

Sensitivity of twin boundary movement to sample shape in Ni-Mn-Ga

Medha Veligatla¹, Christian Titsch², Welf-Guntram Drossel^{2,3}, Carlos J. Garcia-Cervera^{4,5},

Peter Müllner¹

¹ Micron School of Materials Science and Engineering, Boise State University, Boise, ID, 83725, USA

² Institute for Machine Tools and Production Processes, Technische Universität, 09126 Chemnitz, Germany

³ Fraunhofer Institute for Machine Tools and Forming Technology, 09126 Chemnitz, Germany

⁴ Department of Mathematics, University of California, Santa Barbara, CA, 93117, USA

⁵ Visiting Professor at BCAM – Basque Center for Applied Mathematics, Bizkaia, Spain

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Abstract:

When applying a magnetic field parallel or perpendicular to the long edge of a parallelepiped Ni-Mn-Ga stick, twin boundaries move instantaneously or gradually through the sample. We evaluate the sample shape dependence on twin boundary motion with a micromagnetics computational study of magnetic domain structures and their energies. Due to the sample shape, the demagnetization factor varies with the direction of external magnetic field. When the external magnetic field is applied perpendicular to the long edge of the sample, i.e. in the direction in which the demagnetizing field is highest, the magnetic energy intermittently increases when the strength

of the applied magnetic field is low. This energy gain hinders the twin boundary motion and results in a gradual switching, i.e. a gradual magnetization reversal as the applied magnetic field is increased. The formation of 180° magnetic domains offsets this effect partially. In contrast, when the applied magnetic field is parallel to the long edge of the sample, i.e. in the direction in which the demagnetizing field is lowest, the energy decreases with each subsequent magnetization domain reversal and the twin boundary moves instantaneously with ongoing switching. The actuation mode with the field parallel to the long sample edge lends itself for on-off actuators whereas the actuation mode with the field perpendicular to the long sample edge lends itself to gradual positioning devices.

Introduction

Macroscopic deformation in magnetic shape memory (MSM) alloys occurs when the material is subjected to an external magnetic field or a mechanical stress. Lattice reorientation via twinning in the martensite phase causes this shape change. An MSM single crystal with one twin boundary consists of two twin domains sharing the boundary. These twin domains have different magnetization and crystallographic orientations[1,2]. Depending on the direction of the external magnetic field, one variant grows at the expense of the other as the twin boundary ~~to~~ moves across the sample. The maximum magnetic-field-induced strain depends on the martensite structure and lattice parameters and varies between 6 and 12 %[2–5] . With a few microseconds response time[6], these materials have great potential as actuators. Numerous research groups have studied the material properties of Ni-Mn-Ga single crystals and its response to variable magnetic fields to improve the performance of MSM actuators[7–14].

In 1995, Ullakko [15] introduced the concept of using magnetic field induced reorientation of martensite variants for magnetically powered actuators. Ullakko suggested that with several

percent strain and rapid control, the magnetic shape memory alloys may outperform piezoelectric and magnetostrictive materials. Subsequently, Ullakko *et al.* demonstrated the deformation in a Ni_2MnGa with magnetic fields[16]. In 2004, Suorsa *et al.*[17] measured various properties that determine the dynamic behavior of a 10M Ni_2MnGa MSM material. For a sample dimension of 1 mm x 2 mm x 10 mm, the authors reported the acceleration of the sample surface, rise time and actuation velocity to be 5000 m/s^2 , 0.2 ms and 1.3 m/s respectively. The switching behavior of the material dictates the response of the actuator. Recently, Saren *et al.*[6] and Smith *et al.*[18] reported twin boundary velocities of 39 and 82 m/s implying actuation speeds of 2.4 and 4.8 m/s. Pagounis *et al.* summarized some recent progress on MSM actuators[19].

The goal of this paper is to study extrinsic factors that influence twin boundary motion in MSM actuators. We apply experimental and numerical methods to study the macroscopic and mesoscopic magnetic response of elongated Ni-Mn-Ga single crystals in a magnetic field. We are particularly interested in the response of the material when exposed to magnetic fields in different directions, namely parallel and perpendicular to the long sample axis. While we performed switching field tests to demonstrate the macroscopic magnetic response, the micromagnetics simulations were carried out to demonstrate the mesoscopic magnetic interactions.

Micromagnetics

The field of micromagnetics was pioneered by Brown[20] and a comprehensive review was presented by Chantrell *et al.* [21]. Many research groups have used micromagnetics to characterize mesoscale magnetic properties of Ni-Mn-Ga alloys[22–29]. The theory of solving the Landau-Lifshitz dynamic equation was applied with various methods such as phase field modeling [23–25,28–30]. This method has been used to study the twin boundary mobility[23], magnetic domain evolutions[30], demagnetization effects[29], and magneto-mechanical properties[24,25] of Ni-

Mn-Ga. These research groups studied the magnetic domain evolution as the twin boundary moves along the sample length. In the present study, we simulate the domain evolution with respect to time with a fixed twin microstructure, i.e. with static twin boundaries. Neglecting twin boundary mobility allows us to study the interactions of magnetic domains and twin boundaries in greater detail. Hobza *et al.* applied a code developed by Garcia-Cervera[31] to study the torque generated by a magnetic field on Ni-Mn-Ga samples with various twin microstructures[22,32]. This code evaluates the actual dynamics (Landau-Lifshitz equation). In our method, we only solve linear systems of equations with constant coefficients. The cost per step of our method is $O(N \log N)$, where N is the number of cells. Using this code we obtained magnetic energies for magnetic equilibrium structures that summarize the switching behavior for a single twin boundary system in Ni-Mn-Ga. We map the different energy contributions in the process of magnetic domain evolution. In order to qualitatively compare these energies, the simulations were arranged such that they replicate the experimental setup of a switching field test at small scale. The equilibrium magnetic structures and energies obtained through these simulations take into account the anisotropy, exchange, Zeeman, and stray field energies. The code solves the Landau-Lifshitz-Gilbert equation to approach the minimum energy state:

$$d\mathbf{M}(\mathbf{r})/dt = -\left(\mu_0\gamma/M_s\right)\mathbf{M} \times \mathbf{H} - \alpha\left(\mu_0\gamma/M_s\right)\mathbf{M} \times [\mathbf{M} \times \mathbf{H}] \quad (1)$$

where $\mathbf{M}(\mathbf{r})$ is the magnetization density at position \mathbf{r} , γ is the gyromagnetic ratio, α is the dimensionless damping parameter, and $\mathbf{H}(\mathbf{r})$ is the magnetic field, which is the negative derivative of total energy with respect to magnetization:

$$\mathbf{H} = -\frac{\delta E}{\mu_0 \delta \mathbf{M}} = -\left(2K_u/\mu_0 M_s^2\right)(M_2 + M_3) + \left(2C_{ex}/\mu_0 M_s^2\right)\Delta \mathbf{M} - \nabla \mathbf{U} + \mathbf{H}_{\text{ext}} \quad (2)$$

where K_u is the anisotropy constant, M_s is the saturation magnetization, M_2 and M_3 are magnetization components that are orthogonal to the axis of easy magnetization, C_{ex} is the exchange constant, μ_0 is magnetic permeability of free space, and \mathbf{H}_{ext} is the external magnetic field.¹ The individual summation terms in equation (2) are the energies associated with magnetocrystalline anisotropy, exchange interaction, stray field, and external applied field (Zeeman energy). In short, the magnetocrystalline energy is the energy associated with the orientation of magnetic domains with respect to the axis of easy magnetization, the exchange energy is the short range interaction energy between neighboring magnetic moments, and the stray field and Zeeman energies are associated with magnetic domain splitting and the external magnetic field respectively. Hobza *et al.* provide a detailed description of these energy terms and the micromagnetics code [22].

Experiments and simulations:

All the experiments were conducted on a Ni-Mn-Ga single crystal with 10M martensite structure and composition $\text{Ni}_{49.5}\text{Mn}_{28.8}\text{Ga}_{21.7}$ (Goodfellow). A rectangular sample with dimensions 3.93 mm x 2.86 mm x 1.06 mm was cut with all faces parallel to $\{100\}$. X-ray diffraction and energy dispersive X-ray spectroscopy were done with a Bruker D8 Discover diffractometer and a Hitachi S-3400N-II scanning electron microscope equipped with an Oxford Instruments Energy EDS to confirm the crystal structure and the composition. Magnetic switching field experiments were conducted with a ADE model 10 vibrating sample magnetometer (VSM). For the VSM experiments, the sample was mounted to a quartz tube and exposed to an increasing magnetic field. The experiment were done with two configurations, namely such that the magnetic field was

¹ Equations 1 and 2 are given in SI unites and differ from those given in Ref. [31].

parallel to the longest (designated “parallel”) and the intermediate (designated “perpendicular”) edge of the sample. First the sample was placed in the field with parallel sample configuration. The field was increased from 0 to 1.2 T and reduced to 0 T. Then, the electromagnet was rotated such that the sample was in perpendicular configuration. In this setup the field was increased from 0 to 1.2 T and reduced to 0 T. Then the magnet orientation was rotated back to the parallel configuration. We conducted 6 experiments with alternating parallel and perpendicular configurations and measured the magnetization as a function of magnetic field strength. At the beginning of the experiments in parallel and perpendicular configurations, the samples were fully extended (6% strain) and fully compressed (0% strain), respectively.

We conducted micromagnetics simulations to assess the equilibrium magnetic structure and to calculate the magnetic energies of Ni-Mn-Ga samples for magnetic fields in parallel and perpendicular configuration and for various deformation states. The strain was varied from fully compressed state (i.e. 0%) to fully elongated state (i.e. 6%) in increments of 0.5%. The sample dimensions used for simulating 0% and 6% correspond to $1.55\text{ }\mu\text{m} \times 0.53\text{ }\mu\text{m} \times 0.36\text{ }\mu\text{m}$ and $1.64\text{ }\mu\text{m} \times 0.50\text{ }\mu\text{m} \times 0.36\text{ }\mu\text{m}$. A twin boundary at 45° to the sample was introduced when the strain percent was varied from 0.5 to 5.5 %. The entire simulation had 73728 cells defined such that 384 were along the longest sample dimension and 192 are along the intermediate dimension. Thus at 3% strain, each cell size along the long and intermediate dimensions were 4.17 nm and 2.70 nm respectively. Each simulation ran for 20,000 iterations. To obtain a magnetic structure with minimum energy configuration, we added multiple runs that continue from the previously ended run. In total, we did 380,000 iterations for each simulation condition to obtain the minimum energy state. Simulations were conducted at 100 mT, 150 mT, 200 mT, 250 mT, and 300 mT for all strain

values. The direction of the magnetic field was applied parallel and perpendicular to the longest dimension of the sample (Figure 1). Figures 1a and 1b illustrate the initial sample size and the direction of the magnetic field for perpendicular (fully compressed to 0% strain) and parallel (fully elongated to 6 % strain) sample configurations. The lines inside these rectangular schematics marked as 'c' represent the orientation of the axis of easy magnetization in their fully compressed and elongated states.

Results:

Results of the VSM switching field experiments in parallel and perpendicular sample configuration are shown in Figure 2. The plot is a record of magnetization *vs* external magnetic field $\mu_0 H$. For the sample setup with parallel configuration the magnetization increased linearly until 0.4 T followed by a sudden rise to near saturation. The quick and complete rise indicates that twinning occurred throughout the entire sample. (This event is often referred to as switching.) For the sample setup with perpendicular configuration the increase in magnetization up to saturation occurred gradually over multiple small steps from 0.25 T to 0.38 T.

Figure 3 is a plot of numerically calculated magnetic energy densities with respect to sample deformation at various magnetic fields for the parallel configuration. In this setup since we started the experiment with a fully elongated sample, the deformation started at 6% and proceeded to 0% and the energy density decreased monotonically with deformation. With increasing magnetic field, the slope magnitude increased.

Figure 4 is a plot showing the numerically calculated magnetic energy densities with sample deformation at various magnetic fields for the perpendicular configuration, starting from the fully compressed state. The strain on the abscissa goes from 0 to 6%. At lower field values i.e. at 100

and 150 mT, the energy density decreased initially, went through a local minimum, then increase and went through a local maximum before it decreased again. The local energy maximum was between 4.5 and 5% strain for 100 mT and at 5.5% strain for 150 mT. At magnetic fields equal to or larger than 200 mT, the energy density decreased monotonically with increasing strain. We obtained energy plots for all simulated states, which were 130 (13 strain states, 2 field directions, and 5 field values). In the following, we selected all 13 strain states for the lowest (100 mT) and highest (300 mT) magnetic field values to display the magnetic domain structures (Figure 5). From these states, we selected the states with 100 mT in parallel configuration, fully compressed (0% strain, Figure 6) and fully expanded (6% strain, Figure 7), respectively and with 100 mT in perpendicular (Figure 8) and parallel (Figure 9) configuration at 4.5% strain to highlight the impact of field direction on magnetic energy distributions.

Figure 5(a, b) are the equilibrium magnetic domain structures obtained for parallel sample configuration. Figure 5a and Figure 5b show the magnetic structure evolution for a single twin boundary system starting from 6% to 0% at 100 mT and 300 mT respectively. In both cases, one twin domain had a single magnetic domain structure (represented in red, magnetic moments pointing to the left) while the other twin domain across the twin boundary had multiple magnetic 180° domains (yellow, magnetic moments pointing up, and green, magnetic moments pointing down). The magnetic domain boundaries within the right twin were 180° domain boundaries. The twin boundary carried 90° magnetic domain boundaries where the yellow/red boundary was a head-to-tail boundary and the green/red domain boundary was a tail-to-tail boundary. Figures 5(c) and (d) are the equilibrium magnetic structures obtained for perpendicular sample configuration. Figure 5d shows the magnetic domain structure evolution for a single twin boundary system from 0% to 6% strain at 300 mT. It is similar to Figure 5(a, b) where one variant had a single domain

structure and the other variant across the twin boundary had multiple magnetic domains. In this case, however, the left twin domain with the axis of easy magnetization horizontal had multiple magnetic domains. These results agree with the experimental characterization of magnetization reported by Faran *et al.*[33]. Whereas at 100 mT (Figure 5c) as the magnetic structure evolution occurred from 0% to 6% strain both twin domains contained multiple magnetic domains. Up to a strain of 3.5%, the left twin domain contained one blue magnetic domain (magnetic moments pointing to the right) and the right twin domain had one green magnetic domain. The blue and green magnetic domains met at the twin boundary in head-to-tail configuration. From 4 to 6% strain, additional green magnetic domains formed in the right twin domain. These green magnetic domains extended across the entire sample.

The four energy terms (anisotropy, exchange, stray field, and Zeeman) that are associated with the total magnetic energy calculation are shown for selected cases in Figures 6, 7, 8, and 9. Figure 6 and Figure 7 demonstrate the energies corresponding to 0% and 6% strain respectively at 100 mT for parallel sample configuration. Since there were no magnetic domain boundaries for the 0% strain case (see Figure 5a) all the energies were uniformly distributed across the sample (Figure 6). In the case of 6% strain, since there were multiple magnetic domains (see Figure 5a), the anisotropy and exchange energy was high at magnetic domain boundaries compared to the regions within the domains (Figure 7). Figure 8 and Figure 9 show the energies associated with 4.5 % strain at 100 mT for perpendicular and parallel configuration respectively. In Figure 8, at the twin boundary and magnetic domain boundaries (where the domain orientation changes due to transition) the anisotropy and exchange energies are high compared to the regions that have a uniform orientation of magnetic moments. The stray field energy was quite uniform across the sample but the alternating domain regions (light blue in Figure 8 bottom right, green in Figure 5)

which had the magnetic moments pointing in the direction opposite to the external field had significantly heightened Zeeman energy and lowered stray field energy. Figure 9 represents the same strain state (4.5%, 100 mT) in parallel configuration. While the anisotropy, exchange and stray field energies were high at transition regions compared to the regions with uniform orientation of magnetic moments, the Zeeman energy on the whole fell on the lower end of the energy scale with no distinguished change in energy from one magnetic domain to another.

Figures 10 (a) and (b) show the contribution from each energy term (anisotropy, exchange, stray field and Zeeman energy) towards the total magnetic energy for different strain states at 100 mT in parallel and perpendicular configurations. From comparing the energies in Figure 10 with the magnetic domain evolution in Figure 5 follows that the anisotropy and exchange energies increased with increasing number of magnetic domains in the structure whereas the stray field and Zeeman energies decreased. For the perpendicular field configuration (Figure 10b), at 4% strain and 4.5% strain the Zeeman energy increased while the stray field energy decreased.

Discussion:

Here we qualitatively compare the experimental and numerical results. We do not attempt to compare the experimental and numerical results quantitatively because the volume of the simulated sample is orders of magnitude smaller than that of the experimental sample. Experimental data (Figure 2) shows that for parallel sample configuration switching in the material is abrupt whereas for perpendicular sample configuration it occurs gradually in a step like behaviour. Results from numerical calculations for parallel sample configuration show that the magnetic energy density monotonously decreases with increasing magnetic field and strain (Figure 3). This decrease in energy density explains the spontaneous switching that we see in the

experimental data. Once the magnetic field provides enough driving force to nucleate a twin boundary, the twin boundary moves through the entire sample since the energy continuously decreases as the twin boundary advances. In the case of perpendicular sample configuration (Figure 4), results from numerical calculations at 100 mT show a localized increase in energy from 4 % to 5.5 % strain. With increasing magnetic field, the overall energy becomes lower and so does the local energy maximum at large strain. Above 200 mT magnetic field, the total energy follows the same monotonously decreasing trend as for the parallel configuration. This means that when a twin boundary forms at low magnetic field strength, it can advance only as long as the energy decreases and stops at a strain where the energy is a local minimum. To overcome the energy barrier (i.e. the local maximum) the magnetic field must increase. Since the material remains at the energy valley until the required magnetic field is applied, the twin boundary movement is retarded. This results in gradual, step-like switching.

The shape of the sample plays an important role for the twin boundary motion. The motion of the twin boundary magnetizes the sample and, thus, reduces the Zeeman energy. This is the main driving force for twin domain switching. As the sample gets magnetized, the stray field energy increases. For a parallelepiped bar (present study), in the perpendicular configuration, the magnetic field is perpendicular to the long axis of the sample, which results in a higher demagnetization factor (and, thus, higher stray field energy) compared to the parallel configuration (where the field is parallel to the long axis of the sample). This effect is shown in Figure 10 where the stray field energy increases strongly with increasing strain between 1 and 3.5 % strain for the perpendicular configuration (Figure 10b). In contrast, the stray field energy increases only moderately with ongoing deformation in the parallel configuration (Figure 10a).

To lower the stray field energy the magnetic structure tends to form multiple domains separated by 180° domain walls. This happens for perpendicular and parallel sample configurations (Figure 5). However, in parallel configuration, 180° magnetic domains form only in that twin domain where the axis of easy magnetization is perpendicular to the magnetic field. In this case, the 180° magnetic domains reduce the stray field energy without changing the Zeeman energy. In contrast, because of the large demagnetization factor perpendicular to the longest edge of the sample, 180° magnetic domains form in both twin domains for the perpendicular configuration. This means that in the twin domain with the easy axis of magnetization parallel to the magnetic field, the green domains (in Figure 5c) are magnetized opposite to the direction of the magnetic field. These domains increase the Zeeman energy. The increase in Zeeman energy partially compensates for the decrease in stray field energy.

These results have implications for the design of magnetic shape memory alloy actuators. If one attempts to build an on-off actuator, i.e. a device that switches abruptly between two states, the magnetic field must be applied parallel to the long direction of the magnetic shape memory element. In this configuration, the device switches instantaneously from fully elongated to fully contracted. To switch abruptly from fully contracted to fully extended requires a strong magnetic pulse perpendicular to the sample where the field strength of the pulse is sufficient to saturate the sample. If one attempts to build a positioning actuator capable of adjusting a position gradually, the magnetic shape memory element must be long and the magnetic field must be applied perpendicular to the stroke. Gradual resetting with a magnetic field parallel to the direction of the stroke is not possible since such actuation results in instantaneous and complete switching. Instead, resetting can be achieved with second actuator in “push-push” configuration[34].

Conclusions:

We combined experiments and numerical calculations to study the effect of sample shape on twin boundary mobility. Lowering the Zeeman energy is the main driving force for twin domain switching. As the twin boundary moves through the sample, the sample gets magnetized, which increases the stray field energy. This effect is stronger when the magnetic field is perpendicular to the longest sample extension because of a higher demagnetization factor. The demagnetizing field hinders twin domain switching in the perpendicular configuration. The formation of 180° magnetic domains partially offsets this shape effect. The perpendicular configuration lends itself for a gradual positioning device while the parallel configuration is ideal for an on-off switch.

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Figures:

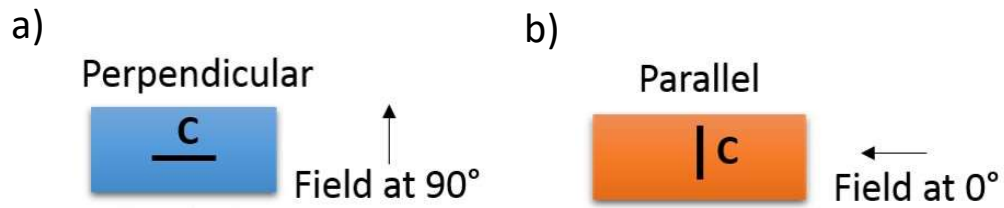


Figure 1: Schematics of samples used for micromagnetics simulations and experimental set up with respect to magnetic field. The direction of magnetic field is indicated by the arrows. (a) Initial sample dimension for perpendicular sample configuration: starting with fully compressed (0% strain) sample and (b) initial sample dimension for parallel configuration: starting with fully elongated (6% strain).

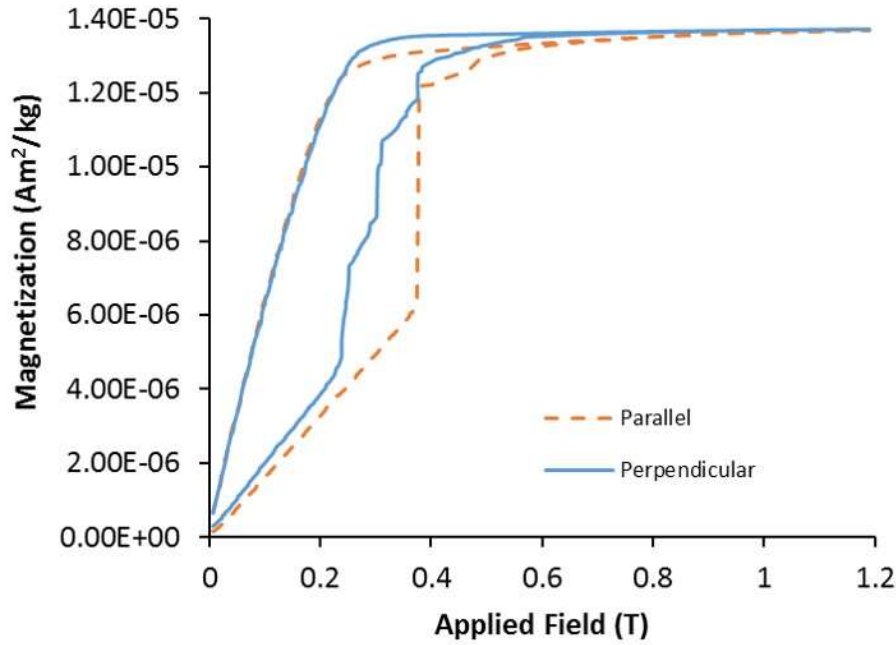


Figure 2: Switching behavior of $\text{Ni}_{49.5}\text{Mn}_{28.8}\text{Ga}_{21.7}$. The curves represent the change in magnetization as a function of applied magnetic field for magnetic fields applied in different directions. The dotted and solid curves correspond to perpendicular and parallel sample configurations.

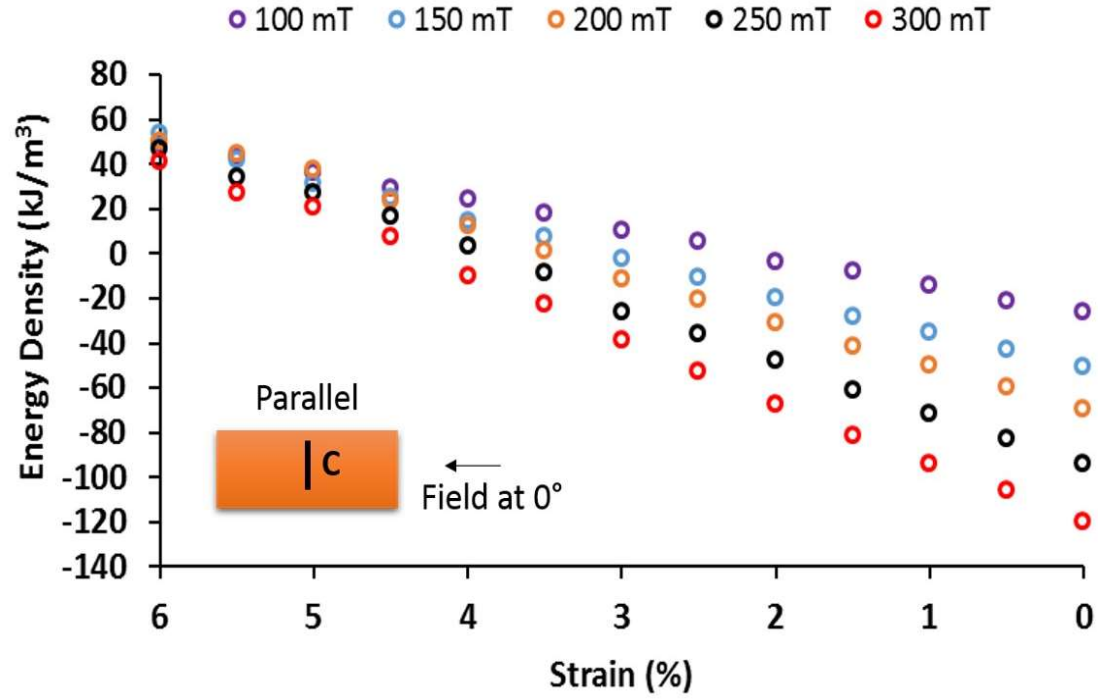


Figure 3: Numerical calculation of magnetic energy densities for different strains in parallel sample configuration. The energy densities are plotted against sample deformation as they occur during an experiment (*i.e.* starting from fully elongated to fully compressed). The inset shows a sample with the direction of easy magnetization (represented by c) and the orientation of the external magnetic field.

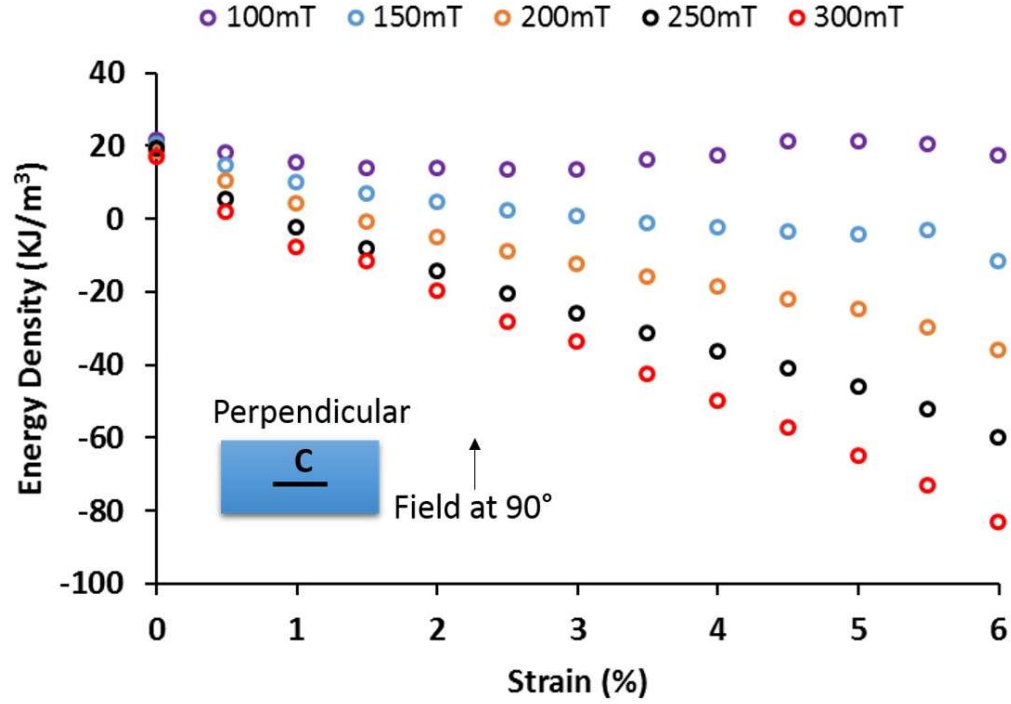


Figure 4: Numerical calculation of magnetic energy densities for as a function of strain in perpendicular sample configuration. The energy densities are plotted against sample deformation as they occur during an experiment (*i.e.* starting from fully compressed to fully elongated). The figure inset shows a sample with the direction of easy magnetization (represented by *c*) and the orientation of the external magnetic field.

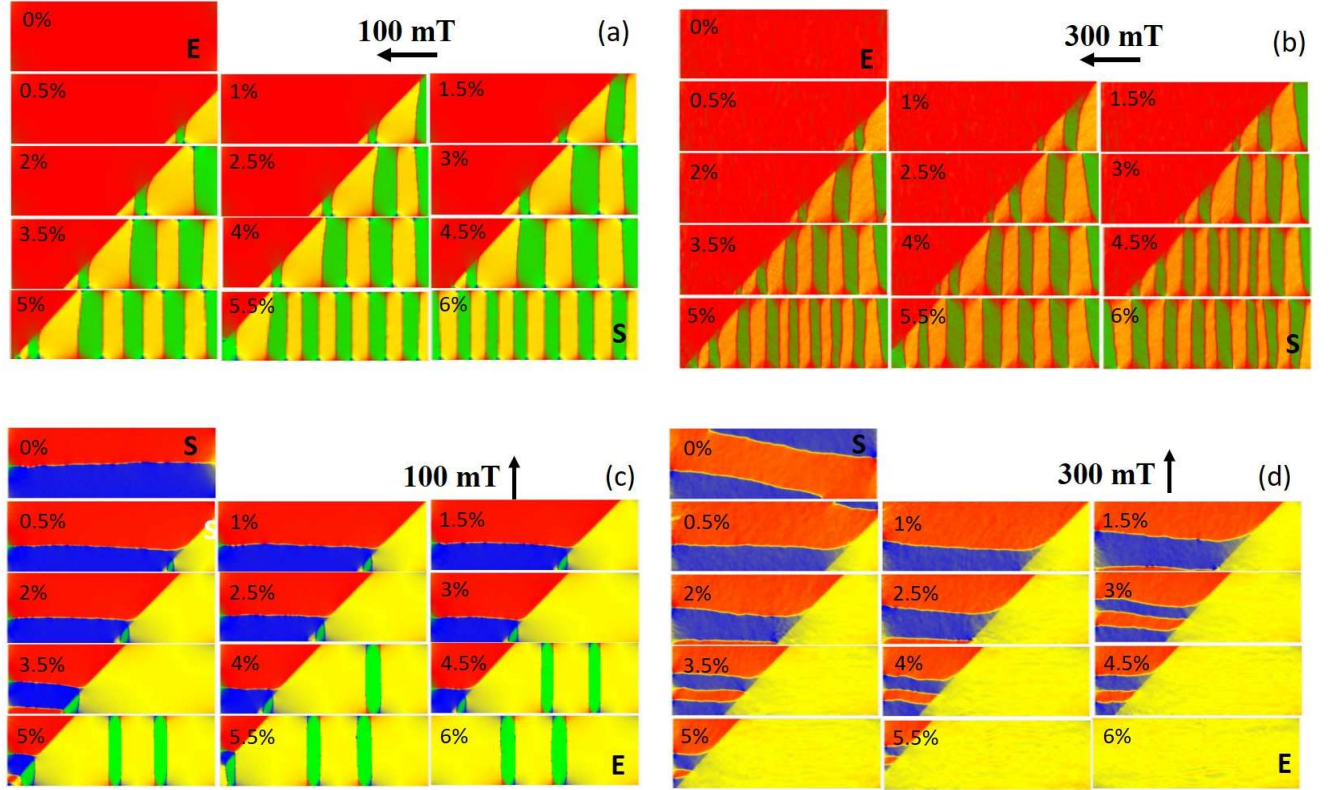


Figure 5: Evolution of the magnetic domain structure for a switching field test at 100 and 300 mT in a single twin boundary state. (a, b) are the equilibrium domain structures for parallel sample configuration and (c, d) are the equilibrium domain structures for perpendicular sample configuration. The letter ‘E’ and ‘S’ indicate the start and end of deformation as the switching field test is performed i.e. the sample deforms from 6 to 0% for parallel and 0 to 6% for perpendicular sample configuration. The colors here indicate the direction of magnetization in the magnetic domains: red (\leftarrow), blue (\rightarrow), yellow (\uparrow), and green (\downarrow)

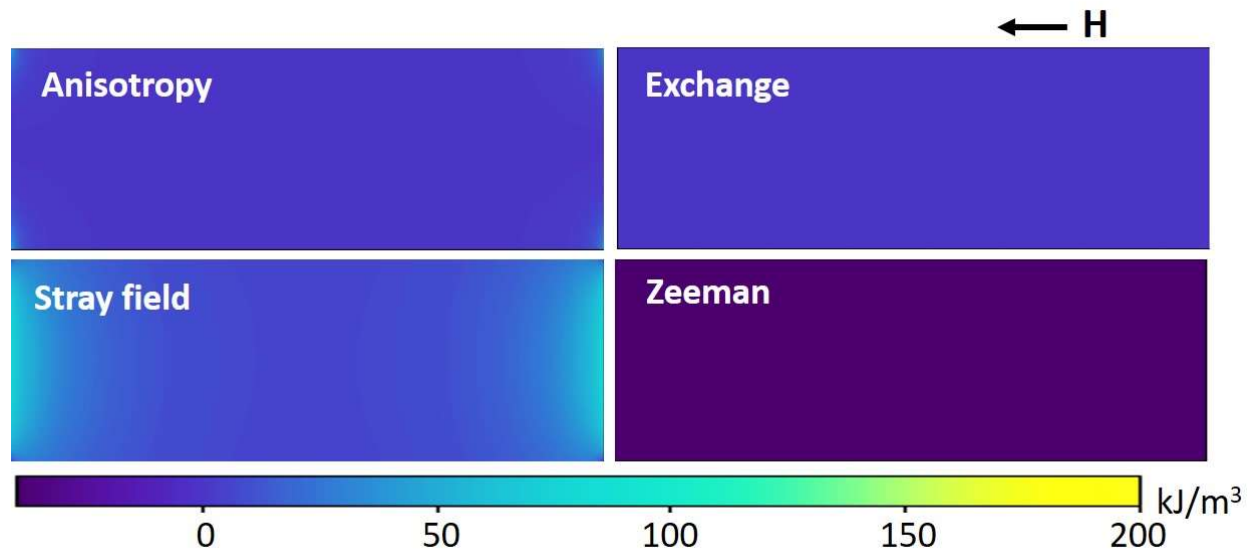


Figure 6: Energy maps at 0% strain in a 100 mT magnetic field for sample setup in parallel configuration. The direction of the field is indicated by the arrow. Each plot as labeled represents the anisotropy, exchange, stray field, and Zeeman energy associated with the magnetic domain structure at equilibrium. The maps are homogeneous because the samples has n twin and magnetic domain boundaries.

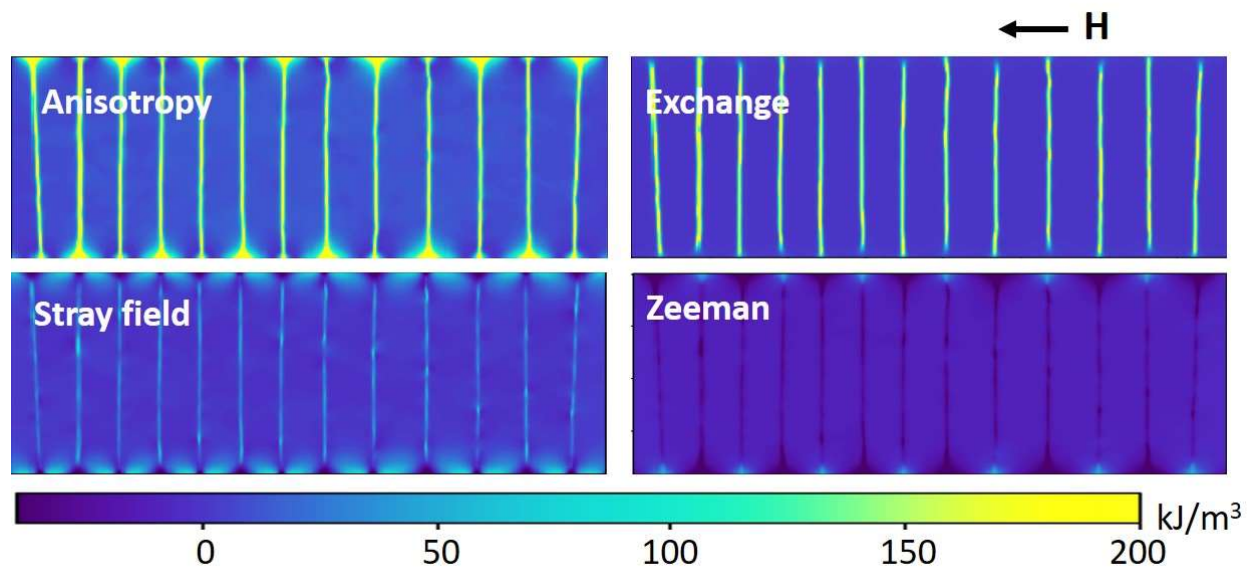


Figure 7: Energy maps at 6% strain in a 100 mT magnetic field for sample setup in parallel configuration. High densities of anisotropy and exchange energy decorate the magnetic domain boundaries.

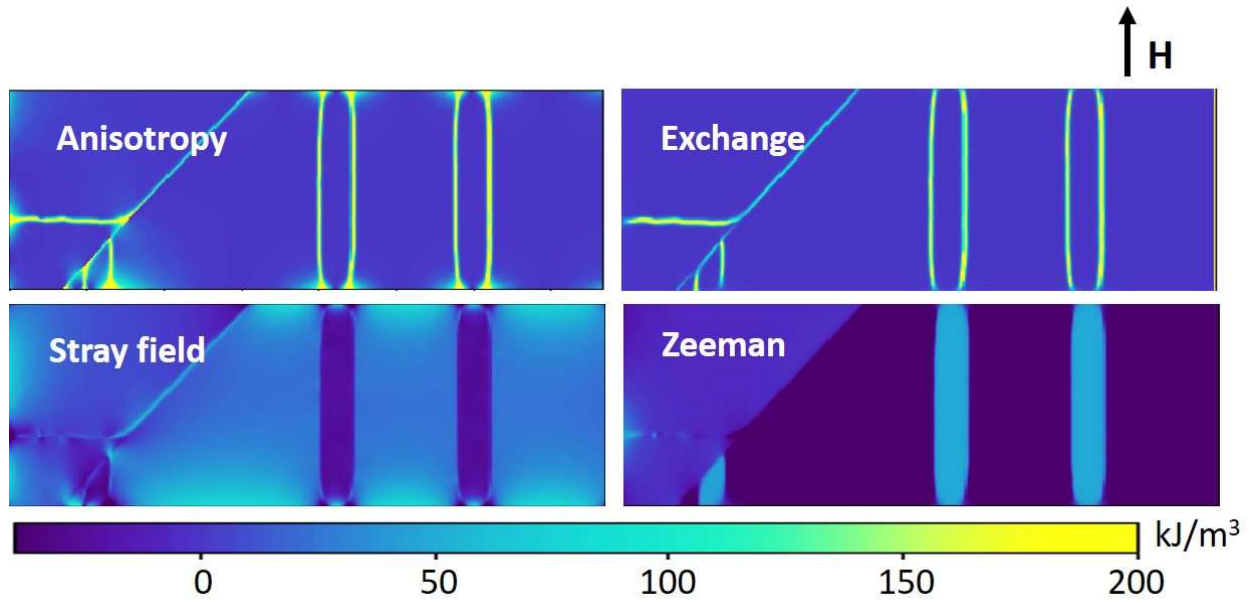


Figure 8: Energy maps at 4.5% strain in a 100 mT magnetic field for sample setup in perpendicular configuration. The twin boundary has lower energy than the magnetic domain boundaries. The vertical magnetic domains with magnetization pointing down (green in Fig. 5) have low stray field and high Zeeman energy.

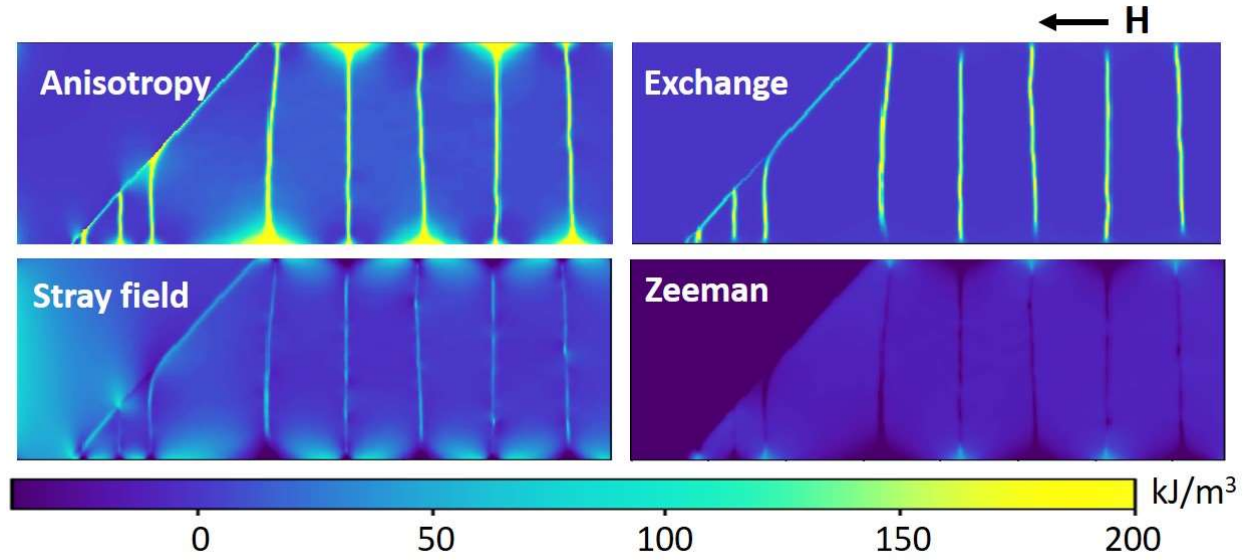


Figure 9: Energy maps at 4.5 % strain in a 100 mT magnetic field for sample setup in parallel configuration. The twin domain with c parallel to the longest edge (red in Fig. 5) has low Zeeman energy.

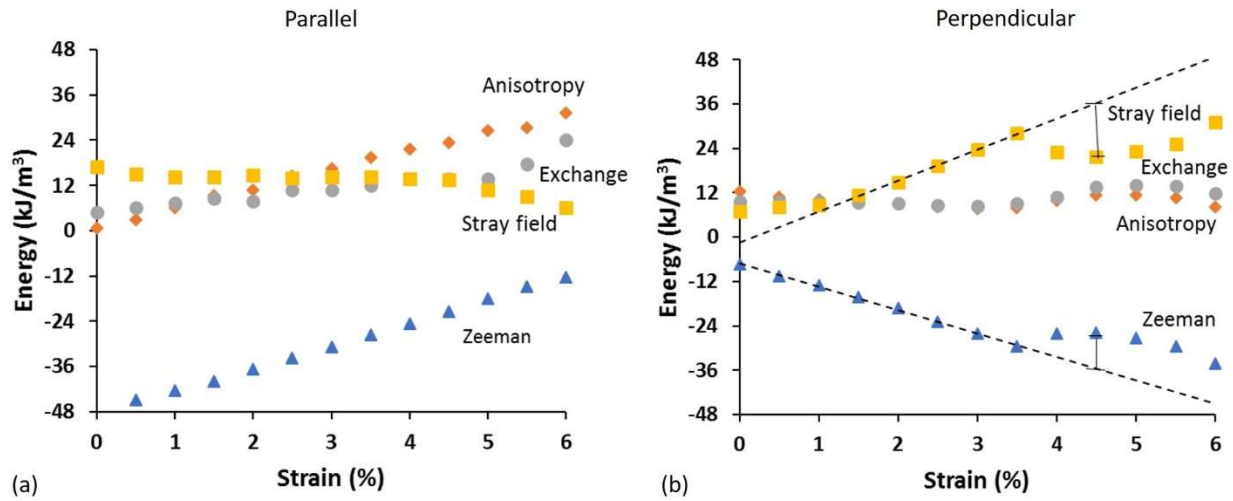


Figure 10: Contributions from anisotropy, exchange, stray field and Zeeman energies to the total magnetic energy of equilibrium magnetic structures obtained at different strain states at 100 mT. (a) In parallel configuration and (b) In perpendicular configuration.