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Breaching the work output limitation of ferromagnetic shape memory alloys

Yaniv Ganor,[1,a)] Doron Shilo,[1] Thomas W. Shield,[2] and Richard D. James[2]
[1]*Department of Mechanical Engineering, Israel Institute of Technology—Technion, Haifa 32000, Israel*
[2]*Department of Aerospace Engineering and Mechanics, University of Minnesota, Minneapolis, Minnesota 55455, USA*

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One important parameter that quantifies the performance of ferromagnetic shape memory alloys is the blocking stress. To date, the low blocking levels (<5 MPa) impede the utilization of these alloys in applications where high work output is required. In this paper, we demonstrate an increase in the blocking stress by more than *100%* by reducing the actuator size. A new theoretical model shows that smaller specimens have increased values of the blocking stress due to an enhancement in the energy barrier to magnetization rotation and indicates on a fundamental relationship among the specimen size, its microstructure, and its physical properties. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2982916]

In recent years, a special class of active materials known as ferromagnetic shape-memory (FSM) alloys has received much attention due to its large actuation strains and fast response.[1–5] Despite the large strains, the work output of FSM alloys is limited by the FSM alloys' relatively low blocking stress of 2–6 MPa,[5–7] above which magnetic field induced strain vanishes. This level of stress impedes the utilization of these alloys in practical applications where high work output by small components is required. To date, attempts to enhance the work output limitation have been mainly focused on changing the concentrations of the off stoichiometric compounds of the FSM alloys[8] to increase the magnetocrystalline anisotropy energy. In this paper we suggest that the work output limitation can be breached by reducing the size of the actuator and present evidence for a fundamental relationship between size and work output.

The actuation mechanism of FSMs differs from that of conventional, temperature driven shape memory actuators. In temperature driven shape memory actuation, the reversible strain is related to a structural phase transformation from austenite to martensite.[9,10] FSM actuation is due to a fast rearrangement of twin variants of the martensite induced by a magnetic field.[11] In a typical specimen of five-layer modulated Ni_2MnGa, a cubic to tetragonal martensitic phase transformation occurs when the material is cooled below a characteristic temperature. Different parts of the crystal transform to different variants of martensite. Each pair of variants is compatible, and they meet at one of two well-defined interfaces called twin boundaries. These boundaries are also coincidentally pole-free when the magnetization is parallel to the easy axis of each variant. An applied field and stress can competitively bias the material toward one or the other of these variants.[1] When an external magnetic field H is applied, it aligns the magnetization M toward this direction. The resulting alignment of magnetization can take place either by switching the variant or by a rotation of the magnetization. The variant switching is the basic actuation mechanism in FSM alloys and it results in a large strain of up to 6.5%[2,3] which is more than an order of magnitude larger than giant magnetostrictive materials such as Terfenol-D and piezoelectric materials such as PZT.[12,13] On the contrary, the magnetization rotation results in strains only of the order of 10^{-5}.

The amount of energy required to rotate the magnetization away from the easy axis is quantified by the magnetocrystalline anisotropy constant. In Ni_2MnGa this constant is high enough such that at low external loads, the variant switching process is favorable. However, when a high enough stress is applied, variant switching is inhibited and magnetization rotation becomes energetically favored. At this point, the blocking stress is reached, and only small ordinary magnetostrictive strain is produced upon application of the field. This determines the limit on the amount of work output that can be generated by the FSM actuator.

Variant switching is a complicated physical process in which the volume fraction of the energetically favored variant is increased at the cost of the others. This process takes place by nucleation of new domains/variants and twin boundary motion.[1] Hence, it is significantly influenced by the martensitic microstructure. In ferromagnetism there is a fundamental relation between the specimen size and the average domain size, which originates from the competition between exchange, demagnetization, and anisotropy energies.[14] This gives rise to the hypothesis that specimens of different scales will affect the martensitic/domain microstructure and might exhibit different mechanical behavior.

To establish that the microstructure is influenced by the size of the specimen, we performed a preliminary calculation of a multivariant periodic microstructure of alternating vertical twin bands, as depicted in Fig. 1(a). When magnetization rotation occurs in one variant, the magnetization divergence-free condition across neighboring twin boundaries is no longer fulfilled. This results in strain and magnetic incompatibilities at the vicinity of twin boundaries that cost a significant increase in the energy per unit area of the twin boundary.[1] Accordingly, we suggest the existence of horizontal fine arrays of magnetic domains within twin bands, inspired by the observations of Lai *et al.*[15] and illustrated in Fig. 1(b), such that the average magnetization jump at the

a)Author to whom correspondence should be addressed. Electronic mail: yanivg@tx.technion.ac.il.

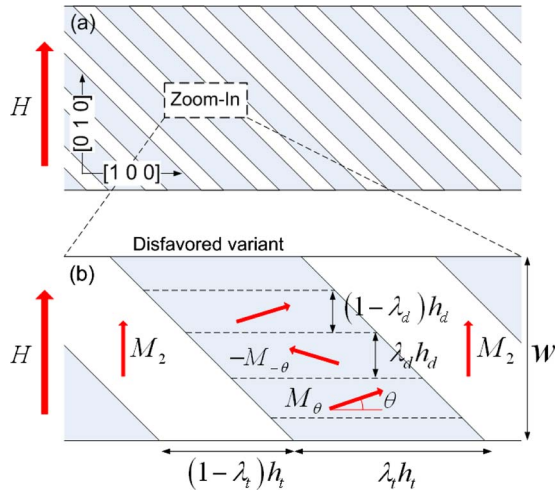

FIG. 1. (Color online) Configuration of domains assumed in the domain theory calculation.

twin boundaries will be divergence-free. A minimization of the total energy of the system is sought to determine the volume fractions $0<\lambda_t<1$ and $0<\lambda_d<1$, length scales h_t and h_d, and the angle of rotation θ [Fig. 2(b)]. The rotation of magnetization at high stress occurs within the fine arrays of magnetic domains in the twin bands containing the disfavored variant.[16] The total energy was computed from the sum of anisotropy, demagnetization, domain/twin boundary, and Zeeman energies.

We have seen from the domain theory calculation summarized below that the fineness of these domains and the fineness of the twins are significantly affected by the specimen dimensions, with smaller specimens favoring finer twins (domains within twins) with thicknesses proportional to $\sqrt{w}$ ($\sqrt[4]{w}$), where w is the specimen width. The same calculation predicts an increase in the height of the energy barrier accompanying magnetization rotation with decreasing w.

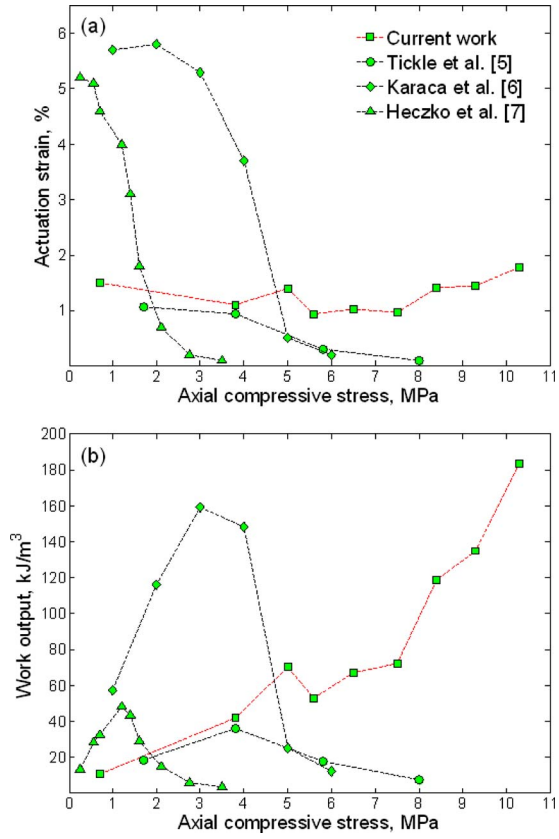

FIG. 2. (Color online) (a) Actuation strain and (b) work output for varying axial compressive loads.

The preliminary calculations, based on the model shown in Fig. 1, indicated that the twin bands with easy axes aligned with the applied field would not subdivide into domains at moderate fields. Thus we only allowed subdivision of the bands having horizontal easy axes [Fig. 1(b)]. Closure domains were first introduced at the ends of the horizontal domains, but then it was found that because of the relation $M_s^2 \ll K_u$ in this alloy, that main contribution to the energy at these ends is the demagnetization energy due to the fluctuations of the magnetization, which was estimated. A relation $\lambda_d=f(\theta)$ emerged from energy minimization: this relation is equivalent to $(M_2-\langle M\rangle)\cdot n=0$, i.e., the *average magnetization* in the subdivided domains produces no poles on the twin boundaries. The values of h_t and h_d that minimize the total energy are

$$h_t=\sqrt{\frac{2\sqrt{2}\gamma_t w}{M_s^2 b_t}}, \quad h_d=\sqrt{\frac{2\gamma_d \lambda_t h_t}{M_s^2 b_d}}, \tag{1}$$

where $M_S=600$ emu/cm^3 is the saturation magnetization and $\gamma_t\approx 3$ J/m^2 and $\gamma_d\approx 2$ J/m^2 are interfacial energy densities of the twin/domain boundaries.[17] The symbols b_t and b_d denote certain explicit functions of θ and λ_t that emerge from the estimation of the demagnetization energy of the fluctuations. These functions obtain values between 0 and 1. Substituting the above values in Eq. (1) for the approximation $b_t=b_d=0.5$ shows that for a specimen width of 100 μm the microstructure periods are $h_t=5$ μm and $h_d=500$ nm. This means that the width of the magnetic domains becomes comparable or even smaller than the thickness of two domain walls, which is between 100 and 200 nm in most ferromagnetic materials.[16] For example, at small values of $\theta\leq 0.2$ the fraction $(1-\lambda_d)$ is smaller than 0.1 and the width of one type of the magnetic domain $(1-\lambda_d)h_d$ is smaller than 50 nm. Evidently, this is not a possible solution, and it indicates that the magnetization rotation cannot occur gradually. Therefore, a nucleation of an array of magnetic domains with relatively large values of θ is a necessary step in the magnetization rotation process. The energy barrier for this nucleation increases as the specimen size decreases and disfavors magnetization rotation as the specimen size is decreased.

These results are qualitatively consistent with the recent observations of Lai *et al.*[15] where subdivision of only the disfavored twins is observed. Also, in the range where significant rotation has occurred, our relation $\lambda_d=f(\theta)$ predicts $\lambda_d\sim\frac{1}{2}$ [the full relation is $f(\theta)=(1+\cos\theta-\sin\theta)/2\cos\theta$] which is also qualitatively similar to the observations of Lai *et al.* This consistency indirectly lends support to the predictions about the energetic barrier given above.

In light of these predictions a novel experimental setup was developed[11] to test whether specimen size affects the blocking stress and to provide initial measurements on the behavior of microscale actuators. The experimental system comprises an alternating magnetic field generator and a mechanical loading and sensing system. The strain is determined by measuring the angle of a reflected laser beam off the surface of a silicon cantilever held against the free sur-

face of the Ni_2MnGa sample. The experiments were performed using a single crystal Ni_2MnGa specimen cut to a square cross section with $h=200$ μm and a length of 4 mm with faces parallel to the [100] directions, and cooled down to −28 °C (about 30 °C below its martensitic final temperature).

A series of tests was performed with increasing values of mechanical load to determine the blocking stress of the microspecimen. Our results (dashed red line), together with previous results (black dashed lines) by Tickle and James,[5] Karaca *et al.*,[6] and Heczko *et al.*,[7] are shown in Fig. 2 in terms of the actuation strain [Fig. 2(a)] and work output [Fig. 2(b)]. The earlier experiments were done using Ni_2MnGa single crystal specimens cut to cross section dimensions of at least $h=2$ mm with different stoichiometric concentrations. The differences in actuation strain at low stress appearing in Fig. 2(a) are commonly observed in Ni_2MnGa and are likely related to differences in specimen quality in terms of surface and bulk defects. However, Fig. 2(b) shows a significant difference between earlier results and the current results. While earlier experiments show a continuous decrease in the work output, which begins at stresses of 1.5–4 MPa, our results show a continuous increase in the work output up to a load of 10 MPa, which was the technical limit of the experimental setup.

To conclude, the results demonstrate an increase in the blocking stress by more than 100%, which theory suggests is related to the size reduction of the FSM specimen in our experiments. Theory indicates that this is due to an enhancement in the energy barrier to magnetization rotation in the disfavored twins at small specimen sizes. A more complete study is underway, both theoretically and experimentally, which also includes the effect of changing the aspect ratio of the specimens.

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