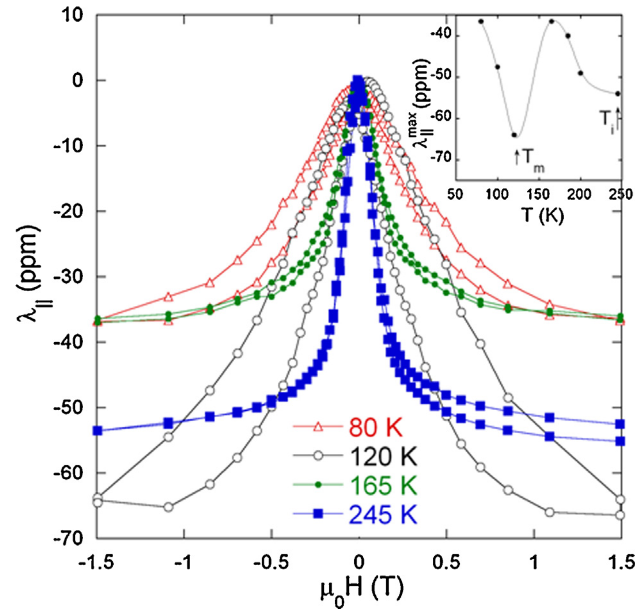

Fig. 2. Magnetostriction of Ni—Mn—Ga (alloy A1) as a function of the field at different temperatures corresponding to the austenite, premartensite and martensite. Inset: magnetostriction values at the saturating field as a function of temperature showing deep minimums in the vicinity of martensitic and premartensitic transformations. Line is a guide for the eye.

2. Experimental

Two FSMAs with compositions (in at.%) $Ni_{49.7}Mn_{24.3}Ga_{26.0}$ (A1) and $Ni_{50.1}Fe_{18.6}Co_{4.1}Ga_{27.2}$ (A2) and one MMSMA, $Ni_{50.0}Mn_{35.0}Sn_{15.0}$ (A3), were prepared by an induction melting/casting method. Samples were spark-cut from ingots, heat-treated at 900 °C during 72 h in argon and quenched into cold water. The thermomagnetization curves revealed pre-martensitic ($T_I \approx 200$ K) and martensitic ($T_m \approx 130$ K) transformations as well as Curie temperature ($T_C = 366$ K) for A1. The alloy A2 exhibited MT at $T_m \approx 290$ K and $T_C = 355$ K while A3 showed $T_m \approx 230$ K and $T_C = 318$ K (for austenite). According to neutron and X-ray diffraction data, the martensitic structure was tetragonal with $c/a < 1$ for A1 [13], tetragonal with $c/a > 1$ for A2 (unpublished), and 4-layered orthorhombic for A3 [14].

Magnetostriction measurements have been performed in slabs of 3 mm × 5 mm × 1 mm cut out of different parts of the ingots, by means of two strain gauges (one was cemented on the sample, the other one served as reference) using a magnetic platform of Cryogenic Ltd. CFMS in the temperature range from 100 to 320 K and in fields up to 5 T. In-plane strains, both parallel and perpendicular to the applied field, were recorded as a function of field and temperature.

3. Results and discussion

$Ni_{49.7}Mn_{24.3}Ga_{26.0}$: Fig. 1 shows the parallel deformation of sample A1, under cooling–heating ramps, in the vicinity of martensitic transformation measured sequentially under 1 T and zero of applied field. When cooling under the applied field (FC), a rather large contraction (≈0.25%) occurs at the MT temperature as a consequence of the preferential orientation growth of the martensitic variants. This behaviour can be explained within the magnetoelastic model of ferromagnetic martensite (see [1,2,6] and references therein). Due to the strong magnetoelastic coupling, the application of a magnetic field creates an equivalent compressive stress in the cubic phase. Although not very high (few MPa), this magnetostress obviously affects the process of nucleation/growth of martensitic plates during cooling through MT, in the same way as the uniaxial mechanical compression does. The relatively small strain in this case is easily accommodated elastically so, the strain obtained is normally recoverable during heating across the reverse MT, which is seen in Fig. 1. The zero field (ZFC) curves do not show a significant change of the sample length during MT as martensitic variants tend to accommodate the strain to maintain the shape of the sample. The small maximum on the ZFC shape variation at MT can be explained by the minimum of elastic modulus accompanying MT and some internal tensile stresses in the sample produced, e.g., by gauge gluing. Perpendicular FC leaves the ZFC case unaffected. A small contribution from spontaneous volume contraction of the sample A1 at the MT, typically about 0.06% in the low-temperature Ni—Mn—Ga alloys [15], is presumably masked by the shape change and can be neglected. The negative slope in the high-temperature phase is due to an uncorrected instrumental drift.

Fig. 2 shows representative magnetostriction curves recorded in parallel magnetic field at different temperatures, corresponding to different phases of the alloy A1. The magnetostriction is negative, with a magnitude at saturation between 30 and 70 ppm. The curves show appreciable hysteresis below the martensitic transformation at 180 K. In the martensitic state, curves do not seem to manifest any MFIS contribution (thus implying pure rotation mechanism), primarily, because of much larger elastic modulus and twinning stress than in any Ni—Mn—Ga single crystalline samples [16]. A possible volume magnetostriction effect in the martensite has to be ruled out, as the evolution of the strain under the applied field does saturate whereas the volume magnetostriction should grow linearly with the field. The Inset to Fig. 2 shows large temperature variation of the absolute value of magnetostriction in saturating field in the vicinity of martensitic and premartensitic transformations which is also observed in the single crystalline Ni_2MnGa and explained by the elastic modulus behaviour [12].

$Ni_{50.1}Fe_{18.6}Co_{4.1}Ga_{27.2}$: The measurements on this alloy were hindered by its enhanced brittleness stemming from a particular grain structure. The samples exhibited a columnar grain structure with grains of nonuniform shape and approximate dimensions of about 0.5 mm in cross-section and few millimetres in length. Such a structure was prone to intergranular cracks, both in as-received state and after thermal cycling through MT. Some preliminary results are worth showing taking into account the lack of similar studies on this alloy system.

The cooling–heating behaviour of the parallel strain at MT in sample A2 is shown in Fig. 3. The strain gauge was glued along the sample texture. Much higher peaks than in the case of alloy