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chapter 1

GIANT MAGNETOSTRICTIVE MATERIALS

O. SÖDERBERG, A. SOZINOV, Y. GE, S.-P. HANNULA
and V.K. LINDROOS

*Laboratory of Materials Science, a member laboratory of the Center for New Materials,
Helsinki University of Technology, Helsinki, Finland*

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1. Introduction

When a magnetostrictive material is set in the magnetic field it strains while its magnetic moments reorientate. The present review focuses on the ferromagnetic Magnetic Shape Memory (MSM) alloys with a giant magnetic-field-induced strain (MFIS). MFIS is based on the rearrangement of the martensite twin variants by the twin boundary motion. This is a result of the interaction of the magnetic and crystal structure domains. The MSM alloys or Ferromagnetic Shape Memory Alloys (FSMA) have a ferromagnetic thermoelastic twinned martensite phase, high magnetocrystalline anisotropy and highly mobile twin boundaries. These materials have potential for actuator and sensor applications since they combine a large strain with rather high frequencies without a temperature change. The obtainable strains are greater than those of magnetostrictive, piezoelectric or electrostrictive materials.

Ullakko (1995, 1996a) presented the idea of redistribution of twin variants by magnetic field, i.e. MSM behaviour in martensitic phase. Experimental confirmation showed a 0.2% MFIS in an unstressed single crystal of Ni_2MnGa in 800 kA/m magnetic field applied along [001] at 265 K (Ullakko et al., 1996b, 1997). By applying the constantly increasing knowledge of the Ni-Mn-Ga system, the obtainable strain and the service temperature is raised and the applied magnetic field is reduced to some extent (Tickle and James, 1999; Tickle et al., 1999; Murray et al., 2000a, 2000b; Heczko et al., 2000; Sozinov et al., 2001a, 2001b, 2001c, 2002a, 2002b, 2003a, 2003b, 2004a). To achieve the maximum MFIS, the martensite should be in a single-variant state. In Ni-Mn-Ga alloys with the modulated five-layered martensite structure (5M) a MFIS of 6% is obtained. In alloys with a modulated seven-layered martensite structure (7M) the corresponding MFIS is 10%. The actuation of the 5M alloys is demonstrated with 2.5% strains up to 500 Hz (Henry et al., 2001, 2002a, 2002b, 2003a, 2003b; Tellinen et al., 2002; Marioni et al., 2003a, 2003b). The reverse application of the Ni-Mn-Ga MSM element as a sensor or a generator is studied by Müllner et al. (2003a) and Suorsa et al. (2004).

The possibility for MFIS in other than Ni-Mn-Ga alloys is also studied in several alloy systems with thermoelastic martensite transformation (James and Wuttig, 1998; Furuya et al., 1998, 1999; Kakeshita et al., 2000, 2003; Yabe et al., 2000; Oikawa et al., 2001a, 2001b, 2002, 2003; Wuttig et al., 2001, 2002; Craciunescu et al., 2002a, 2002b, 2002c; Kakeshita and Fukuda, 2002, 2003; Morito et al., 2002, 2003; Zhou et al., 2002a, 2003; Inoue et al., 2003; Karaca et al., 2003; Sato et al., 2003; Li et al., 2004a, 2004b; Fukuda et al., 2004; Efstathiou et al., 2004). Some of the alloys apply the actual MSM phenomenon, but in most of them the idea is to control the stress-induced martensite (SIM) formation by the magnetic field. SIM based strain is also possible in Ni-Mn-Ga (Kokorin et al., 1992; Martynov and Kokorin, 1992; Chernenko et al., 1995a, 1998a; Vasil'ev et al., 1999; Gonzàlez-Comas et al., 1999; Inoue et al., 2000; Li et al., 2004a, 2004b; Heczko and Straka, 2004b).

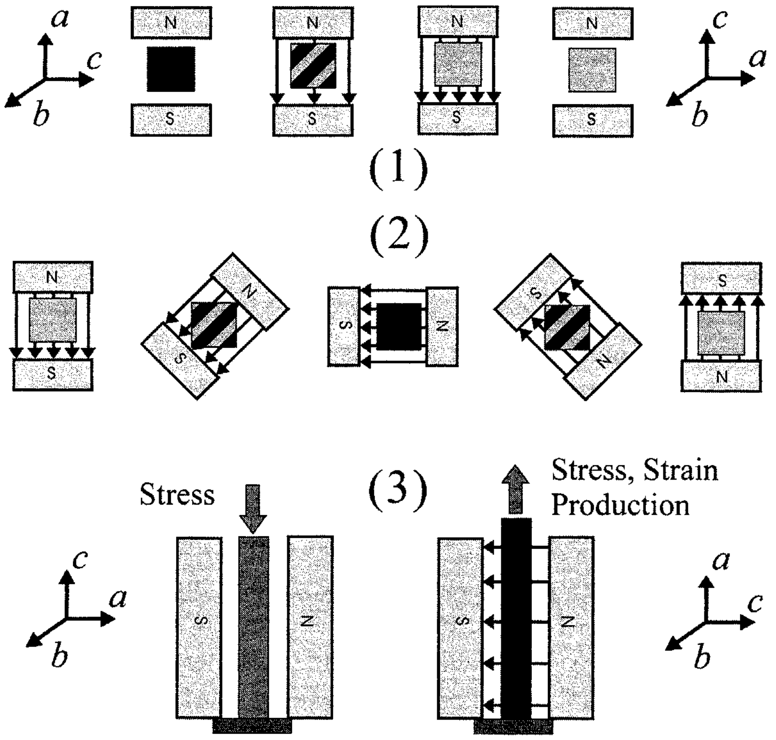


Fig. 1.1. A MSM element in (1) a unidirectional and (2) a rotating magnetic field; and (3) in the actuator. a, b and c are the crystallographic axes of martensite ($c < a, b$); c-axis is the easy axis of magnetization. (Courtesy of Söderberg et al., 2005, reproduced by permission of Elsevier Science).

1.1. Magnetic shape memory effect

The magnetic shape memory effect (MSME) is demonstrated with a Ni-Mn-Ga MSM alloy in the magnetic field (fig. 1.1). In the 5M and 7M martensite the shortest crystallographic c-axis is also the easy axis of magnetization. Because the easy directions of magnetization are different in the adjoining twins, the applied magnetic field creates a difference in energy of the variants (O'Handley et al., 2000; O'Handley and Allen, 2001; Vasil'ev and Takagi, 2004; Kiang and Tong, 2005). This energy difference is a driving force for the growth of those twin variants that are favourably oriented to the applied field. The structural change occurs via twin boundary motion. Those martensite variants with c-axis along the magnetic field become dominant and the material contracts in the direction of the applied field. The shape change remains after removing the magnetic field. Actuation is achieved if the magnetic field is turned perpendicular to its original orientation—for example with rotating the magnetic field—or with an external spring-back load. Then one single-variant martensite structure changes to another and generates the cycling of the giant MFIS (Söderberg et al., 2005).

1.2. Selected active materials

Active materials change their properties according to external impulses and they can work as actuators, sensors or simultaneously both. Such materials in addition to the MSM alloys are piezoelectrics (PZT), magnetostrictive materials (MS), shape memory alloys (SMA) and electroactive polymers (EAP). Piezoelectric materials are widely used in sensing and actuation, especially with high frequencies. They change dimensions in an electric field, or when loaded mechanically, they create an electric current (Muralt, 2001; Boller, 2001; Neurgaonkar, 2001; Quandt, 2001). Piezos can be polymers—such as PVDF—or ceramics. The most utilized is Pb-Zr-Ti (PZT) with 0.16% strain. With single crystals of $Pb(Zn_{1/3},Nb_{2/3})O_3$ (PZN), $Pb(Mg_{1/3},Nb_{2/3})O_3$ (PMN) and $PbTiO_3$ (PT) the strain can be much higher. Magnetostrictive materials change dimensions in a magnetic field (Guruswamy et al., 2000; Quandt and Claeyssen, 2000; Peuzin, 2001; Quandt, 2002). The new promising MS candidates are Fe-Ga-based alloys (Guruswamy et al., 2000; Clark et al., 2003; Kumagai et al., 2004), but the most applied industrial MS material is Terfenol D ($Tb_{0.3}Dy_{0.7}Fe_2$) operating at ambient temperature up to 20 kHz with 0.2%–0.24% strain in 40 kA/m magnetic field (Clark, 1980; Clark et al., 1992; Clark, 1994; O'Handley and Allen, 2001). With the shape memory alloys (SMAs) one can obtain large strains and good stress output with changing temperature, but in actuation the frequencies are smaller than with other active materials due to the lengthy cooling process (Duerig, 1990; Otsuka and Wayman, 1998; Friend, 2001; Quandt, 2001). The most applied SMAs are Ni-Ti-, Cu-Zn-Al- and Fe-Mn-Si-based alloys. Some of these materials have in austenitic state a superelastic behaviour just above the martensite transformation temperature where stress induced martensite can form. Also, electroactive polymers and ionic polymeric–metal composites (IPMCs) have great potential as soft robotic actuators, artificial muscles, and dynamic sensors in micro-to-macro size range (Shahinpoor, 2003).

2. Modelling the behaviour of MSM materials and the giant magnetic-field-induced strain (MFIS)

Modelling the behaviour of MSM materials in simultaneously applied stress and magnetic field is complicated since these materials consist of multidomain crystallographic and magnetic subsystems and their behaviour is affected by the interaction between these subsystems. The first-principles numerical calculations have been applied to reveal the microscopic origin of the lattice instabilities and the phase transformations in Ni-Mn-Ga alloys (Ayuela et al., 1999, 2002; Godlevsky and Rabe, 2001; Bungaro et al., 2003; Zayak et al., 2003; Zayak and Entel, 2004, 2005; Wan et al., 2005a, 2005b), their magnetocrystalline anisotropy (Enkovaara et al., 2002a), and some particularities in the magnetic subsystem (Enkovaara et al., 2002b, 2003, 2004). The low temperature phase diagram for Ni-Mn-Ga was calculated based on the phenomenological theory of phase transformations in ferromagnets (review by Vasil'ev et al., 2003). The premartensitic phenomenon and the intermediate phase in it are reviewed by Planes and Mañosa (2001). The general scheme considering the multistage structural transformation with the modulated phases is suggested by Castán et al. (2003). Chernenko et al. (2004a) and Hirsinger et al. (2004) have been modelling the stress-induced martensitic transformation.

According to the first theoretical considerations of the large magnetically induced strain, the total energy density of the ferromagnetic martensite in a magnetic field contains the Zeeman terms $-\boldsymbol{H} \cdot \boldsymbol{m}_i$ (Ullakko et al., 1996b; James and Wuttig, 1998). Here $\boldsymbol{H}$ is the magnetic field and $\boldsymbol{m}_i$ the magnetic moment of the martensite variant i with changing value in the differently oriented variants. High magnetocrystalline anisotropy leads to

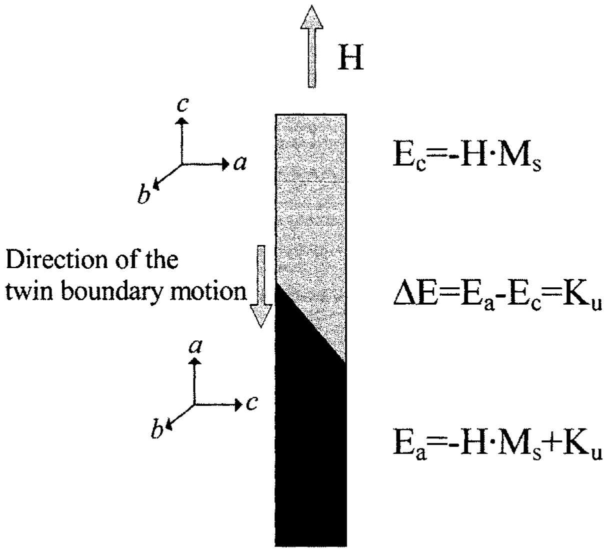


Fig. 2.1. Limitation of the magnetic field-induced driving force of the twin boundary motion. The difference in the magnetic energy density of the twin variants is independent of the magnetic field value at high field, since the magnetic moments of martensitic variants are oriented along the field. The deviation of a few degrees from the 90° between the c-axis inside the variants is neglected.

large differences in Zeeman energies and, thus, may promote the redistribution of martensite variants, when twin boundaries move easily. O'Handley's model (1998) takes into account the magnetization rotation. Due to it, the different martensite variants saturate at a higher field than the anisotropy field, $2K_u/M_s$ (K_u—the uniaxial magnetic anisotropy constant, M_s—saturation magnetization). Consequently, the driving force for the martensite variant redistribution by the magnetic field is restricted (fig. 2.1). James et al. (1999) also acknowledged the importance of magnetization rotation. O'Handley's model has been the basis for the later models of the MSM materials in the alternating magnetic field (Henry et al., 2002b; Marioni et al., 2003a).

Likhachev and Ullakko (2000a, 2000b) pointed out the importance of the hysteretic mechanical behaviour in the stress-induced martensite variant rearrangement. The mechanical stress–strain relationship has been extended with the magnetic stress $\sigma_{\mathrm{mag}}(H)$ when the magnetic field is applied. The Zeeman, magnetostatic and magnetic anisotropy energies are all included. The saturation value is predicted to be $\sigma_{\mathrm{mag}}(H) = K_u \cdot \varepsilon_0^{-1}$, where $\varepsilon_0 = (1 - c/a)$, and a and c are the lattice parameters of the tetragonal phase. With this model the magnetic stress $\sigma_{\mathrm{mag}}(H)$ and deformation $\varepsilon(\sigma_{\mathrm{mec}}, H)$ for the two-variant martensite structure can be calculated from the experimental mechanical testing data $\varepsilon_{\mathrm{mec}}(\sigma_{\mathrm{mec}})_{H=0}$ and the field dependencies of magnetization along easy and hard directions in the single-variant state (Likhachev et al., 2004a). Thus, the large MFIS is possible only in materials where the maximal magnetic stress exceeds the zero-field twinning stress σ_{tw}, i.e. $K_u \cdot \varepsilon_0^{-1} > \sigma_{\mathrm{tw}}$.

The approach of Hirsinger and Lexcellent (2003a, 2003b) and Creton and Hirsinger (2005) is based on a thermodynamic model of irreversible process with internal variables. The magnetic subsystem is non-linear reversible, while the mechanical response is decomposed to reversible and irreversible parts. Bogdanov et al. (2003) have applied the phenomenological theory of magnetoelastic interactions to describe the two-variant tetragonal martensite in a magnetic field. The elastic energy for tetragonal lattice and the magnetostatic together with the magnetic anisotropy energies are included. The field and stress dependencies of strain are quantitative. Experimental data on elastic constants in the martensitic phases is important for the model. This model can be improved by including the mechanical hysteresis in it.

The Landau approach is widely used for modelling the structural and magnetic phase transitions (Vasil'ev et al., 2003). Buchel'nikov et al. (2001, 2005) and L'vov et al. (2002) apply it to the magnetic field-induced strain in MSM materials. The L'vov model was developed further by taking into account the statistical nature of the obstacles for twin boundary motion (Glavatska et al., 2003b). It was also applied to explain the different Curie temperatures of cubic and martensitic phases as well as the magnetic anisotropy properties of the tetragonal martensites in Ni-Mn-Ga and the mechanical behaviour of the 5M in a constant magnetic field (Chernenko et al., 2003a, 2003b, 2004b, 2004c). The main difference between the others and the model of L'vov is that the normal magnetostriction is considered responsible for the giant MFIS. All other above-mentioned models neglect the normal MS deformation.

The microscopic details concerning the behaviour of MSM materials are proposed only by Paul et al. (2003) and Müllner et al. (2003b). According to Paul et al. the twin boundary motion is controlled by the motion of the magnetic domain walls and the model analyses the region close to the twin boundary in the micro-magnetic scale. Müllner et al. explain the MFIS of martensite by the effect of the magnetic force on the dislocations and, therefore, the magneto-mechanical hysteresis is explained with a microscopic model dealing with twinning dislocations, their mutual interaction and interaction with interfaces.

3. Ni-Mn-Ga alloys

The structural, magnetic and mechanical properties of the Ni-Mn-Ga alloys—the currently best performing MSM materials—are highly sensitive to the chemical composition and temperature. Their phase transformations, magnetic properties, behaviour in the magnetic field and some selected physical and chemical properties are introduced here.

3.1. Martensitic and reverse phase transformations

The high temperature parent phase of near-stoichiometric Ni_2MnGa alloys has the highly ordered $L2_1$ cubic structure with lattice parameter $a = 0.576–0.597$ nm depending on the alloy composition and the external temperature (Webster et al., 1984; Chernenko et al., 1995b; Brown et al., 1999; Inoue et al., 2000; Ge et al., 2003). The degree of atomic order effects the structure and the magnetic properties. The ordering can be changed, for example, by fast cooling from homogenisation temperature resulting in 100 K lower martensitic transformation temperature (Kreissl et al., 2004). The dendritic microstructure formed during casting of Ni-Mn-Ga alloys can be homogenized if the annealing temperature is high enough and the duration of the heat treatment long enough—too low temperature and too short time do not have the desired effect (Gupta et al., 2004; Pirge et al., 2004). A variety of martensite crystal structures have been observed in Ni-Mn-Ga: non-modulated (marked with T or NM in references), and modulated, so-called, 5-layered (5M), 7-layered (7M), 8-layered (8M) or 10-layered (10M) (Martynov and Kokorin, 1992; Chernenko et al., 1998b; Pons et al., 2000; Lanska et al., 2004). Modulation is observed as extra diffraction maxima between the fundamental spots in $\langle 110\rangle^*$ direction of reciprocal space and the crystal structure can be interpreted either as a long period stacking of closed-packed planes $(1\underline{1}0)$ (Pons et al., 2000) or a periodic shuffling of basal planes (110) along $[1\bar{1}0]$ (Martynov and Kokorin, 1992). When the crystal lattice is presented in the cubic parent phase co-ordinates the ratio of the lattice parameters (of the basic or average lattice) in the 5M and 7M structures is $c/a < 1$ and in the NM structure $c/a > 1$. The premartensitic or intermediate structure appearing in certain Ni-Mn-Ga alloys before the martensitic transformation (Cesari et al., 1997; Kokorin et al., 1997; Planes et al., 1997; Khovailo et al., 2001; Vasil'ev et al., 2003; Seguí et al., 2005) may have relevance in the formation of the modulated phases, but it is not discussed in detail here, since this presentation focuses on the behaviour of the martensitic phases.

The stoichiometric Ni_2MnGa transforms from the $L2_1$ phase (the parent phase P) into a martensite structure approximately at 200 K (Webster et al., 1984; Ooiwa et al., 1992; Pons et al., 2000; Brown et al., 2002). In the off-stoichiometric Ni-Mn-Ga alloys the martensitic transformation is highly composition dependent and occurs at temperatures below 630 K (Chernenko et al., 1995b, 1999; Mañosa et al., 1999; Lanska et al., 2004). The martensite transformation temperatures M_s together with the Curie points T_c are mapped according to the valence electron concentration in fig. 3.1 (see for example Chernenko, 1999; Tsuchiya et al., 2001; Lanska et al., 2004). Ternary Ni-Mn-Ga alloys having their Curie points approximately at 370 K transform to the 5M structure at 343 K at the highest, while for the 7M alloys the highest reported martensite start temperature (M_s) is 356 K (Lanska et al., 2004). Since the MSM occurs in certain martensite structures, the intermartensitic reactions limit its lowest service temperature. Intermartensitic transformations may occur thermally or stress induced (Martynov and Kokorin, 1992; Chernenko et al., 1995a, 1997; Wang et al., 2001; Heczko et al., 2002a; Soolshenko et al., 2003; Seguí et al., 2003; Dai et al., 2004; Khovailo et al., 2004a; Sozinov et al., 2004b; Söderberg, 2004). The final structure is usually the NM phase and the transformation sequence depends on the alloy composition. It occurs via modulated structures, for example P → 5M → 7M → NM or P → 7M → NM. Attempts to increase the transformation temperatures and the Curie point, i.e. rise the service temperature, as well as suppressing the intermartensitic reactions are carried out by alloying with quaternary elements (Kokorin et al., 1989; Tsuchiya et al., 2000a; Liu et al., 2002a, 2002b; Cherechukin et al., 2004; Khovailo et al., 2003, 2004b; Lu et al., 2003; Yamaguchi et al., 2003; Kikuchi et al., 2004; Koho et al., 2004; Söderberg et al., 2004a; Tsuchiya et al., 2004).

3.2. *Mechanical properties*

The crystal structure has a remarkable influence on the mechanical properties of the Ni-Mn-Ga alloys. Mechanical behaviour is relevant for the possibility of MSM effect and obtainable strains.

Three elastic constants of the Ni-Mn-Ga cubic parent phase C_{11}, C_{12} and C_{44} are calculated from the stiffness constants measured in different crystal directions with the

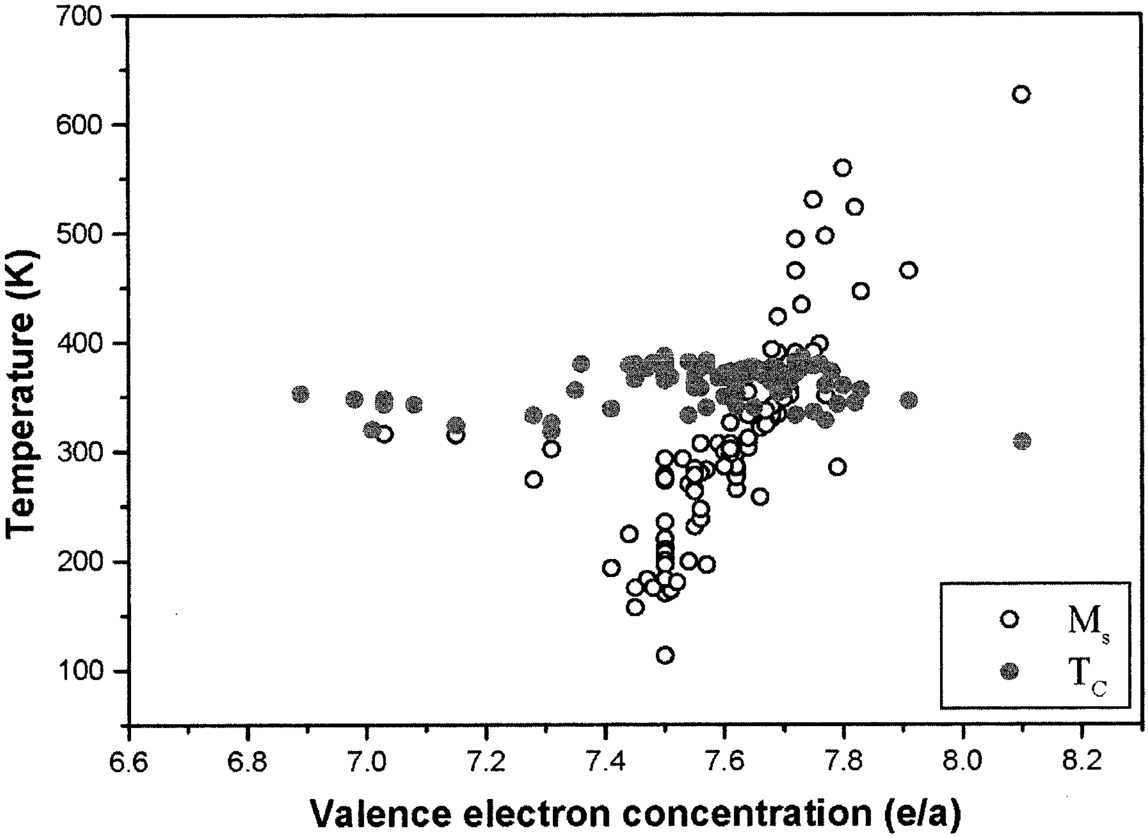


Fig. 3.1. The martensitic transformation temperatures and the Curie points vs. the valence electron concentration in Ni-Mn-Ga. References: Albertini et al. (2001a, 2001b), Aliev et al. (2004), Brown et al. (1999, 2002, 2004), Cesari et al. (1997), Chernenko (1999), Chernenko and Vitenko (1994), Chernenko et al. (1994, 1995a, 1995b, 1997, 1998a, 1998b), Elfazani et al. (1981), Ezer et al. (1999), Fritsch et al. (1994), Ge et al. (2002), Glavatska et al. (2002, 2003a), Gonzàlez-Comas et al. (1999), Inoue et al. (2002), Heczko and Straka (2003), Heczko et al. (2001a, 2002a, 2002b), James et al. (1999), Kokorin et al. (1992, 1996), Kudryavtsev et al. (2002), Lanska et al. (2004), Ma et al. (2000), Mañosa et al. (1997), Martynov (1995), Martynov and Kokorin (1992), Matsumoto et al. (1999), Mogylnyy et al. (2003), Murray et al. (1998), Obradó et al. (1998), Ooiwa et al. (1992), Pakhomov et al. (2001), Park et al. (2003), Pasquale et al. (2002), Planes et al. (1997), Pons et al. (2000), Shanina et al. (2001), Sozinov et al. (2001a), Stenger and Trivisonno (1998), Stuhr et al. (1997, 2000), Tickle and James (1999), Tickle et al. (1999), Tsuchiya et al. (2000b, 2003a, 2003b), Ullakko et al. (1996b, 1997), Wang et al. (2001, 2002), Webster et al. (1984), Wirth et al. (1997), Zasimchuk et al. (1990), Zheludev et al. (1995a, 1995b), Zuo et al. (1998).

ultrasonic continuous-wave method and show softening of $C' = 1/2(C_{11} - C_{12})$ in the vicinity of the structure phase transformation (Worgull et al., 1996; Planes and Mañosa, 2001; Stipcich et al., 2004). This indicates on the lattice instability to shear wave propagated in [110]. For the martensitic tetragonal phases due to the lower symmetry the amount of independent elastic constants is six. Experimental studies of it in Ni-Mn-Ga have been recently started (Dai et al., 2003, 2004). It is important to mention that a low shear elastic constant was found in the 5M phase (9 GPa) as well as in the cubic phase (7 GPa). The rhombohedral constants C_{44} and C_{66} are close to each other, 51 GPa and 49 GPa (Dai et al., 2003). The temperature dependence of the elastic constants C' and C^* in a magnetic field are plotted in fig. 3.2 from above the Curie point down to 200 K for the

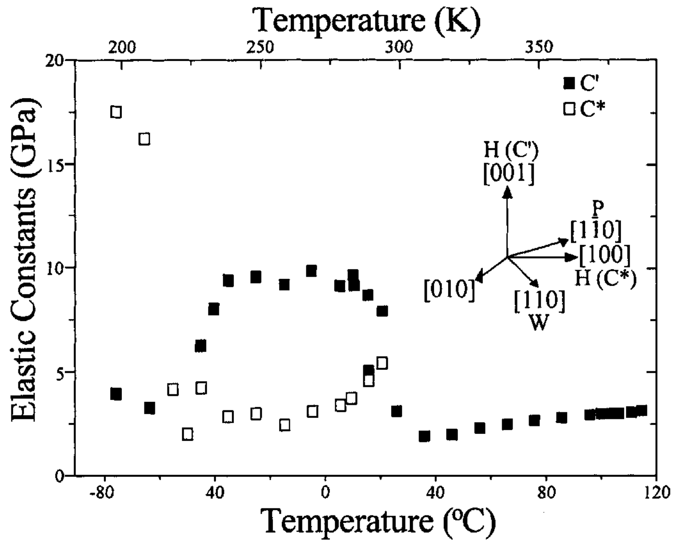
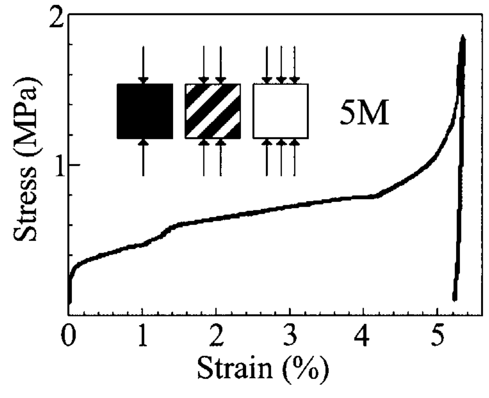


Fig. 3.2. The elastic constants C' and $C^* = (a+b)^{-1}[cC_{44}+a(C_{33}-C_{44}-C_{13})]$, where $a = C_{11}-2C_{44}-C_{13}$, $b = C_{11} - 2C_{66} - C_{12}$ and $c = C_{33} - 2C_{44} - C_{12}$, of the alloy $Ni_{50}Mn_{28.4}Ga_{21.6}$ in the temperature range of 200–380 K (above the Curie point). C' was determined with magnetic field along [001] and C^* with the magnetic field parallel to [100]. In the austenite (above 318 K) the two modes are degenerate. (Courtesy of Dai et al., 2004, reproduced by permission of American Institute of Physics).

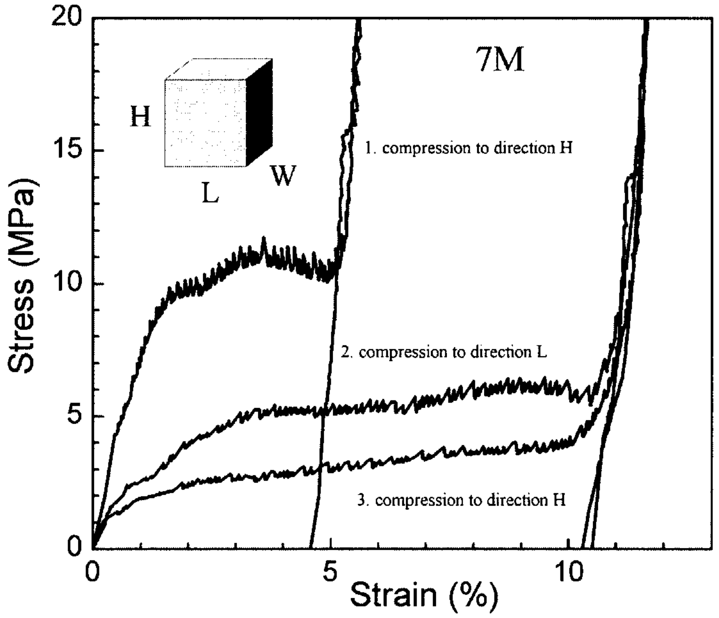
single-variant sample of the alloy $Ni_{50}Mn_{28.4}Ga_{21.6}$ (Dai et al., 2004). Here, the upper mode C' describes the stiffness of the tetragonal distortion in the xy plane, while the lower C^* represents physically the stiffness for the further tetragonal distortion in the xz or yz plane.

In Ni-Mn-Ga alloys the hydrostatic pressure stabilizes the parent phase (Kanomata et al., 1987), while the shear stress favours the martensite and causes the stress-induced martensite (SIM) formation (Kokorin et al., 1992; Martynov and Kokorin, 1992; Chernenko et al., 1998a, 2003b; Gonzàlez-Comas et al., 1999). Stresses of 40–80 MPa or higher give approximately a 4% strain connected to the SIM formation in the parent phase; furthermore, if the intermartensitic reactions are involved, strains may be 6–10% (Martynov and Kokorin, 1992). SIM formation is possible also in a high magnetic field, for example a field of 10^4 kA/m at 1 K above the martensite start temperature (Kakeshita et al., 1999; Vasil'ev et al., 1999; Gonzàlez-Comas et al., 1999; Inoue et al., 2000; Cherechukin et al., 2001, 2003). Training in the magnetic field increases the obtainable shape change in such a transformation (Cherechukin et al., 2003).

The thermally formed martensite has a multi-variant structure. The rearrangement of the twins occurs when sufficient external stress is applied. This is observed as a stress plateau in the stress–strain-curve (Otsuka and Wayman, 1998; Murray et al., 1998; O'Handley and Allen, 2001). At the end of the stress plateau, the twin structure of the martensitic phase is simpler. The single-variant state in the 5M structure can be obtained with a single compression and, the twinning stress, σ_{tw}, needed for variant reorientation can be less than 1 MPa (fig. 3.3a). The twinning stress of the 7M structure can be decreased close to 1 MPa by the pre-straining including three compression-cycles to two different crystallographic orientations (fig. 3.3b, Sozinov et al., 2004a). The single variant state of the NM structure can be obtained with three successive compressions to the three crystallographic



(a)



(b)

Fig. 3.3. The martensite variant rearrangement in the Ni-Mn-Ga alloys in compression. (a) 5M-, (b) 7M- and (c) NM-martensite. (Courtesy and compilation from Heczko and Straka, 2003 and Sozinov et al., 2003b, 2004a, 2004b, reproduced by permission of American Institute of Physics).

orientations or by tensile-compressive cycling. The lowest observed twinning stress for NM martensite is about 6 MPa, which is much higher than the one required in MSME (fig. 3.3c, Sozinov et al., 2004b; Söderberg et al., 2004b). The twinning stress decreases with increasing temperature (Heczko et al., 2002b; Koho et al., 2003; Heczko and Straka, 2003; Sozinov et al., 2003a, 2004a, 2004b; Söderberg et al., 2004b). The maximum obtainable strain connected to a certain single variant martensite structure depends on the crystallographic distortion of the structure and can be calculated as $\varepsilon_0 = 1 - c/a$, where c and a are given in the parent phase coordinates (O'Handley and Allen, 2001). This grants for the maximum strains approximately 6% in the 5M-, 10% for the 7M- and 20% for the NM-structure.

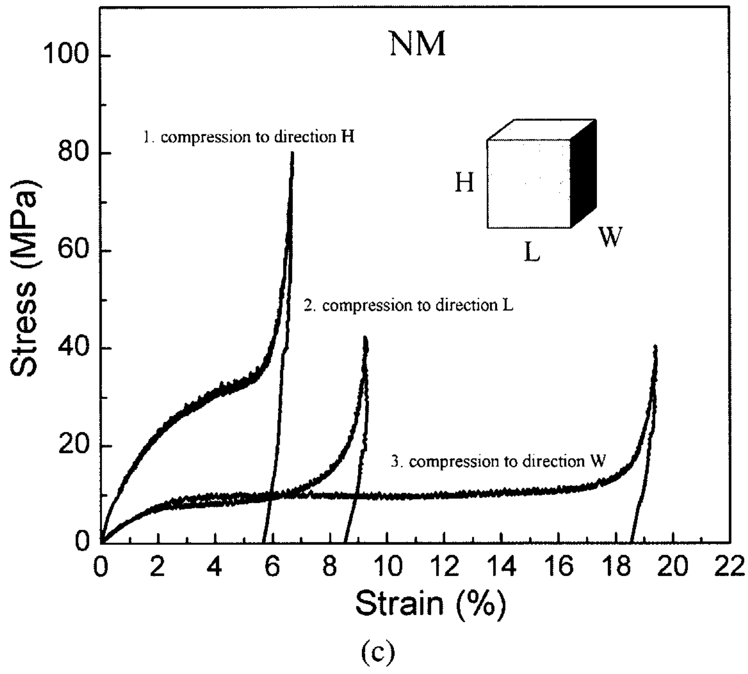


Fig. 3.3. (*Continued.*)

3.3. *Magnetic properties*

The magnetic anisotropy as well as the magnetization of the material are important factors for the MSM effect. Also, it is important to notice the interaction of the twin domains and the magnetic domains of the structure.

Magnetocrystalline anisotropy has been studied in the Ni-Mn-Ga system experimentally both for the cubic phase and for the different martensites (Tickle and James, 1999; Likhachev and Ullakko, 2000b; Murray et al., 2000a; Heczko et al., 2000, 2002b; Sozinov et al., 2001b, 2002a; Straka et al., 2002, 2004; Straka and Heczko, 2003b; Heczko and Straka, 2003, 2004a). Also, the summarised experimental results for Ni-Mn-Ga martensitic phases are available (Sozinov et al., 2002c, 2003a; Heczko et al., 2003; Straka and Heczko, 2003a).

Magnetization curves at 300 K for the 5M, 7M and NM martensites measured in different crystallographic directions are shown in fig. 3.4. The magnetic anisotropy energy density is calculated as the area between the easy and the hard magnetization directions. For reliable results, the martensite must be in the single-variant state (Tickle and James, 1999). For the 5M martensite the easy axis of magnetization is its crystallographic short c-axis. For the uniaxial anisotropy constant K_u at ambient temperature values in the range of 1.2–2 × 10^5 J/m^3 have been reported, while the second anisotropy constant is negligible (Heczko et al., 2002b; Enkovaara et al., 2004). The big difference in the K_u values reflects probably the composition dependence (Albertini et al., 2002a; Heczko and Straka, 2004a, 2004b). Also, in the 7M martensite the shortest c-axis is the axis of easy magnetization (fig. 3.4c). The longest a-axis coincides with the axis of the hard magnetization and the magnetization of the b-axis is the intermediate one. Therefore, two magnetic anisotropy constants are needed to characterize this orthorhombic crystal structure. Their respective

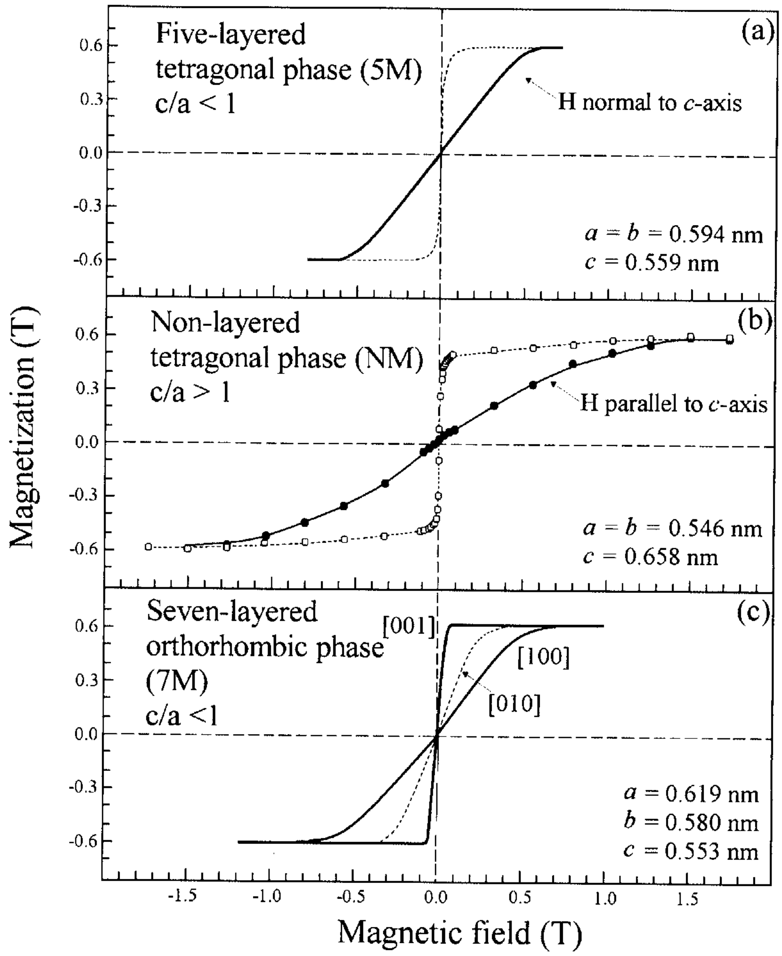


Fig. 3.4. Magnetization curves for different martensitic phases in the Ni-Mn-Ga system at 300 K. 1 Tesla corresponds with approximately 800 kA m^{-1}. (Courtesy and compilation from Sozinov et al., 2002c, reproduced by permission of Institute of Electrical and Electronics Engineers (IEEE)).

values (K_b, K_a) can be calculated from the magnetization data as the area cross-sections between curves for the c-axis and for the b- or a-axis respectively. $K_b = 0.7 \times 10^5$ J/m^3 and $K_a = 1.6 \times 10^5$ J/m^3 for $Ni_{48.8}Mn_{29.7}Ga_{21.5}$ and $K_b = 1.02 \times 10^5$ J/m^3 and $K_a = 1.74 \times 10^5$ J/m^3 for $Ni_{50.5}Mn_{29.4}Ga_{20.1}$ are established at ambient temperature (Sozinov et al., 2002a; Heczko et al., 2003). In the NM martensite the c-axis is the hard axis of magnetization and there is an easy plane of magnetization. This is connected to the different kind of tetragonal distortions of the NM phase ($c/a > 1$) as compared with the distortion of the 5M phase ($c/a < 1$). At ambient temperature the value of magnetic anisotropy energy density for the NM martensite is close to -2×10^5 J/m^3. The magnetic anisotropy increases, when the temperature decreases, as shown in fig. 3.5 (Heczko et al., 2002b; Heczko and Straka, 2003; Straka and Heczko, 2003b; Straka et al., 2004).

The saturation magnetization in the Ni-Mn-Ga system depends on composition and temperature (Jin et al., 2002; Takeuchi et al., 2003; Heczko and Straka, 2004a). Upon cooling the magnetization increases as shown in fig. 3.6, but during the structural phase transformations the magnetization shows an abrupt change (Webster et al., 1984; Marcos et al., 2002;

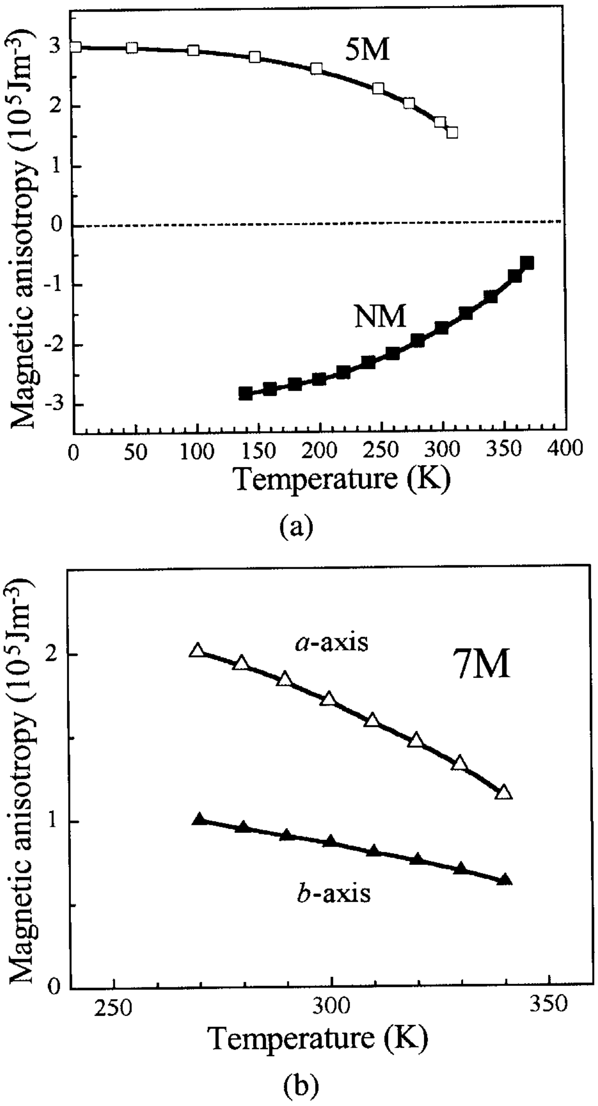


Fig. 3.5. Temperature dependences of magnetic anisotropy energy density of different martensitic phases in Ni-Mn-Ga. (Courtesy and compilation from Straka and Heczko, 2003b, reproduced by permission of American Institute of Physics).

Vasil'ev et al., 2003). In the Ni-Mn-Ga martensite the magnetic anisotropy is larger than in the cubic phase; therefore, the magnetization of the martensite at low fields is smaller than the one of the cubic phase (Tickle and James, 1999). In strong fields the saturation magnetization of martensite is higher than that of the cubic phase and this can be applied in the giant magnetocaloric effect (Marcos et al., 2002, 2003).

The martensitic transformation (*cubic* → 5M) of the single-crystalline Ni-Mn-Ga material produces a multi-variant structure, where the *c*-axes of the variants are close to the $[100]_P$, $[010]_P$ and $[001]_P$ directions of the parent cubic phase. According to Likhachev and Ullakko (2000a) when the magnetic field is applied along the $[100]_P$ direction and the saturation value in one martensite variant is reached, this variant is taken over by one large magnetic domain. A rising magnetic field increases the total magnetization of the structure by rotation of local magnetic moments inside other martensite variants. This explains

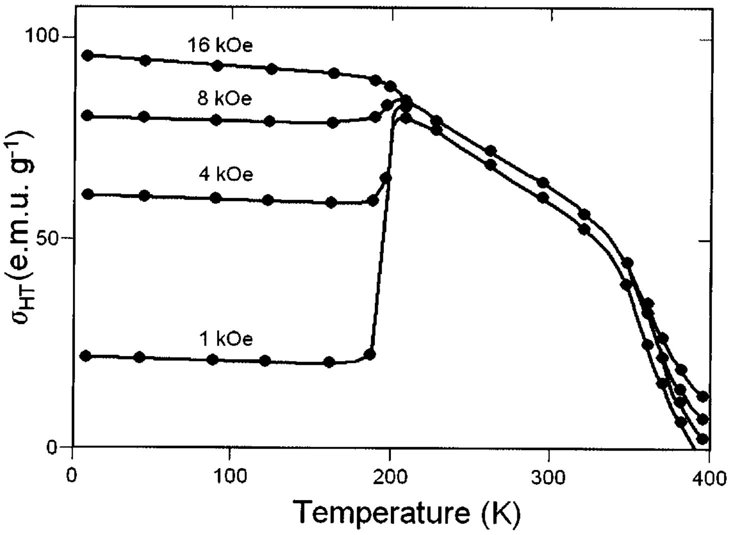


Fig. 3.6. Temperature dependencies of magnetization for Ni_2MnGa. The abrupt change at $T = 202$ K is connected with the structural cubic to tetragonal phase transformation. (Courtesy and compilation from Webster et al., 1984, reproduced by permission of Elsevier Science).

the kink (fig. 3.7) in magnetization data observed at the field value corresponding to the saturation field of the first variant (with *c*-axis along the field) reported by Ullakko et al. (1996b) for a single-crystal sample.

When there is no constraint and the twinning stress is low enough, the MFIS connected with twin boundary motion can be measured simultaneously with the magnetization data (fig. 3.8). It is shown by a jump in the first quadrant of the magnetization curve. The critical magnetic field for MFIS decreases when the magnetic anisotropy constant of the material increases or when the twinning stress decreases.

The martensite of the MSM alloy $Ni_{51.3}Mn_{24}Ga_{24.7}$ has a hierarchical magnetic domain structure of herringbone patterns, where 180° magnetic domains form laminas within the martensite variants (James et al., 1999). When magnetic fields up to 732 kA/m are applied during the martensitic transformation the fir-tree magnetic domain structure changes to one big domain (Pan and James, 2000). In the austenitic and premartensitic phase the 180° domain is dominant, while in the martensitic phase the magnetic domain coincides with the twin boundaries and the microtwins of the structure (DeGraef et al., 2001, 2003; Heczko et al., 2001a; Sozinov et al., 2001a; Park et al., 2003; Tsuchiya et al., 2003b; Grechishkin et al., 2004). The magnetization of the main variant may distort the internal twins into a zig-zag shape. (Ge et al., 2004). During the magnetic-field-induced twin boundary motion the magnetic domain is observed to superimpose upon the martensite twin domains (Chopra et al., 2000). In fig. 3.9 the 180° stripe domain follows the *c*-axis within one variant, the 90° domain wall coincides with the twin boundary when the *c*-axis is in plane in the adjoining variants and a labyrinth domain forms if the *c*-axis is out of plane. Here the macroscopic domain width is 5–40 μm. The Lorenz microscopy has given domain widths less than 0.1 μm, which may be due to the fact that magnetic domains might also have an internal structure. Sullivan et al. (2004a, 2004b) investigated the reconfiguration of the magnetic domains during the structural phase transformation of Ni-Mn-Ga by using a new magnetic transition spectrum method (Chopra and Sullivan, 2005).

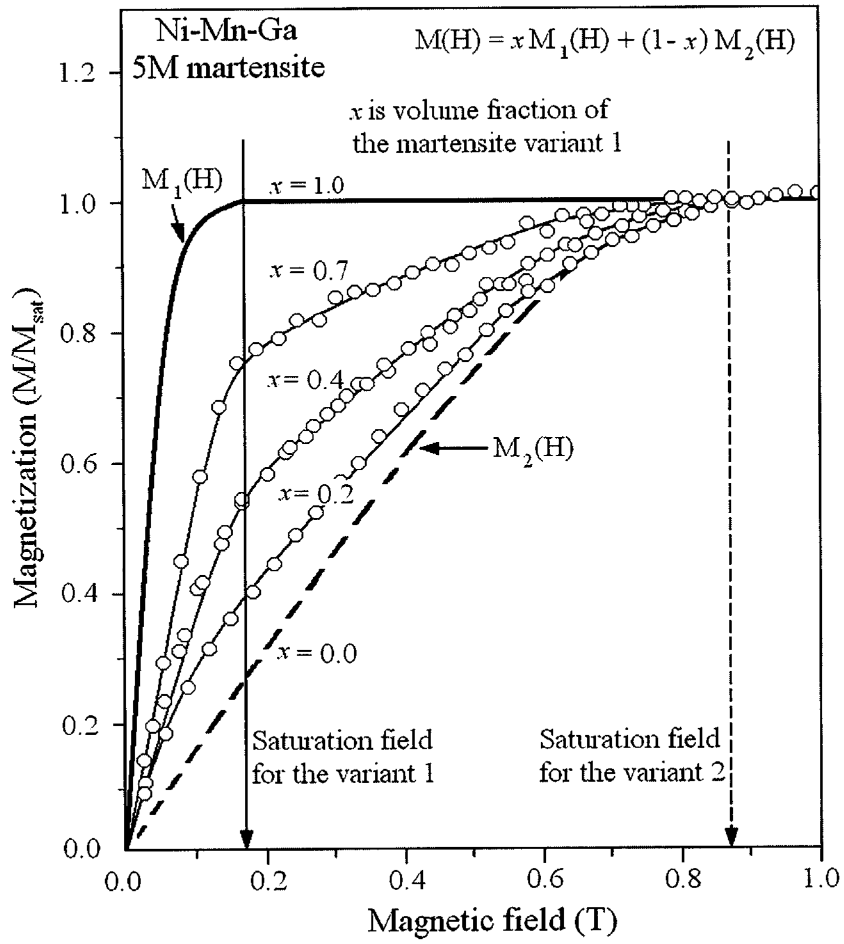


Fig. 3.7. Illustration of a simple additive model valid for the magnetization in the Ni-Mn-Ga 5M martensite. The magnetization curves of the two-variant constrained sample are measured along the *c*-axis of one variant. The *c*-axis of the second variant is perpendicular to the field direction within the accuracy of a few degrees. Due to the constraint, x is constant in the applied magnetic field. 1 Tesla corresponds with approximately 800 kA m^{-1}. (Courtesy and compilation from Likhachev et al., 2004a, reproduced by permission of Elsevier Science).

4. Martensite variant rearrangement in an applied magnetic field

Stress and magnetic field influence the MFIS of the MSM materials. Both the mechanical behaviour in a constant magnetic field and the field-induced strain under a constant stress are considered here.

The material parameters of the MSM alloys obtained in mechanical testing with a constant magnetic field can be applied for the device design. The mechanical behaviour of the 5M martensite in a constant magnetic field is studied by Jääskeläinen (2001), Straka and Heczko (2003a, 2005), Müllner et al. (2003b), Chernenko et al. (2004a, 2004b, 2004c), Suorsa and Pagounis (2004) as well as by Likhachev et al. (2004b). Figure 4.1 shows schematically the mechanical behaviour of a 5M single-variant sample, when the compressive stress is applied along the [100] direction (*a*-axis) and the magnetic field to the [001] direction (*c*-axis). Since the long *a*-axis is the hard axis of magnetization, growth of the twin variant, which has the *a*-axis along the field direction, is retarded. Therefore,

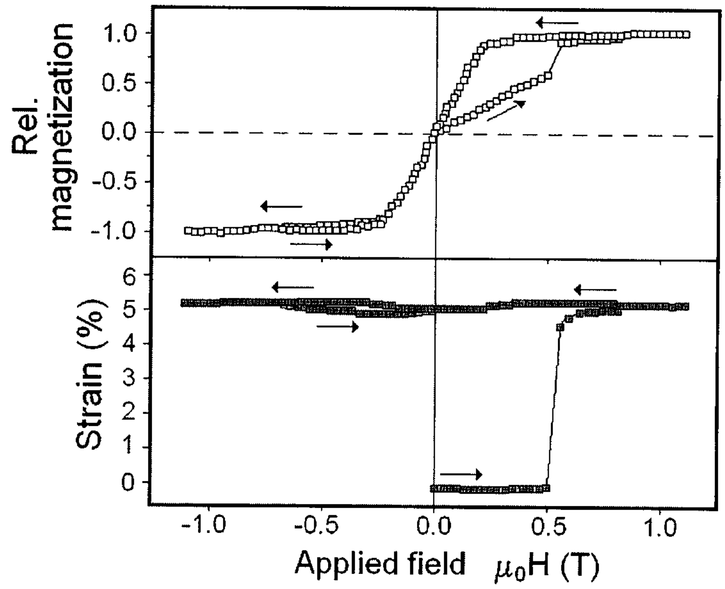


Fig. 3.8. The magnetization and straining of a single-variant 5M material as a function of the magnetic field. Initially the field is applied along the *a*-axis. An abrupt change in magnetization and strain at a moderate field is due to the twin variant redistribution. 1 Tesla corresponds with approximately 800 kA m^{-1}. (Courtesy and compilation from Heczko et al., 2002b, reproduced by permission of American Institute of Physics).

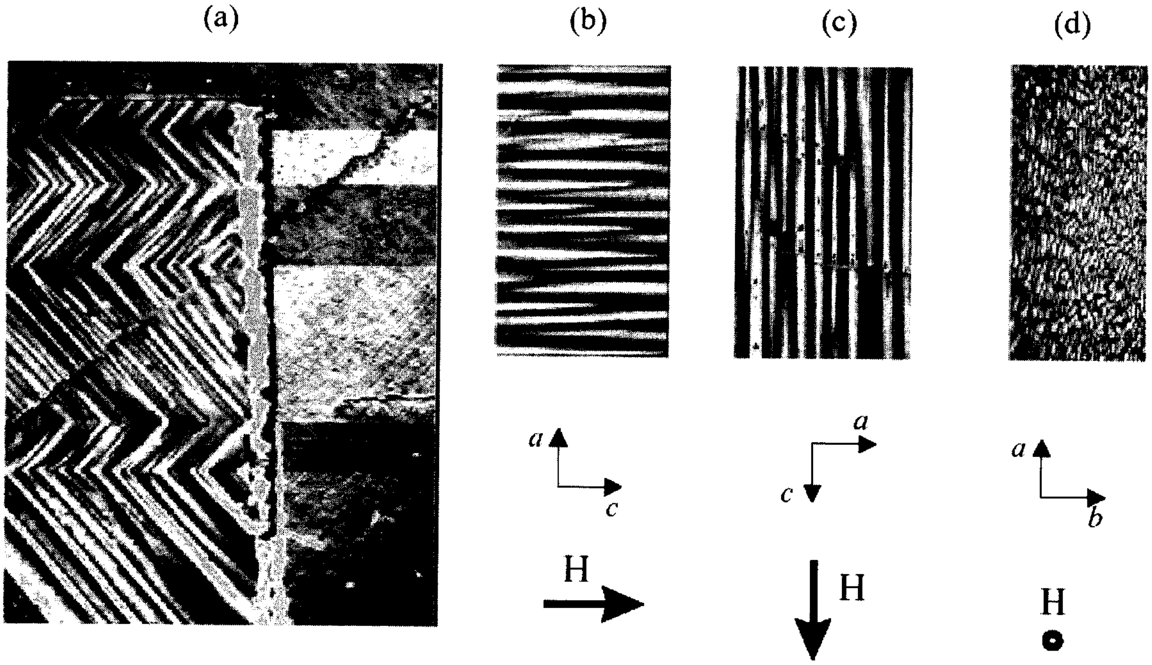


Fig. 3.9. (a) The structure of a two-variant 5M martensite structure as seen in a magneto-optic image (MO) at *left* and an optical image at *right*. The MO-images of a single variant 5M martensite structure obtained in a magnetic field applied along (b) the horizontal direction, (c) the vertical direction and (d) perpendicular to the plane of observation. The H—direction of the magnetic field, which was applied and removed after the reorientation of the crystal (*a*, *b* and *c*—crystallographic axes of martensite lattice). (Courtesy of Sozinov et al., 2001a, reproduced by permission of EPD Sciences).

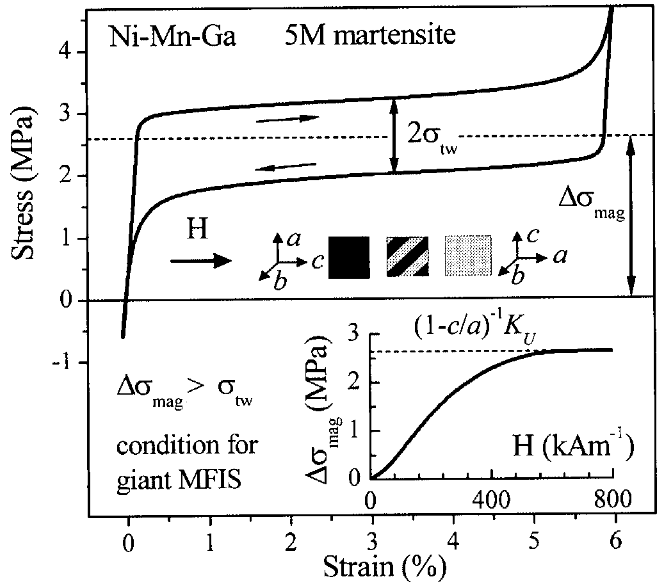


Fig. 4.1. The magnetically assisted pseudo-elastic behaviour of a 5M martensite single-variant sample in compression at the constant magnetic field $H = 800\,\mathrm{kA\,m^{-1}}$. Inserts show the field dependence of $\Delta\sigma_{\mathrm{mag}}$ and the crystallographic reorientation by the twin boundary motion (change between axes $a \leftrightarrow c$, b-axis is common for both variants). The reorientation is illustrated schematically by the sample colour (black → gray during loading; gray → black during unloading). (Courtesy of Söderberg et al., 2005, reproduced by permission of Elsevier Science).

more stress (shown as $\Delta\sigma_{\mathrm{mag}}$ in fig. 4.1) is needed in the magnetic field to obtain the same deformation as in the mechanical testing without the field. This addition, $\Delta\sigma_{\mathrm{mag}}$, is the magnetically induced stress. It increases with H and has the saturation value of $\Delta\sigma_{\mathrm{mag}} = (1 - c/a)^{-1}K_{\mathrm{u}}$. If the mechanical deformation is carried out without the magnetic field, the 6% strain would remain in the 5M structure after unloading. However, in the magnetic field, if the saturation value of $\Delta\sigma_{\mathrm{mag}}$ exceeds the twinning stress (σ_{tw}), the material behaves pseudo-elastically and the obtained shape change recovers totally in unloading. In alloys having this spring-like behaviour, the giant MFIS and the full size MSM effect is possible. This magnetically assisted pseudo-elastic behaviour of the martensitic Ni-Mn-Ga alloys exists in a broad temperature range. The 7M martensite shows a similar behaviour with the maximum magnetic stress of approximately 1.6 MPa at ambient temperature and approximately 10% pseudoelastic strain (Sozinov et al., 2002a, 2004a; Likhachev et al., 2004a). In the NM martensite the twinning stress exceeds the magnetic stress and, consequently, the pseudo-elastic strain has not been observed in it (Sozinov et al., 2004b).

4.1. *Effect of load on magnetic-field-induced strain (MFIS)*

The effect of the constant transversal stress on the magnetic-field-induced strain of the 5M martensite has been studied by Murray et al. (2000b, 2001); Heczko et al. (2000, 2001b); Jääskeläinen (2001); Jääskeläinen et al. (2003); Likhachev et al. (2001, 2002), O'Handley and Allen (2001); O'Handley et al. (2003); Henry et al. (2001, 2002a, 2002b) and Tellinen et al. (2002), Straka and Heczko (2005). If the martensitic single-variant sample is put into

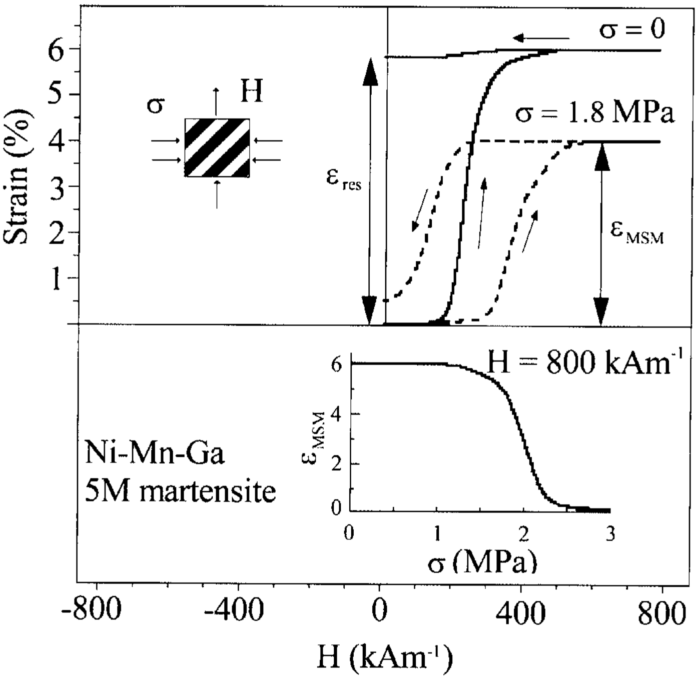


Fig. 4.2. The MFIS of a single-variant sample under a constant transversal stress. Insert shows the influence of the stress on ε_{MSM} with $H = 800\ \mathrm{kA\,m^{-1}}$.

the magnetic field freely ($\sigma = 0$ MPa), the obtained MFIS (ε_{MSM}) remains as a residual stress (ε_{res}) when the magnetic field is removed (fig. 4.2). The difference between ε_{MSM} and ε_{res} is very small. If the compressive stress is applied together with the magnetic field, the obtained MFIS (ε_{MSM}) decreases and the saturation field value for the ε_{MSM} increases. Also, the residual strain ε_{res} decreases, since a stress induced variant rearrangement occurs when the field is removed. If the applied stress is tensile, the magnetic field required for the twin boundary motion is less than in the case of compressive loading (Gans et al., 2004). The giant MFIS is totally suppressed when the transversal stress exceeds 2.5–3 MPa. This is explained by the competition between the applied stress (σ) and the magnetically induced stress ($\Delta\sigma_{mag}$) as the latter one can not exceed the value $K_u \cdot \varepsilon_0^{-1}$ (see Chapter 2). However, at compressive stresses higher than 3 MPa the conventional magnetostriction is still observed (Tickle and James, 1999; Heczko, 2005).

In actuation the reversible strain (ε_{rev}) is important (Likhachev et al., 2001) and ε_{rev} has a maximum at a certain optimal value of the applied stress (fig. 4.3). With the correct stress, the MSM-element is brought back to its initial state when the magnetic field is removed. If the stress is too small, it cannot move the twin boundaries totally back and the shape change is only partial. With too high stress the MFIS and the ε_{rev} are suppressed.

4.2. Dynamical actuation and fatigue of MFIS

For the dynamical actuation of Ni-Mn-Ga MSM elements, usually an axial movement is applied, even though bending would also be possible (Suorsa et al., 2002; Tellinen et al., 2002; Suorsa and Pagounis, 2004). The mechanical components of an ordinary actuator are the MSM element, the moving mass and a spring for spring-back load. The MSM

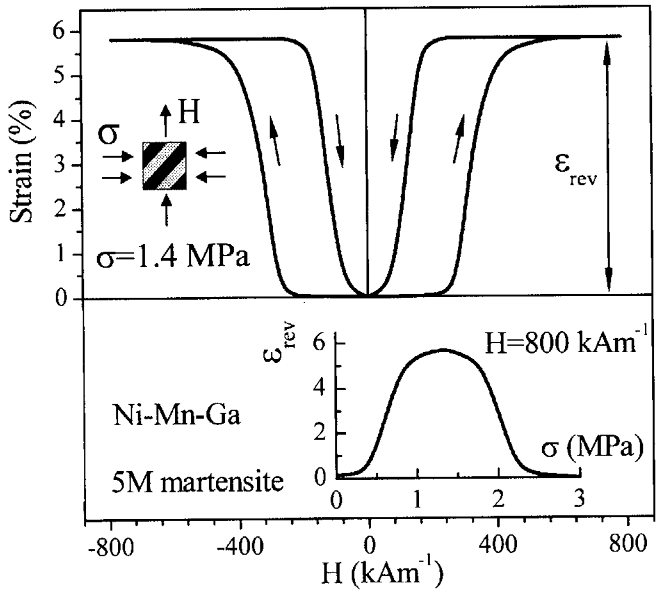


Fig. 4.3. The actuation of a MSM element with the constant transversal stress in an alternating magnetic field. Insert shows the influence of the stress on $\varepsilon_{\mathrm{rev}}$ in $H = 800\ \mathrm{kA\,m^{-1}}$. (Courtesy of Söderberg et al., 2005, reproduced by permission of Elsevier Science).

element is in the same magnetic circuit with the ferromagnetic core and the magnetic field is generated by magnetising coils. The system is usually driven by a current, since with a voltage drive there will be a delay in operation. During one full circle of the magnetic field change the shape of the MSM element changes twice. The maximum change increases with the growing magnetic field until the saturation value is reached (Aaltio and Ullakko, 2000). The magnetic-field-induced force of the actuator depends on the cross-sectional area of the MSM element, while the stroke is related with the length of the element. At high frequencies the eddy currents have to be reduced and the system may also reach the resonance frequency.

The Ni-Mn-Ga single crystals are used for actuation of 2%–4% strains with frequencies decreasing with the generated stroke (Henry et al., 2001, 2003a; Suorsa et al., 2002; Tellinen et al., 2002). The Ni-Mn-Ga element contracts along the field direction and extends in the plane normal to the magnetic field while conserving the volume during actuation. Figure 4.4 indicates that the Ni-Mn-Ga samples can be driven at least up to 320 Hz (Henry et al., 2003b). Beyond 100 Hz the peak output strain seems to drop down sharply; this is a response to the reduction in the applied field.

The fatigue of the MSM actuation is a less studied field. A demo-actuator driven by a 1.8 A current showed unchanged behaviour after 40 000 cycles (Aaltio and Ullakko, 2000). A high-frequency actuator with 10 mm MSM element with about 2% strain and 1 MPa constant stress kept the stroke value approximately constant till 200 million cycles of the alternating magnetic field (Tellinen et al., 2002). The evolution of the structure and the MSM properties in the 5M specimens have been studied during testing in a rotational magnetic field (6 Hz drive) with a 0.2 MPa bias stress (Heczko, unpublished). During the first million cycles the initial 6% MSM strain decreased to approximately 3% and then stayed approximately constant until the end of the test up to 36 million cycles. Müllner et

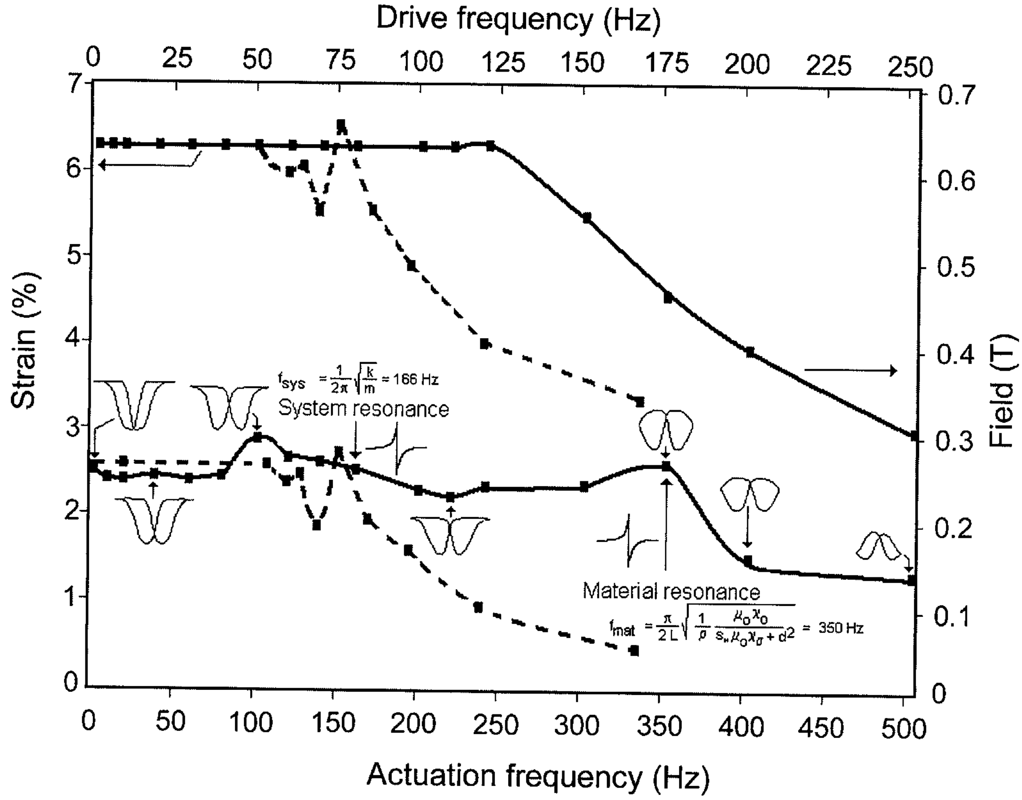


Fig. 4.4. Dependence of the maximum field applied to the MSM element and the strain measured at the maximum field as functions of the drive frequency and the actuation frequency for two different test systems (dot and solid lines). 1 Tesla corresponds with approximately 800 kA m^{-1}. (Courtesy and compilation from Henry et al., 2003b, reproduced by permission of SPIE—The International Society for Optical Engineering).

al. (2004) showed that the 5M material (in ref. 10M) showed a constant magnetic-field-induced strain in cycling, while in the 7M material (in ref. 14M) the MFIS decreased. Müllner et al. (2004) and Xiong et al. (2005) have studied the fracture mechanism of Ni-Mn-Ga martensites in thermal and magnetic cycling.

4.3. Temperature dependence of MFIS

The MSM effect is limited by the phase transformations and affected by the temperature dependence of the twinning stress, the tetragonality of lattice and the magnetic anisotropy (Heczko and Straka, 2003). In the 5M Ni-Mn-Ga martensite the lattice parameter a increases slightly, while the parameter c decreases with decreasing temperature (Ma et al., 2000; Glavatska et al., 2002, 2003a) resulting in increasing of the lattice distortion $(1-c/a)$ and, consequently, in an increase of the largest possible MFIS. However, the temperature dependencies of the magnetic anisotropy of the material and the twinning stress are more important. They both increase during cooling, but the twinning stress increases considerably (Heczko et al., 2002a, 2002b, 2003). Consequently, the lowest MSM service temperature is reached when the magnetically induced stress is no longer able to exceed the twinning stress. Another lowest limit is the possible intermartensitic transformation. The highest service temperature is limited by the reverse transformation. The service temperature region of the MSM effect for the alloy $Ni_{49.7}Mn_{29.1}Ga_{21.2}$ is shown in fig. 4.5

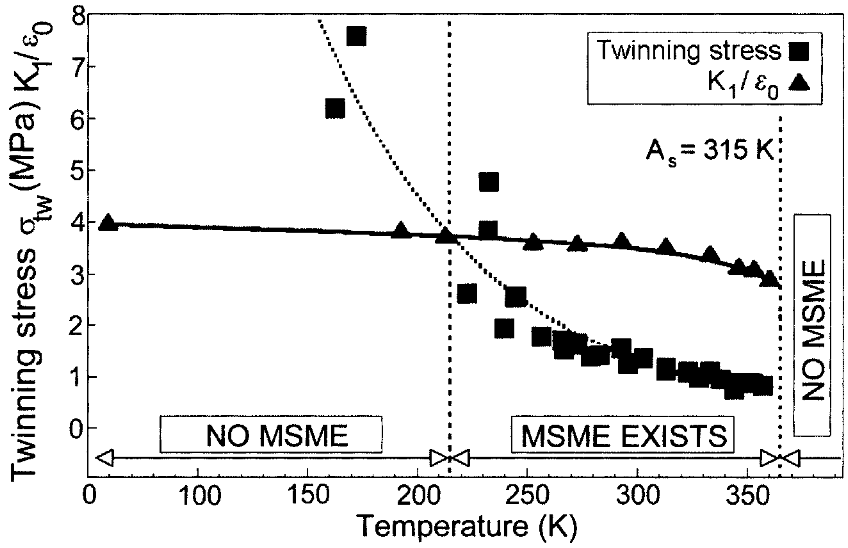


Fig. 4.5. The existence of the MSME in the alloy $Ni_{49.7}Mn_{29.1}Ga_{21.2}$ is limited by the reverse phase transformation at a high temperature and by the temperature where magnetically induced stress ($\sigma_{mag} = K_1/(1 - c/a)$, see part 2) becomes lower than the twinning stress σ_{TW}. (Courtesy of Heczko and Straka, 2003, reproduced by permission of American Institute of Physics).

(Heczko and Straka, 2003). Here, it was found that the obtainable MFIS increases with the decreasing temperature, but it also has been reported that the MFIS decreases in cooling of the alloy $Ni_{52.6}Mn_{23.5}Ga_{23.9}$ (Pasquale et al., 2002).

5. Selected properties of Ni-Mn-Ga alloys

The unique transport properties (Zhou et al., 2002b; Lee et al., 2003) or the magnetocaloric effect (Pakhomov et al., 2001; Marcos et al., 2003, 2004a, 2004b; Aliev et al., 2004; Albertini et al., 2004; Zhou et al., 2004, 2005) may create possibilities for using some Ni-Mn-Ga alloys in such applications as spin electronics and solid state refrigeneration. When Ni-Mn-Ga alloys were grown by molecular beam epitaxy, the resulting heterostructures showed in-plane magnetization and Curie temperatures of approximately 300 K (Dong et al., 2000a; Palmstrom et al., 2002). In a magnetocaloric effect the applied magnetic field may cause enhanced heat production or absorption in a material showing the simultaneous occurrence of structural and magnetic transitions. A large magneto-entropy change has been observed in some Ni-Mn-Ga alloys (Hu et al., 2000, 2001a, 2001b; Pareti et al., 2003) as well as the simultaneous occurrence of the structural and magnetic transition approximately at 370 K (Chernenko et al., 2002; Khovailo et al., 2002; Jiang et al., 2002; Söderberg et al., 2004a).

In corrosion testing in NaCl-water-solution the non-modulated NM structure showed the best chemical properties among the Ni-Mn-Ga martensites and the cubic parent phase was at the same level (Liu et al., 2002a, 2002b, 2003a, 2003b). Ni-Mn-Ga behaved better than the common low-alloyed steel, but not so well as the AISI316L stainless steel.

6. Promising MSM materials

In addition to the bulk materials the research of the Ni-Mn-Ga MSM includes also thin films (Suzuki et al., 1999; Dong et al., 2000a, 2000b, 2004; Ahn et al., 2001; Tello et al., 2002; Wu et al., 2002a; Wu and Tseng, 2002b; Castaño et al., 2003; Kohl et al., 2003; Rumpf et al., 2003; Chung et al., 2003; Hakola et al., 2004a, 2004b; Dubowik et al., 2004; Golub et al., 2004; Kim et al., 2004), ribbons (Chernenko and Vitenko, 1994; Chernenko et al., 1994; Kanada et al., 1998; Albertini et al., 2002b; Heczko et al., 2002c; Algarabel et al., 2004) and composite structures (Feuchtwanger et al., 2003a, 2003b; Hosoda et al., 2004). Also, the investigation of MSM powders has intensified, since they are important especially while preparing composites (Berkowitz et al., 2004; Solomon et al., 2004). However, Ni-Mn-Ga alloys are rather expensive, brittle and currently their MSME service temperature range is below 340 K. The search for possible new MSM materials is targeted to the alloys with a thermally formed or stress induced ferromagnetic thermoelastic martensite phase. The martensite should have extremely mobile twin boundaries. The high magnetocrystalline anisotropy and the high saturation magnetization are also desirable properties. The Fe-Pd and Fe-Pt alloys have shown MFIS, while the Fe-Ni-Co-Ti group has not been a success. Co-Ni and Co-Ni-Ga have turned out to be promising candidates, while Co-Ni-Al and Ni-Mn-Al have not been suitable.

Alloys close to Fe_3Pd show a martensite transformation up to 273 K and also a SIM formation resulting in a tetragonal phase with a short c-axis (Sohmura et al., 1980; Matsui et al., 1981; Sugiyama et al., 1984; Kato et al., 2002). The Curie point of the $Fe_{70}Pd_{30}$ is 573 K (Matsui and Adachi, 1983; Koeda et al., 2001; Cui and James, 2001). The saturation magnetization of the martensitic phase is 1400 kA/m and the magnetocrystalline anisotropy constant K_u is approximately 1 MJ/m^3 (Matsui and Adachi, 1983, 1989; Klemmer et al., 1995; James and Wuttig, 1998). The single crystalline Fe-Pd has shown a free MFIS of 0.5% at 256 K in a 1 T cyclic field as well as a single strain of 3% and subsequent cycles with 0.1% MFIS at 77 K in a 4 T field (James and Wuttig, 1998; Koeda et al., 2001; Yamamoto et al., 2004). At ambient temperature in a 1 T field, ribbons prepared by melt-spinning showed 0.18% free strain and 0.08% strain with a 10 MPa tensile stress (Furuya et al., 1998; Kubota et al., 2001, 2002a). The good MFIS of ribbons is based on the fine columnar grains in which there was developed a $\langle 100 \rangle$ fiber texture and a $\langle 100 \rangle$ tilt grain boundary (Kubota et al., 2002b; Yasuda et al., 2002). In $Fe_{70.4}Pt_{29.6}$ melt-spun ribbons the magnetostriction at 300 K has been 0.065% (Kubota et al., 2002b), while a larger shape change of 1.5–2.3% has been observed in the single crystalline Fe_3Pt in a magnetic field of 4 T at 4.2 K (Kakeshita et al., 2000; Kakeshita and Fukuda, 2002; Sakamoto et al., 2004). The Fe-Pt alloys have a martensitic transformation to a tetragonal phase up to 450 K, but the thermoelastic structure is obtained only in annealed materials with a drop of the T_m and an increase of T_C (Wayman, 1971; Dunne and Wayman, 1973a, 1973b; Kajiwara and Owen, 1974; Tadaki, 1977; Kakeshita et al., 1984). Close to the composition Fe_3Pt both martensite and austenite are ferromagnetic and the Curie temperature is approximately 450 K (Wasserman, 1990). Fukuda et al. (2004) showed that the $Fe_{68.8}Pd_{31.2}$ samples expand at 77 K in a 1.25 T field and at 4.2 K in a 4 T field the Fe_3Pt samples contract along the field direction [001], since the easy axis of magnetization in the Fe-Pd alloy is the a-axis and in the Fe-Pt alloy the c-axis.

The Co-Ni alloys show both the thermal and the SIM transformation when the Ni content is less than 35 wt.%. In the single-crystalline Co–33 wt.% Ni approximately 3% reversible MFIS due to SIM formation was obtained in a 2 T field below 180 K, and at ambient temperature Co–32 wt.% Ni showed a MFIS of 4.2% during the first 4–5 cycles in a 1.2 T field along the [001] orientation (Jiang et al., 2001; Liu et al., 2001, 2003a; Zhou et al., 2003). Also, Co-Ni-Ga alloys close to the Heusler composition show a thermoelastic martensite transformation and a shape memory effect (Oikawa et al., 2001a; Wuttig et al., 2001; Craciunescu et al., 2002a, 2002c; Kishi et al., 2003). The higher Ni/Ga increases the transformation temperatures and decreases the Curie temperature—in the alloy $Co_{47}Ni_{23}Ga_{30}$ T_m and T_C merge at 370 K. The phase transformation temperatures increase also when Al is added instead of Ga or by water quenching of the alloys having less than 26 at.% Ga. In the [001] single crystalline sample of $Co_{50}Ni_{22}Ga_{28}$ (T_C 400 K, T_m 343 K) the two-way shape memory can be continuously adjusted from -2.3% to 0 with a bias field of 0.8 T and when increasing the bias field to 2.0 T a positive shape deformation of $+2.2\%$ occurs (Li et al., 2004b). In Co-Ni-Al shape memory alloys the martensite transformation temperature increases and the Curie point decreases as the Ni content changes from 30 to 45 at.%, when the Al content is 30 at.% and the martensitic transition merges to the magnetic one at approximately 250 K when the Ni content is 35 at.% (Kainuma et al., 1996b; Oikawa et al., 2001a, 2001b; Murakami et al., 2002). The magnetocrystalline anisotropy of the $Co_{37}Ni_{34}Al_{29}$ is 3.9 MJ/m^3 and the reversible MFIS is limited to 0.06% (Morito et al., 2002).

In the Ni-Mn-Al system the ordered $L2_1$ phase transforms to martensite the crystal structure of which depends on the alloy composition (Inoue et al., 1994; Kainuma et al., 1996a, 2000; Otsuka and Morito, 1996; Sutou et al., 1998). Ferromagnetic ordering with a Curie temperature of approximately 330 K can be obtained by aging the quenched samples (Gejima et al., 1999). A magnetostrain of 1% in a single crystalline sample has been obtained in a 7 T magnetic field (Fujita et al., 2000).

From other material groups Ni-Ga-Fe (Oikawa et al., 2002; Ota et al., 2002; Sutou et al., 2004), Co_2NbSn (Garde and Ray, 2002; Neumann et al., 2002; Wolter et al., 2002), Fe-Rh (Ibarra and Algarabel, 1994), Mn-As (Chernenko et al., 1999), $La(Fe_xSi_{1-x})_{13}$ (Fujieda et al., 2001), $Gd_5(Si_xGe_{1-x})_4$ (Morellon et al., 2000) and in a antiferromagnetic γ-Mn-Fe(Cu) (Zhang et al., 2005) have been studied to some extent.

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