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Abstract

Field-induced strains of 6% are reported in ferromagnetic Ni–Mn–Ga martensites at room temperature. The strains are the result of twin boundary motion driven largely by the Zeeman energy difference across the twin boundary. The strain measured parallel to the applied magnetic field is negative in the sample/field geometry used here. The strain saturates in fields of order 400 kA/m and is blocked by a compressive stress of order 2 MPa applied orthogonal to the magnetic field. The strain versus field curves exhibit appreciable hysteresis associated with the motion of the twin boundaries. A simple model accounts quantitatively for the dependence of strain on magnetic field and external stress using as input parameters only measured quantities.

Keywords

magnetostriction, magnetoelastic effects, twin boundaries, shape memory effects, stress-strain relations, nickel alloys, manganese alloys, gallium alloys, twinning, strains, magnetic fields, ferromagnetic materials, hysteresis

Disciplines

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Comments

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6% magnetic-field-induced strain by twin-boundary motion in ferromagnetic Ni–Mn–Ga

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Field-induced strains of 6% are reported in ferromagnetic Ni–Mn–Ga martensites at room temperature. The strains are the result of twin boundary motion driven largely by the Zeeman energy difference across the twin boundary. The strain measured parallel to the applied magnetic field is negative in the sample/field geometry used here. The strain saturates in fields of order 400 kA/m and is blocked by a compressive stress of order 2 MPa applied orthogonal to the magnetic field. The strain versus field curves exhibit appreciable hysteresis associated with the motion of the twin boundaries. A simple model accounts quantitatively for the dependence of strain on magnetic field and external stress using as input parameters only measured quantities. © *2000 American Institute of Physics.* [S0003-6951(00)00832-9]

Ferromagnetic shape memory alloys (FSMAs) moved from a hypothetical new class of active materials, to join piezoelectric and magnetostrictive materials, upon observation of a 0.2% magnetic-field-induced strain in a single crystal of Ni_2MnGa in 1996.[1] By way of comparison, piezoelectric materials[2] show strains of order 0.1%[2] and the leading magnetostrictive material, Terfenol-D,[3] shows a field-induced strain of about 0.24%. This 0.2% strain represents only a small fraction of the 6% or 7% transformation strain expected to be accessible by field-induced twin selection in Ni–Mn–Ga FSMAs. Extensional strains of 0.5% have been observed in martensitic $Fe_{70}Pd_{30}$ crystals[4] at -17 °C and 2.3% in Ni_2MnGa martensitic crystals[5] at -8 °C. Shear strains of 5.7% have been reported at room temperature in Ni–Mn–Ga martensites.[6]

Here we report measurements of field-induced generation of the full extensional strain associated with the crystallographic distortion in a ferromagnetic shape memory alloy. An initial single-variant state is achieved by application of a compressive stress of order 1 MPa to a sample of an off-stoichiometry crystal of Ni–Mn–Ga. The strain is produced by application of a field of typically 400 kA/m (5 kOe) orthogonal to the compressive stress direction. High-speed photographs reveal the nearly complete transformation of the sample from one twin variant to another by twin boundary motion during application of the field. The form of the strain as a function of applied field and stress is quantitatively well described by a thermodynamic model[7,8] that includes Zeeman energy, magnetocrystalline anisotropy, external stress, and an internal restoring force. The observed hysteresis is due to the threshold stress needed to initiate twin boundary motion and can be determined independently from a stress–strain curve. There are no adjustable parameters in the model.

The composition weighed out for growth, $Ni_{47.4}Mn_{32.1}Ga_{20.5}$, was chosen slightly off the Heusler stoichiometry, Ni_2MnGa. This was done in order to render alloys having Curie temperatures greater than the martensite transformation temperature, which in turn was to be greater than room temperature.[9] Crystals were grown by the Bridgman technique at Ames Laboratory. High purity nickel, manganese, and gallium were arc melted into buttons and then drop cast into a chilled copper mold. The as-cast ingots were placed in alumina crucibles and were heated to 1350 °C for 1 h to allow homogenization before withdrawing the sample from the heat zone at a rate of 5.0 mm/h. In order to minimize evaporation of the Mn during crystal growth, the furnace was backfilled to a positive pressure of 6.8×10^5 Pa with purified argon after the chamber and sample had been outgassed at 1350 °C under vacuum. The single crystal boule measured 1 cm in diameter and 5 cm in length. Electron probe microanalysis showed the composition to vary along the boule axis with Mn content increasing and Ga content decreasing in the growth direction.

After Laue orientation, several samples were cut from the crystal boule by electric discharge milling. Rectangular prismatic samples measuring about 6 mm by 6 mm by 20 mm, were cut to have their faces parallel to the faces of the high-temperature, $L2_1$ (Fm3m) unit cell.[8] The sample described here has an analyzed composition of $Ni_{49.8}Mn_{28.5}Ga_{21.7}$, with a Curie temperature of 95 °C. Upon transformation to the tetragonally distorted ($I4/mmm$) martensitic phase below 45 °C, twinning can occur along {112} planes ({101} or {011} in $Fm3m$). A separate disk-shaped sample measuring 3 mm diameter by 1 mm thick was made of essentially the same composition as the prism. This disk sample has a ⟨100⟩ direction in its plane and was prepared for magnetic anisotropy measurements. The single-variant state was established in the disk by application of a magnetic field while the sample was cured in epoxy. (A twin-free state can be achieved either by application of a stress of order 1 MPa or a magnetic field of order 400 kA/m.) After curing of

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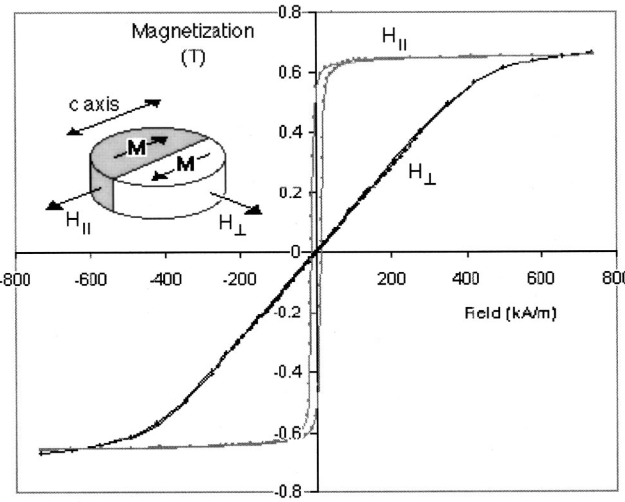

FIG. 1. Field dependence of magnetization at room temperature in single-variant, martensitic Ni–Mn–Ga crystal with field applied parallel and perpendicular to [001]—Ref. 11.

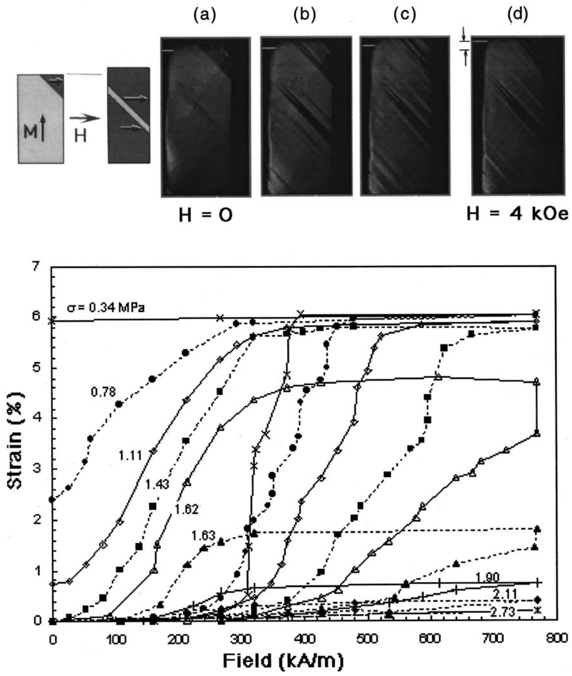

FIG. 2. Top: Orientation of **M**, **H**, and σ relative to the twinned sample. Selected high-speed video frames show the sample in the initially compressed ($\sigma \approx 0.2$ MPa) state ($H \approx 0$), in two intermediate states, and in the magnetically saturated, fully strained state, respectively. Bottom: Field-induced strain at various external opposing stresses at room temperature for a $Ni_{49.8}Mn_{28.5}Ga_{21.7}$ single crystal.

the epoxy, the single-variant state was retained. Room temperature magnetization curves taken parallel and perpendicular to the *c* axis and in the plane of the disk are shown in Fig. 1. They indicate the strength of the uniaxial magnetic anisotropy to be $K_{u1} = 1.5 \times 10^5$ J/m^3. The saturation magnetization at room temperature is $\mu_0 M_s = 0.62$ T. Tickle and James[10] measured the anisotropy $Ni_{51.3}Mn_{24.0}Ga_{24.7}$ single crystals constrained to be in the single-variant state. They reported $K_u = 2.45 \times 10^5$ J/m^3 at $-17\,°C$.

For the field-induced strain measurements, the prismatic sample was placed with its short dimension along the field axis of an electromagnet while its long dimension (vertical) supported a nonmagnetic push rod extending above the electromagnet and ending in a platform. Dead weights were placed on the platform to apply a stress to the sample. The displacement of the platform referenced to the sample base was measured with an eddy current proximity sensor. A stress of 1.0 MPa was found to render the sample in a nearly single-variant state.

Figure 2 shows the results of field-induced strain measurements in a $Ni_{49.8}Mn_{28.5}Ga_{21.7}$ single crystal at room temperature for various axial external stresses that oppose the field-induced strain. The sketch shows the orientation of the magnetization, magnetic field, and external stress relative to the twinned sample. The magnetization vectors are shown parallel to the tetragonal *c* axis in each variant. The photographs are frames from a high-speed (1000 frames/s) video taken on the sample in the initially stressed state (approximately 0.34 MPa) at $H = 2 \times 10^5$ A/m (a), at two intermediate states of actuation (b), (c), and at saturation after about 23 ms (d). The images show that upon application of a transverse field, twins magnetized parallel to the field (darker contrast) appear and grow, causing the sample to extend vertically against the applied stress. The graph shows that the strain increases sharply beyond about 200 kA/m and saturates at about 400 kA/m. Upon removal of the field, the sample remains in an essentially transversely magnetized (vertically extended) state for $\sigma < 0.5$ MPa. Repeating the field cycle under incrementally greater stress results in an increase in the threshold field at which most strain occurs. At stress levels in excess of 1.1 MPa, the initial stress-stabilized state is reestablished upon removal of the field. At still higher stresses, the $e-H$ curves are sheared over and the full transformation strain is no longer achieved. A stress of 2 MPa (44 lb/cm^2) reduces the maximum field-induced strain to 0.6%. The hysteresis appears nonmonotonic in applied stress. Essentially the full transformation strain is achieved in this case for stresses less than 1.0 MPa. Other samples from the same boule have shown strains of 5.7% at saturation.

Phenomenological models describing the field-induced motion of twin boundaries in FSMAs generally include the Zeeman energy, magnetic anisotropy energy, an internal restorative elastic energy, and an external stress.[7,8,12] The field-dependent strain may be expressed as a function of the volume fraction, f_i, of each variant: $e(H) = e_0 \delta f(H)$, where e_0 is the transformation strain and $\delta f = f_1 - 1/2$. The equivariant state, $f_1 = f_2 = 1/2$, is defined as $e_0 = 0$.[7] Micromagnetic models[5,10,13] may also include a magnetostatic energy that tends to restore *M* to zero when the field vanishes. The action of the external stress depends on its orientation relative to the field-induced deformation. The stress here is oriented to oppose the field-induced strain. The results of the model are summarized by

$$e(H) = e_0 \delta f = \frac{2K_u h(1-h/2) - \sigma e_0}{C_{\mathrm{eff}} e_0}, \qquad (1)$$

where $h = M_s H / 2K_u$ and C_{eff} is the effective modulus of the twinned state that accounts for the elastic energy stored in parts of the material that do not respond to the field by twin-boundary motion.[7,12] The data of Fig. 2 are described with this model using the measured Ni–Mn–Ga parameters: $\mu_0 M_s = 0.6$ T, $K_{u1} = 1.8 \times 10^5$ J/m^3, $\epsilon_0 = 0.06$, and $C_{\mathrm{eff}} = 2$

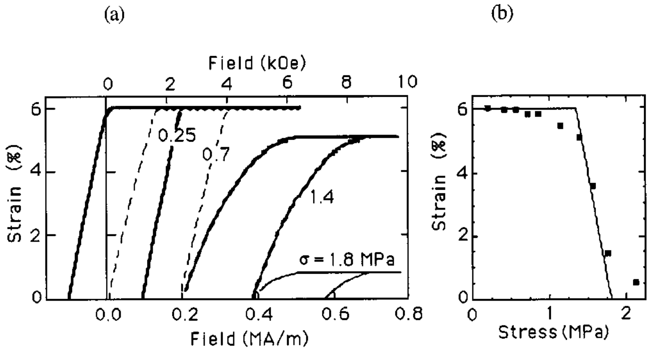

FIG. 3. Left, calculated strain vs applied field curves from Eq. (1) and right, calculated strain vs stress with overlaid experimental data.

$\times 10^7\ \mathrm{N/m^2}$. The model results displayed in Figs. 3(a) and 3(b) show that these parameters give a reasonable reproduction of the major trends in the experimental data, namely the shape of $e(H)$, as well as the increase in threshold field and decrease in field-induced strain with increasing external stress. The model does not account for the observed change in coercivity with external stress. The predicted decrease in saturation strain with increasing stress [Fig. 3(b)] also gives a reasonable fit to the data.

The coercivity is introduced into the model in an *ad hoc* manner (external field is expressed as $H \pm H_c$ for decreasing or increasing field strength, respectively) but its value is derived from the mechanical stress strain curves of the crystal (Fig. 4). The mechanical properties of FSMAs in the martensitic phase are similar to those of conventional SMAs. With increasing stress, the material first shows a modulus, C_0, characteristic of the single-variant state. Above a critical stress, σ_0, at which deformation by twin boundary motion initiates, the modulus decreases to C_{tb} and the material may strain to its full transformation strain, e_0, after which it is mechanically detwinned. For larger stresses, the modulus reverts to its single-variant value. Values of the parameters in Fig. 4 measured for Ni_2MnGa crystals are typically of order[11,14] $\sigma_0 \approx 1$ MPa, $C_0 \approx 2$ GPa, $e_0 = 0.06$, and $C_{\mathrm{tb}} \approx 18$ MPa. The coercivity is given by $H_c = \sigma e_0/\mu_0 M_s = 100$ kA/m.

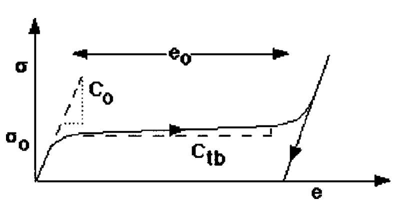

FIG. 4. Stress–strain behavior of martensitic Ni-Mn-Ga alloys.

In summary, strains of 6% have been produced at room temperature in single crystals of Ni–Mn–Ga by application of fields of order 400 kA/m under an opposing stress of order 1 MPa. The strain is the result of field-induced twin boundary motion. The form of the dependence of the strain on field and stress is well described by a phenomenological model with no adjustable parameters; all parameters are independently measured.

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