**FINAL REPORT: FG02-01ER-45906 - A novel class of artificially modulated
magnetic multilayers based on magnetic shape memory alloys**

Description/Abstract:

The temperature dependent micromagnetic behavior, structural phase transitions, magnetic transition, and the dynamics of phase transitions have been investigated using magnetic shape memory alloys. Results provide a novel concept of 'polymagnets'. In thin film form the number of martensite variants is greatly reduced. A new technique to study dynamics of magnetic phase transitions has been developed applicable to bulk, thin films or multilayers. Transition pathways in magnetic shape memory alloys are as follows: Structural transitions followed by magnetic transition on cooling, and magnetic transition followed by structural transition on heating. The anisotropy of exchange spring multilayers is greatly sensitive to substrate constraints, and shows a marked rotational hysteresis at lower temperatures. Results also show that a large reduction in coercivity of multilayers is a direct result of broadening of domain wall width by interlayer magnetostatic interactions.

§ A.1. Final Report

§A.1.1. Temperature Dependent Evolution of Martensite Structure

The tetragonal martensite in Ni-Mn-Ga Heusler alloys, Fe-Pd, and Co-Ni-Ga systems forms as twins across the $\{101\}$ planes.^{1,2,3} As is the case with SMAs in general,⁴ the cubic to tetragonal martensite transformation in these alloys results in a complex martensite structure caused by the simultaneous nucleation and growth of a multiplicity of martensite bands across the length of the sample and their subsequent intersection with each other. This is shown sequentially for both forward and reverse martensite transformation in a Ni-Mn-Ga sample in Figure 1 [(a-f) for cooling and (g-l) for heating]. Starting from the high temperature cubic phase in Figure 1(a), a martensite band first nucleates and grows diagonally, as shown sequentially in Figure 1(b-c). This is followed by the growth of another twin band orthogonal to the first band, as shown in Figure 1(d-e). The fully transformed state of the sample is shown in Figure 1(f). Figure 1(g-l) shows the reverse transformation from tetragonal to cubic phase on heating the sample across A_s and A_f temperatures. A comparison of Figure 1(g-l) with micrographs in Figure 1(a-f) shows that although both the forward and reverse transformation occurs by the motion of multiple interfaces, the transformation pathways are an almost identical inverse of each other [for example, compare Figure 1(b) during cooling with Figure 1(k) during heating or Figure 1(e) with Figure 1(i)].

Figure 1. Martensitic transformation in a Ni-Mn-Ga alloy on cooling (a-f), and heating (g-l). Nominal composition of the alloy was $\text{Ni}_{54.35}\text{Mn}_{23.18}\text{Ga}_{22.47}$.

While the micrographs in Figure 1 are aesthetically beautiful, the microstructure is complex. In order to make a meaningful interpretation of the magnetic domain structure, its field and temperature dependence, as well as their dynamic properties, a well defined and controlled structure is required.

Fortunately, from the pioneering work of Roytburd,⁵ we know that the martensite structures can be organized in a hierarchy according to their complexity. This is shown

schematically in Figure 2 using a hypothetical cubic→tetragonal transformation. As a result of reduction in crystal symmetry accompanying the martensitic transformation, structural domains are formed, which are differently oriented variants of the martensite phase. These variants are generally related to each other in a twin-like manner, and are referred to as the ferro-elastic domains. These ferro-elastic domains form a hierarchical polydomain structure to minimize the overall elastic energy of the system.

Figure 2. Hierarchy of polydomain structures for a cubic→tetragonal transformation [5].

For example, in Figure 2, any one of the three cubic axes can transform to become the tetragonal c-axis, giving rise to three spatially distinct variants of the tetragonal phase. These are called zeroth order polydomains. Any two of these three variants can form a plane-parallel assembly of twins, called a polysynthetic twin or simply, a polytwin (1st order polydomains), as shown in the penultimate outer circle in Figure 2. The next level of complexity (2nd order polydomains) consists of taking any two of the three polytwins to form a structure whose elastic energy is lower than that of 1st order polydomains, as shown in the outer circle in Figure 2.

In addition to this complexity, in case of magnetic SMAs the ferromagnetic Weiss domains are magneto-elastically coupled to, and superimposed on the polydomains.

Using the Ni-Mn-Ga, Fe-Pd, and Co-Ni-Ga systems, we have systematically investigated the properties of SMAs according to the hierarchy of polydomains, as described in the following.

§A.1.2. Micromagnetic Structure of Zeroth, 1st, and Higher Order Polydomains

Zeroth Order Structures: Figure 3 shows a Co-Ni-Ga single crystal undergoing martensite transformation. In this particular crystal, the transformation occurs by the motion of a single interface, and the resulting martensite is structurally homogenous, i.e., without the formation of any twins. This constitutes a zeroth order polydomain.

Figure 3. A $\text{Co}_{50}\text{Ni}_{20}\text{Ga}_{30}$ single crystal held between M_s - M_f temperature interval, shown transforming into a single variant martensite. The critical temperatures for this alloy were: $M_s \sim 255$ K, $M_f \sim 237$ K, and $A_s \sim 250$ K, $A_f \sim 264$ K, and Curie temperature ~ 400 K.

The transformation strain ($\sim 6.6\%$) in this alloy is evident in Figure 3 from the sharp bend in the crystal at the austenite-martensite interface. Unlike a twinned crystal where the magnetocrystalline anisotropy axis changes spatially from one crystallographic twin to another, the structurally homogeneous martensite in Figure 3 has a fixed direction of the magnetic easy axis throughout its volume. Viewed along the magnetic easy axis, the micromagnetic structure appears as a series of irregular domains shown in Figure 4(a), with the magnetization vectors going in and out of the plane of the paper. In contrast, the austenite phase consists of a magnetic single domain, as shown in Figure 4(b). [A large single domain austenite state spanning several thousand microns was found to be a common feature of these as well as Ni-Mn-Ga and Fe-Pd alloys, as shown in further detail in the following]. The temperature dependent micromagnetic images in Figure 4 and other figures in the following were taken using the Interference Contrast Colloid (ICC) method, which is discussed in detail elsewhere.^{6,7}

Figure 4. Micromagnetic structure of the Co-Ni-Ga single crystal (a) in the martensite phase, (b) austenite phase.

1st Order Structures: Next consider a {100} $\text{Co}_{49}\text{Ni}_{22}\text{Ga}_{29}$ single crystal that martensitically transforms into a polytwin – a 1st order polydomain. The schematic in Figure 5(b) shows the crystallography of the polytwin (dark and light bands are twins), and the [100] orientation along which the micromagnetic structure was observed in Figure 5(a). The twins in Figure 5(a) are labeledI, II, I, II..., running diagonally from bottom left to top right. Each twin can be seen broken into magnetic domains having anti-parallel magnetization [yellow and green arrows in

Figure 5(a)], separated by 180°-type magnetic domain walls.

Figure 5. (a) Room temperature magnetic structure within the twins of a polytwin in a fully transformed $\text{Co}_{49}\text{Ni}_{22}\text{Ga}_{29}$ {100} single crystal at room temperature. The schematic in (b) shows the twin structure and [100] direction along which sample was viewed; $M_s \sim 341$ K, $M_f \sim 316$ K, and $A_s \sim 346$ K, $A_f \sim 371$ K, and Curie temperature ~ 381 K.

The magnetic arrangement in

Figure 5(a) is exquisite in the manner in which it minimizes its magnetostatic energy - instead of forming closure domains within each domain of a given twin, domains in adjacent twins serve as closure domains for its neighbors, giving the staircase appearance for magnetization vectors running up or down. All magnetization vectors in

Figure 5(a) are in the plane of the twins. When the same polytwin is viewed along the [001] direction [top view in

Figure 5(b)], alternate twins have out-of-plane vectors, as shown in Figure 6.

Figure 6

Figure 6. (a) Micromagnetic structure as seen from the along the [001] direction in the Co-Ni-Ga single crystal of Figure 5. (b) Its corresponding schematic structure.

In Figure 6(a) the twins can be seen running approximately from left to right, and are labeledI, II, I, II.... As in

Figure 5(a), Figure 6(a) shows that each twin is again broken up into magnetic domains, and a corresponding schematic of its micromagnetic structure is shown in Figure 6(b). The out-of-plane projection of magnetization vectors in twins labeled **I** in Figure 6(a) gives rise to conical closure domains within which the magnetization is anti-parallel to

that in the (dark or light) lamellae of twins **I**, as shown schematically in Figure 6(b). For twins labeled **II**, the magnetization lies in-plane, as shown schematically in Figure 6(b). Again the exquisite balance of magnetocrystalline and magnetostatic energy is evident in Figure 6 towards achieving a lowest possible energy state. If the conical closure domains in twins labeled **I** in Figure 6 were to have in-plane magnetization, they would lie along the magnetic hard axis. Instead, by forming conical closure domains with easy axis magnetization, the system avoids paying a price in terms of high magnetocrystalline anisotropy energy in these materials. At the same time, the magnetostatic energy is lowered. It is also of interest to note that conical closure domains were first observed by Chikazumi in high uniaxial anisotropy materials.⁸

a)

b)

Next consider the effect of twin thickness on the micromagnetic structure. Unlike in

Figure 5 or Figure 6, where the twins are tens of microns thick, Figure 7(a) shows the micromagnetic structure of a transformed Ni-Mn-Ga single crystal of nominal composition $\text{Ni}_{54.35}\text{Mn}_{23.18}\text{Ga}_{22.47}$ in which the resulting twins are very fine (of the order of microns, and seen running approximately from left to right). In such a finely twinned martensite, instead of each twin hosting multiple magnetic domains as was the case in

Figure 5 or Figure 6, it is now the magnetic domains that host a large number of fine twins; in Figure 7(a) the magnetic domain walls can be seen traversing the twins from top to bottom. The magnified view in the inset in Figure 7(a) reveals that the domain walls are in fact zigzagged, changing direction with the orientation of the tetragonal c-axis from one twin plate to another. This is shown schematically in Figure 7(b).

Figure 7. Zigzag nature of domain walls from one twin to another in a Ni-Mn-Ga sample at $\sim 21^\circ\text{C}$. Inset shows a magnified view of the zigzag domain walls. (b) Schematic showing the change in tetragonal c-axis and hence magnetic easy axis from one twin to another. The M_s and M_f

temperatures were nominally -18°C and -24°C , respectively and the respective A_s and A_f temperatures were nominally -14°C and -9°C . The Curie temperature was 103°C .

2nd and Higher Order Polydomain Structures: The micrograph in

Figure 8 is taken from another Ni-Mn-Ga single crystal of nominal composition $\text{Ni}_{54.35}\text{Mn}_{23.18}\text{Ga}_{22.47}$. However, it consists of a more complex micromagnetic structure. In

Figure 8, twin bands can be seen running diagonally from bottom left to top right. In addition to the micromagnetic structure within each band (labeled 'intra-domains' in

Figure 8), also note the presence of 'inter-domain' walls that traverse the twin bands. The domain structure in

Figure 8 is a result of a more complex martensite structure, and we have shown that it can be explained by taking into account the fact that these martensite bands are not homogeneous entities but are in fact internally twinned.

Figure 8. Micrograph showing the martensitic magnetic domain structure in a Ni-Mn-Ga single crystal. Note the presence of domains within the bands as well as inter-domain walls spanning several bands.

A striking illustration of the dependence of the micromagnetic structure on twin thickness is shown in Figure 9, which was taken from another area of the above Ni-Mn-Ga sample but where the transformation has resulted in a rapidly varying thickness of the martensite bands. In Figure 9, it can be seen that the magnetic domains in thick twin bands [labeled T1 and T2] are entirely confined within the twins, whereas the micromagnetic structure in finer twins (labeled T3 or T4) is similar to that shown in Figure 7.

Figure 9. Micrograph showing the domain structure in Ni-Mn-Ga, where the sample has transformed into varying thickness of twin bands. Note the presence of inter-domains in thick bands such as T1 or T2, and intra-domain walls in finer twins labelled T3 or T4.

§ A.1.3. Temperature Dependent Evolution of Micromagnetic Structure

In case of zeroth order polydomains such as that shown in Figure 4 above, the effect of temperature is simply to transform the single domain austenite [Figure 4(b)] into a multi-magnetic domain structure in the martensite phase [Figure 4(a)], and *vice versa*.

Figure 10(a-e) shows the temperature dependent evolution of the micromagnetic structure in a Ni-Mn-Ga sample during cooling and heating, and whose composition is similar to the sample shown in Figure 7 above. Figure 10(a) shows that above the M_s temperature, the austenite phase exists as a single magnetic domain. On approaching the M_s temperature, the martensite transformation occurs by the motion of a single interface [shown by black arrows in Figure 10(b)]. In Figure 10(b) the high temperature cubic phase can be seen on the right and the twinned tetragonal martensite on the left. In these ICC micrographs, note the presence of magnetic Weiss domains running normal to the twins. Figure 10(c) shows the sample in the fully transformed state. In Figure 10(c) well defined 180° like magnetic domain walls can be seen more clearly running normal to the twins. [Close to the M_s temperature the tetragonal c/a ratio is close to unity and the manifestation of the transformation strain in the form of surface relief of the twins is faint. Thus the twins are seen more clearly in the fully transformed state in Figure 10(c) in comparison to Figure 10(b)]. The micromagnetic transformation from cubic to tetragonal phase was found to be the reverse of the tetragonal to cubic transformation, as shown in Figure 10(d-e), which leads back to a single domain cubic phase as shown in Figure 10(e).

Figure 10. Micromagnetic evolution as a function of temperature in a Ni-Mn-Ga sample. (a-c) – Cooling; (c-e) – heating.

§ A.1.4. Field Dependent Evolution of Micromagnetic Structure

§A1.4.1. Field induced reorientation of martensite twins

Previously, we had shown preliminary results that the relative volume fraction of the twins changes due to an applied magnetic field.⁹ In this work, the effect of field on mutual accommodation of the twins was studied in detail. This is shown in Figure 11. Domain wall motion due to applied magnetic field in a Co-Ni-Ga sample. The field strength increases from zero in (a) to ~1850 Oe in (i). (a) $H=0$, (b) ~260 Oe, (c) ~380 Oe, (d) ~500 Oe, (e) ~550 Oe (f) ~610 Oe, (g) ~650 Oe, (h) ~1110 Oe, (i) 1850 Oe. The direction of applied field is marked in Error! Not a valid bookmark self-reference.(a), and field strength for each micrograph is given in the figure caption. Starting from zero-field in Figure 11(a), as the field strength increases, the domains in twins labeled I that are favorably oriented (green magnetization vectors in twins labeled I) expand and consume the domains that are unfavorably oriented with respect to the applied field (yellow arrows in twins I). This is shown sequentially in Figure 11(a-c). (a-i) for the Co-Ni-Ga sample described earlier in

Figure 5; the dynamics of wall motion for this sample is discussed separately in a later section.

Figure 11. Domain wall motion due to applied magnetic field in a Co-Ni-Ga sample. The field strength increases from zero in (a) to ~1850 Oe in (i). (a) H=0, (b) ~260 Oe, (c) ~380 Oe, (d) ~500 Oe, (e) ~550 Oe (f) ~610 Oe, (g) ~650 Oe, (h) ~1110 Oe, (i) 1850 Oe.

The direction of applied field is marked in Error! Not a valid bookmark self-reference.(a), and field strength for each micrograph is given in the figure caption. Starting from zero-field in Error! Not a valid bookmark self-reference.(a), as the field strength increases, the domains in twins labeled I that are favorably oriented (green magnetization vectors in twins labeled I) expand and consume the domains that are unfavorably oriented with respect to the applied field (yellow arrows in twins I). This is shown sequentially in Error! Not a valid bookmark self-reference.(a-c). However the

magnetization vectors in all the domains in twins labeled II in Figure 11. Domain wall motion due to applied magnetic field in a Co-Ni-Ga sample. The field strength increases from zero in (a) to ~1850 Oe in (i). (a) H=0, (b) ~260 Oe, (c) ~380 Oe, (d) ~500 Oe, (e) ~550 Oe (f) ~610 Oe, (g) ~650 Oe, (h) ~1110 Oe, (i) 1850 Oe.

The direction of applied field is marked in Error! Not a valid bookmark self-reference.(a), and field strength for each micrograph is given in the figure caption. Starting from zero-field in Error! Not a valid bookmark self-reference.(a), as the field strength increases, the domains in twins labeled I that are favorably oriented (green magnetization vectors in twins labeled I) expand and consume the domains that are unfavorably oriented with respect to the applied field (yellow arrows in twins I). This is shown sequentially in Error! Not a valid bookmark self-reference.(a-c). are orthogonal to the field direction, i.e., along the hard axis of twins II. As a result, the domain walls in twins II do not move at lower fields. In describing the zero-field micromagnetic structure for this sample in

Figure 5, it was noted that the domains in each twins also serve as closure domains for its neighbors. The consequence of this magnetostatic coupling is a pinning of domain walls in twins I by domain walls in twins II, as shown sequentially in Figure 11. Domain wall motion due to applied magnetic field in a Co-Ni-Ga sample. The field strength increases from zero in (a) to ~1850 Oe in (i). (a) H=0, (b) ~260 Oe, (c) ~380 Oe, (d) ~500 Oe, (e) ~550 Oe (f) ~610 Oe, (g) ~650 Oe, (h) ~1110 Oe, (i) 1850 Oe.

The direction of applied field is marked in Error! Not a valid bookmark self-reference.(a), and field strength for each micrograph is given in the figure caption. Starting from zero-field in Error! Not a valid bookmark self-reference.(a), as the field strength increases, the domains in twins labeled I that are favorably oriented (green magnetization vectors in twins labeled I) expand and consume the domains that are unfavorably oriented with respect to the applied field (yellow arrows in twins I). This is shown sequentially in Error! Not a valid bookmark self-reference.(a-c). (c-g), marked within the dotted lines. Starting from Figure 11. Domain wall motion due to applied magnetic field in a Co-Ni-Ga sample. The field strength increases from zero in (a) to ~1850 Oe in (i). (a) H=0, (b) ~260 Oe, (c) ~380 Oe, (d) ~500 Oe, (e) ~550 Oe (f) ~610 Oe, (g) ~650 Oe, (h) ~1110 Oe, (i) 1850 Oe.

The direction of applied field is marked in Error! Not a valid bookmark self-reference.(a), and field strength for each micrograph is given in the figure caption. Starting from zero-field in Error! Not a valid bookmark self-reference.(a), as the field strength increases, the domains in twins labeled I that are favorably oriented (green magnetization vectors in twins labeled I) expand and consume the domains that are unfavorably oriented with respect to the applied field (yellow arrows in twins I). This is shown sequentially in Error! Not a valid bookmark self-reference.(a-c). (c), as the walls in twins I try to move parallel to themselves, pinning due to magnetic domain walls in twins II

causes them to bend, as shown sequentially within dotted lines in Figure 11. Domain wall motion due to applied magnetic field in a Co-Ni-Ga sample. The field strength increases from zero in (a) to ~1850 Oe in (i). (a) H=0, (b) ~260 Oe, (c) ~380 Oe, (d) ~500 Oe, (e) ~550 Oe (f) ~610 Oe, (g) ~650 Oe, (h) ~1110 Oe, (i) 1850 Oe.

The direction of applied field is marked in Error! Not a valid bookmark self-reference.(a), and field strength for each micrograph is given in the figure caption. Starting from zero-field in Error! Not a valid bookmark self-reference.(a), as the field strength increases, the domains in twins labeled I that are favorably oriented (green magnetization vectors in twins labeled I) expand and consume the domains that are unfavorably oriented with respect to the applied field (yellow arrows in twins I). This is shown sequentially in Error! Not a valid bookmark self-reference.(a-c). (c-d). With a further increase in field strength, eventually the walls in twins I snap away from walls in twins II, leaving behind a new set of domains in twins II that have their own closure domains, as shown

sequentially in Figure 11. Domain wall motion due to applied magnetic field in a Co-Ni-Ga sample. The field strength increases from zero in (a) to ~1850 Oe in (i). (a) H=0, (b) ~260 Oe, (c) ~380 Oe, (d) ~500 Oe, (e) ~550 Oe (f) ~610 Oe, (g) ~650 Oe, (h) ~1110 Oe, (i) 1850 Oe.

The direction of applied field is marked in Error! Not a valid bookmark self-reference.(a), and field strength for each micrograph is given in the figure caption. Starting from zero-field in Error! Not a valid bookmark self-reference.(a), as the field strength increases, the domains in twins labeled I that are favorably oriented (green magnetization vectors in twins labeled I) expand and consume the domains that are unfavorably oriented with respect to the applied field (yellow arrows in twins I). This is shown sequentially in Error! Not a valid bookmark self-reference.(a-c). (d-g). Only when the field strength is sufficiently high, the magnetization vectors in twins II begin to rotate along the field direction, as shown in Figure 11. Domain wall motion due to applied magnetic field in a Co-Ni-Ga sample. The field strength increases from zero in (a) to ~1850 Oe in (i). (a) H=0, (b) ~260 Oe, (c) ~380 Oe, (d) ~500 Oe, (e) ~550 Oe (f) ~610 Oe, (g) ~650 Oe, (h) ~1110 Oe, (i) 1850 Oe.

The direction of applied field is marked in **Error! Not a valid bookmark self-reference.**(a), and field strength for each micrograph is given in the figure caption. Starting from zero-field in **Error! Not a valid bookmark self-reference.**(a), as the field strength increases, the domains in twins labeled I that are favorably oriented (green magnetization vectors in twins labeled I) expand and consume the domains that are unfavorably oriented with respect to the applied field (yellow arrows in twins I). This is shown sequentially in **Error! Not a valid bookmark self-reference.**(a-c). (h-i). This is accompanied by a collapse of twins, and the field-induced strain in the material.

§ A.1.4.2. Field induced martensite transformation

Figure 12(a-g) is a sequence of micrographs showing the first direct evidence of field induced martensite transformation using the same Ni-Mn-Ga single crystal as in Figure 7 (and Figure 10) above. The direction of applied field in

Figure 12 is in the vertical direction. The sample was cooled from room temperature and held at 2 °C above the M_s temperature.

Figure 12(a) shows the sample in the absence of any magnetic field, where it is in single domain cubic phase.

Figure 12(a-d) shows that the transformation of the cubic phase into twins with progressively increasing field strength from zero in

Figure 12(a) to 800 Oe in

Figure 12(d).

Figure 12(d-g) shows that the sample reversibly transforms back into the cubic phase as the field strength was reduced to zero. Note the complete reversibility of the martensite transformation as a function of applied field. This behavior is remarkably similar to the stress induced martensite transformation in thermo-elastic SMAs, which is commonly referred to as pseudo-elasticity. Therefore, with this analogy, the present behavior in FSMAs represents a 'pseudo-magneto-elastic' behavior in these alloys.

Figure 12. Field induced transformation 2 °C above the martensite temperature. Approximate field strength is: (a) $H=0$, (b) $H=640$ Oe, (c) $H=720$ Oe, (d) $H=800$ Oe, (e) $H=720$ Oe, (f) $H=640$ Oe, (g) $H=0$.

Also note the difference in twin thickness with respect to the finer twins in Figure 10 as well as the absence of magnetic domains either inside or across the twins in

Figure 12. Also, the ability of the sample to transform from the cubic to tetragonal phase in fields as low as 800 Oe is noteworthy since theoretical calculations based on a modified Clausius-Clapeyron equation relating the field dependence of the critical transformation temperature predicts field strength of several thousand oersteds even a few degrees above the transformation temperature.¹⁰

§ A.1.5. Dynamical Behavior of Magnetic SMAs

The above discussion highlights the static or quasi-static properties of magnetic SMAs. These results describe a given state of the system rather than the path taken in response to a given stimuli (magnetic, elastic, or thermal). The logical next step in understanding the behavior of these complex materials is to study their dynamical characteristics. This refers to a fundamental investigation to identify the various pathways(s) available to the system in going from one state to another, thereby providing a basis for control and optimization. Two case studies done by PI on probing the transition pathways are discussed in the following. The results are significant not only because it is a logical progression in fully characterizing these materials, but because it would ultimately provide a basis for the control and optimization of their dynamic properties.

The high temperature austenite phase in magnetic SMAs is characterized by a different set of magnetic properties (magneto-crystalline anisotropy, moment, etc.) than those in the low temperature martensite phase. Therefore when a magnetic SMA undergoes structural transformation, it also simultaneously experiences a magnetic transition. Our first case study has helped delineate the relative sequence of magnetic transition vis-à-vis martensite transformation in magnetic SMAs, using the Ni-Mn-Ga, Fe-Pd, and Co-No-Ga family of alloys. We have found the sequence of structural and magnetic transitions in all these alloys to be as follows:

On cooling: structural transition followed by magnetic transition

On heating: magnetic transition followed by structural transition

Of course this relative sequence holds for the specific case of zero-field and zero bias stress.

We determined this relative sequence by developing a new high-speed electronic method to study temperature (or pressure) dependent magnetic domain dynamics, called 'magnetic transition spectra' or MTS. It is based on the same principle upon which the well known and established Barkhausen method is based,¹¹ namely, Faraday's law: voltage ξ induced in a pickup coil is proportional to the rate of change of flux with time $\xi = -d\Phi / dt$ in a sample; $d\Phi$ is the flux change over a time interval dt . Whereas the Barkhausen method generates a spectrum of voltage spikes by placing a pickup coil next to a ferromagnetic sample and cycling the sample in an applied magnetic field, see for example, Ref. [12], the MTS method generates a spectrum of

voltage by sweeping the sample across the transformation temperatures instead of an applied magnetic field; the same approach when used by applying load instead of varying temperature gives dynamics of pressure-induced transitions. To illustrate, consider the case of temperature induced MTS. Due to the high speed of the martensite transformation, a catastrophic or avalanche-like reconfiguration of the magnetic domain structure occurs in the crystal on passing from the single domain austenite phase to multi-domain, multi-twinned martensite phase. This gives rise to a spectrum of voltage spikes reflecting the dynamics of the magnetic transition. The resulting spectrum of voltage spikes as a function of temperature is recorded by an adjacent pickup coil. [Details of this technique are given in the following section].

Figure 13(a) shows the acquired magnetic transition spectrum accompanying the martensite transformation in a Ni-Mn-Ga sample. As shown in Figure 13(a) the MTS consists of a very large number of voltage spikes reflecting the dynamics of micromagnetic reconfiguration. The inset in Figure 13(a) is a magnified view of the acquired MTS spectrum, which clearly shows the avalanche-like distribution of the induced pulses. Previously, we have used a ‘jumpsum’ method to analyze and interpret voltage spectra (Barkhausen spectra) in ferromagnetic materials.^{12,13,14} The jumpsum analysis method is well suited because instead of assigning an average or mean value to a given spectrum, it expresses the acquired signal in terms of the profile of the spectrum.

Figure 13. (a) Magnetic transition spectrum for a Ni-Mn-Ga sample on cooling. Note that the jumps occur below the martensite transformation. The critical temperatures for this alloys were also separately measured by calorimetry with *in-situ* optical imaging (using Linkam, UK *in-situ* DSC stage). (b) The JS curve corresponding to the MTS in (a). The inset in (a) is a magnified view of the MTS over a small temperature interval,

and shows the avalanche-like behavior of the jumps. The inset in (b) is a magnified view, which shows that the JS curve consists of a large number of jumps.

Thus with respect to Figure 13(a), one of the signal parameters extracted from the MTS is called the JumpSum JS , which is simply the running total of all the voltage jump heights, as shown in Figure 13(b) – the reverse S-curve. The i^{th} value of JS is equal to the sum of all preceding voltage jumps $\sum_{i=1}^{i=i} \xi_i$ up to that temperature on cooling. The

inset in Figure 13(b) is a magnified view of a portion of the JS curve, which shows that the JS curve is in fact made up of a large number of jumps or steps. The M_s and M_f temperatures for this alloy were measured using both calorimetry and optical observations, and are also indicated in Figure 13. As seen from Figure 13(b), JS value is zero above the M_s temperature and rises to a saturation value within a narrow temperature interval $\Delta T = T_s - T_f$ of 1.5 degrees (the essentially zero initial slope of JS curve in Figure 13(b) is due to small magnitude of jumps at the start of the transformation). In other words, *the structural transition precedes the micromagnetic reconfiguration on cooling*. Another parameter derived is called the JumpSum Rate JSR , which is simply the rate at which flux is being emitted by the sample.

A composite of JS and JSR curves for both cooling and heating cycles for the Ni-Mn-Ga sample is shown in Figure 14; similar curves were obtained for the Fe-Pd alloys. [Note

that the JS and JSR curves in Figure 13 and Figure 14 start from right to left for cooling, and from left to right for heating]. The noteworthy feature of JS and JSR curves in Figure 13 and Figure 14 is the clear indication that during cooling, the magnetic transition occurs after the structural transformation, whereas on heating the magnetic transition is complete before the structural transition begins. In other words, the sequence of structural and magnetic transitions was found to be as follows: *Cooling: structural transition $\rightarrow$ magnetic transition; Heating: magnetic transition $\rightarrow$ structural transition.*

Figure 14. Composite of JS and JSR curves for cooling and heating. Note: For cooling the JS curve goes from right to left whereas for heating the JS curve goes from left to right.

The precedence of the martensite transition over magnetic transition, say, during cooling, is further highlighted in Figure 15, which shows a collage of micrographs that were acquired *in-situ* with the MTS measurements and overlaid on the JS curve of Fe-30at%Pd alloy. Unlike the Ni-Mn-Ga system where the martensite transition occurs by the formation of very fine twins through the motion of a single interface across the surface of the sample, martensite transformation in the Fe-Pd alloy occurs by the abrupt, burst-like transformation of large volumes of the crystal into thick twin bands. This jerky and burst-like mode of transformation makes it easier to further illustrate the relative sequence of magnetic and structural transitions.

Figure 15. Collage showing the JS curve for Fe-30at%Pd sample and the corresponding change in microstructure responsible for each jump in the JS curve. The inset shows JS and JSR curves for cooling and heating.

The collage in Figure 15 shows that the formation of each set of twin bands leads to a sharp increase in the JS value. The micrographs in the collage in Figure 15 were acquired *in-situ* with MTS measurements and they show unambiguously the sequence

of magnetic and structural transitions. The inset in Figure 15 shows the *JSR* curves for Fe-Pd during cooling and heating.

The avalanche-like behavior (as a general phenomenon) and avalanches in the Barkhausen spectrum due to an applied magnetic field are well studied.¹⁵ In particular, Antoni Planes¹⁶ has studied acoustic emission in a *non-magnetic* Cu-Zn-Al shape memory alloy. They have observed avalanche-like behavior associated with the formation of *non-magnetic* martensite plates. In addition, Hardy *et al.*¹⁷ have shown the time dependent evolution of magnetization in manganite in a constant field. The conclusion of the present study is that the magnetic transition is enslaved to the structural transition. Since MTS in the present study was taken in the *absence* of any applied magnetic field, avalanche-like behavior in the MTS signal seems to show an enslaved magnetic signal arising from the avalanche-like behavior of martensite plates. While statistical analysis jump of the spectra for jump amplitudes was beyond the scope of this work, these observations provides an interesting framework for further studying the athermal and time-dependent nature of martensite transformation in ferromagnetic magnetic shape memory alloys in the future.

It is also interesting to note the similarities of the *JSR* curves in the insets of Figure 14 and Figure 15 with curves obtained during magneto-caloric measurements by Planes group.¹⁸ Such a comparison is worthwhile in the future as it could provides insight into the interplay between magnetic, elastic and caloric effects occurring during the transformation in these alloys.

In our second case study, we have looked at the dynamics of domain wall motion at a fixed temperature and zero stress in a Co-Ni-Ga {100} single crystal.

Figure 16. (a) The magnetic structure within the twins of a polytwin in a fully transformed $\text{Co}_{49}\text{Ni}_{22}\text{Ga}_{29}$ {100} single crystal at room temperature. The schematic in (b) shows the twin structure and [100] direction along which sample was viewed; $M_s \sim 341$ K, $M_f \sim 316$ K, and $A_s \sim 346$ K, $A_f \sim 371$ K for heating, and Curie temperature ~ 381 K.

With reference to its zero-field micromagnetic structure as viewed along the [100] axis shown schematically in Figure 16(b), first consider the case when the magnetic field is applied parallel (or anti-parallel) to magnetization vectors in twins labeled I in Figure 16(a). Therefore, the field lies along the easy axis in twins labeled I, whereas it is orthogonal to the easy axis for domains in twins labeled II. Therefore when the field strength is increased, the domain walls in twins I are easily displaced, without exerting any pressure on the twin walls to move. At sufficiently high fields, the magnetization vectors in twins II begin to rotate along the field direction, accompanied by motion of twin walls causing strain. While the micromagnetic (quasi-static) picture of this process, involving extinction of domains in twins I and subsequent nucleation of new sub-domains in twins II was described earlier in Figure 11. Domain wall motion due to applied magnetic field in a Co-Ni-Ga sample. The field strength increases from zero in (a) to ~1850 Oe in (i). (a) $H=0$, (b) ~260 Oe, (c) ~380 Oe, (d) ~500 Oe, (e) ~550 Oe (f) ~610 Oe, (g) ~650 Oe, (h) ~1110 Oe, (i) 1850 Oe.

The direction of applied field is marked in **Error! Not a valid bookmark self-reference.**(a), and field strength for each micrograph is given in the figure caption. Starting from zero-field in **Error! Not a valid bookmark self-reference.**(a), as the field strength increases, the domains in twins labeled I that are favorably oriented (green magnetization vectors in twins labeled I) expand and consume the domains that are unfavorably oriented with respect to the applied field (yellow arrows in twins I). This is shown sequentially in **Error! Not a valid bookmark self-reference.**(a-c). , study of the dynamics of domain walls helps delineate and differentiate the effect of applied field in causing the material to strain. Figure 17(a) shows the jumpsum curve for this particular field orientation. As shown in Figure 17(a), a kink in the curve is now present. The rise in jumpsum on the left of the kink corresponds to simple dynamics of magnetic domain walls in twins labeled I. The rise in jumpsum on the right side

of the kink corresponds to the dynamics of magnetic domains lying in twins labeled II. Since it is the latter that causes strain, this dynamic picture helps reveal the fraction of energy being input to actually cause the useful strain.

In contrast, when the field orientation lies along the twin walls (which run diagonally from bottom left to top right in Figure 16), the field lies along the hard axis for both twins I and II. In this case, the strain is caused by both twins I and II, as reflected by the almost kink-free jumpsum curve in Figure 17(b). In other words, almost all the magnetic energy that is input goes directly to cause useful strain.

These dynamical results pertain to a specific case of fixed temperature and zero bias stress. It reveals the potential for numerous transition pathways that could be mapped,

either by stress-biasing or change in temperature, in order to choose an optimal combination of the given parameters.

Figure 17. Jumpsum curves for the Co-Ni-Ga sample shown in Figure 16. (a) when the field is normal to the twins shown in Figure 16, and (b) when the field is parallel to the twin walls.

§ A.1.6. The Technique of Magnetic Transition Spectra

The structural transformation in FSMAs is martensitic, which owing to its displacive and diffusionless nature proceeds at a speed approaching the transmission of elastic waves in a solid, modified (and slowed) by an appropriate damping factor and softening of the elastic constants in the premartensitic state. Accompanying the martensite transformation in FSMAs is a reconfiguration of the micromagnetic structure. Therefore a suitable method is required to provide simultaneous real-time information on both transitions. Whereas the martensitic transformation can be observed directly in an optical microscope using, say, a Nomarski interferometer, simultaneous monitoring of the high speed magnetic transition using established techniques (such as the polar Kerr effect, magnetic force microscopy, Bitter method, or the ICC method) prove to be too cumbersome and/or slow or inconclusive. This is especially true when the two different transitions occur at different speeds and have to be followed simultaneously as a function of temperature (or pressure) in a sample contained in the protective environment of a heating/cooling stage. Therefore a novel and simple method was developed, which we refer to as the 'magnetic transition spectra' (MTS) method.

The MTS method is an electronic method to monitor the change in micromagnetic structure as a function of temperature or pressure. It is based on the same principle upon which the well known and established Barkhausen method is based, namely, Faraday's law: voltage ξ induced in a pickup coil is proportional to the rate of change of flux with time $\xi = -d\Phi/dt$ in a sample; $d\Phi$ is the flux change over a time interval dt .

Whereas the Barkhausen method generates a spectrum of voltage spikes by placing a pickup coil next to a ferromagnetic sample and cycling the sample in an applied magnetic field, the MTS method generates a spectrum of voltage by *sweeping the sample across the transformation temperatures (or load)* instead of applying any magnetic field.

Figure 18. Schematic of the experimental setup for acquiring the magnetic transition spectrum. (T/C: thermocouple).

As a result of phase transformation (due to temperature, or load), either during forward or reverse direction, change in flux occurs in the sample. The resulting

spectrum of voltage spikes as a function of temperature (or load) is recorded by an adjacent pickup coil. The MTS method is especially well suited for studying magnetic transitions in FSMA's because the local change in flux occurs over short intervals of time, which in turn leads to a high output signal.

The experimental setup used in the MTS method is the same as that for the Barkhausen method. However, unlike the Barkhausen method where an energizing coil is used to generate a magnetic field to sweep the sample, a heating/cooling stage (or tensile stage) is used in the MTS method, as shown schematically in Figure 18. The MTS signal pickup coil is a specially designed and optimized miniature surface probe fabricated out of an insulating ferrite core. Its output is bandwidth limited by a band pass filter to a range of 1-10 kHz. The pickup coil is a miniature probe in the form of a hemispherical ring 2-3 mm in diameter, 1 mm thick and having 50 encircling coil turns. To explain, consider the case of varying temperature (extension to load is straightforward). The sample is attached to the probe and placed in a commercial temperature stage that is capable of heating and cooling the sample from 77 K to 873 K to within ± 0.1 °C. The heating/cooling stage is fully automated and interfaced with an image frame grabber and a synchronized high speed data acquisition card. Furthermore, the experimental setup acquires and labels all images and data automatically following the execution of a pre-programmed heating/cooling cycle. The software operating the stage and acquisition cards also has the ability to embed important experimental information (temperature, heating/cooling rate, etc.) directly on the recorded optical micrographs, which themselves are collated and numbered automatically by the software. This degree of automation ensures unambiguous correlation between the microstructure and the acquired MTS spectrum as a function of temperature. The samples were cooled/heated from rates as low as 0.1 °C/min to 30 °C/min. The MTS spectra were found to be independent of the measured heating/cooling rates because the transformation occurs at speeds much faster than the temperature scans. Moreover, the transition temperatures were found to be unaffected by the presence of the miniature pickup coil lying adjacent to the samples. As the sample is cooled or heated across its critical transformation temperatures, the signal from the MTS coil is digitized by the computer and analyzed subsequently.

Finally, the MTS method is simple to implement and can be used to study other magnetic phase transitions driven by *any* external influence that would cause an abrupt change in the micromagnetic state of the sample (change in temperature, pressure, etc.). For example, we have successfully applied this method to study the onset of coupling in exchange anisotropy ferromagnetic films coupled to an antiferromagnetic substrate, using the Co-CoO systems. Other potential applications of MTS include fundamental investigation of ductile to brittle transition in ferromagnetic structural materials and pressure-induced phase transitions. One current limitation of the method, however, is the upper temperature limit. While it is easy to implement the technique for low temperature studies, we are implementing modifications, both in the probe design and signal processing, but this is not critical to the proposed studies. In short, the MTS method could find applications in many fundamental investigations.

§ A.1.7. Synthesis of Magnetic SMA Thin Films and Multilayers

To undertake similar studies in thin films and multilayers, a combinatorial approach to materials synthesis was used in order to systematically identify useful thin film growth conditions. The PI's UHV deposition chamber is equipped with thin masks of varying aperture sizes that can be translated by high precision micrometers. In this manner, combinatorial libraries consisting of 4x4 arrays of samples and 9x9 libraries were made. Such a 9x9 (=81 samples) library is shown in Figure 19(a), made by dc magnetron co-sputtering of an equi-atomic NiMnGa target with a Ni target. In this particular library, the composition was varied as a function of sputtering power for the two targets, other conditions being the same. Various compositions of interest so obtained are shown in Figure 19(b); several nearly identical compositions so found are not shown.

Figure 19. (a) 9x9 combinatorial library of Ni-Mn-Ga Heusler films made by sputtering. The film diameter is approx. 1 mm. (b) Various compositions of interest in the ternary Ni-Mn-Ga system obtained by this combinatorial method.

Figure 20(a-c) illustrates reversible transformation in one such nearly stoichiometric Ni-Mn-Ga thin film, which undergoes martensitic transition at $\sim -65^\circ\text{C}$.

Figure 20. Reversible transformation in Ni-Mn-Ga SMA from cubic (a) to martensite (b) to (c) cubic states upon cooling and then heating back.

In addition, we have also made Co-Ni thin films by electrodeposition based on the following idea. The threshold stresses for motion of interfaces separating the martensite variants depends upon the crystallography and energy of the interfaces, and the crystal symmetry of the martensite itself, in addition to the usual requirements of control over alloy composition and defect structure. In this regard, the Co-Ni system is of special interest. The Co-Ni-based ferromagnetic SMAs undergo one of the simplest martensitic transformation, *viz.*, from α (face centered cubic) to ϵ (hexagonal close packed), with both fcc and hcp being close packed structures.^{19,20,21} Thus in contrast to the martensite variants that exist as twins in Fe-Pd, Co-Ni-Ga, or the Ni-Mn-Ga alloy systems, martensite variants in case of Co-Ni system exist as a series of stacking faults. Owing to the low stacking fault energy in Co and Co-based alloys such as Co-Ni, α/ϵ interfaces can move with relative ease through

expansion or contraction of the stacking faults. To date, we have successfully determined the optimum thin film growth conditions for which martensite transformation can be observed in thin alloy system. Our results further show that the Co-Ni

ferromagnetic SMA films can be easily processed by electrochemical deposition over a wide range of chemical composition through control over chemical composition of the electrolyte and deposition parameters. As shown in Figure 21, the relative concentration of Co and Ni can be varied over a large range simply by changing the applied voltage for electrodeposition. As a result, Martensite transformations in films having transformation temperatures ranging from -125 °C to -60 °C have been obtained.

Figure 21. Controlled variation in Co:Ni ratio in electrodeposited Co-Ni thin films by changing the bias voltage.

§ A.1.8. Thin Film, Multilayers, and Temperature Dependence of their Magnetic Properties

The thin film geometry imposes special restrictions on the micromagnetics of magnetic

SMA as a direct result of their ferromagnetism. This is illustrated schematically in Figure 22 for a cubic→tetragonal transformation with c/a ratio less than unity (e.g., the Fe-Pd system). If the film lies along the X-Y plane, the Z-axis variant is prohibited due to the exorbitant price that would have to be paid from the associated out-of-plane magnetostatic energy ($\sim 2\pi M_s^2 = 2.1 \times 10^7 \text{ erg/cm}^2$ for Fe-Pd) for such a variant. *This leads to a reduced number of variants for the martensite phase in thin films.*

Figure 22. Schematic showing reduced number of martensite variants possible in thin film geometry due to magnetostatic considerations.

In addition, the substrate imposes additional mechanical constraints on the films. As a result, the interplay between elastic and magnetic interactions governs the magnetic behavior of these films, and was investigated in detail.

Figure 23(a-b) shows in-plane magnetization loops along different directions for {FeCo (10 nm)/TbFe (7 nm)} exchange spring multilayers with 25 and 50 bilayers, respectively²². As seen from Figure 23(a-b), the as-deposited multilayers show nearly isotropic in-plane magnetization behavior characterized by highly square hysteresis loops and a coercivity of $\sim 45\text{-}60 \text{ Oe}$. Given the highly square magnetization loops in all in-plane directions, the easy axis in Figure 23 is defined by the highest value of remanence, whereas the hard axis is defined by the earliest onset of magnetization rotation and lowest value of remanence. To investigate the in-plane anisotropy of these films, torque magnetometry was performed at different temperatures ranging from 298 K to 10 K, and at fields ranging from 10 Oe to 70,000 Oe. For example, Figure 24 shows the field dependence of torque curves at 298 K for the 25 bilayer film whose magnetization loops are shown in Figure 23(a). Figure 24(a) shows that for fields up to 50 Oe, the Tb-Fe and Fe-Co layers are pinned together. The resulting sine curves for unidirectional anisotropy then simply reflects the Zeeman energy at small fields. (Note that the Tb ions have negative exchange with Fe or Co ions. As a result, the larger moment of the Tb ion dominates within the Tb-Fe layer. In addition, the net moment of

the Tb-Fe layer is anti-parallel or ferrimagnetically aligned with the adjacent Fe-Co layers).

Figure 23. Magnetization loops for TbFe (7 nm)/FeCo (10 nm) multilayers with (a) 25 bilayers and (b) 50 bilayers.

The torque curve in Figure 24(b) is above the exchange field but less than the field required for switching the hard Tb-Fe layer. As a result, the magnetization in the soft Fe-Co layers twist in a spiral with respect to the essentially rigid Tb-Fe layers. Also note the distinct hysteresis of $\sim 90^\circ$ in Figure 24(b) at $\sim 119^\circ$ and 213° , which corresponds to irreversible switching of the Fe-Co layers with respect to the ferrimagnetically coupled Tb-Fe layers at these angles. Figure 3(c) shows that at 500 Oe, the Tb-Fe layer begins to rotate, and Figure 24(d-f) shows switching of both the Tb-Fe and Fe-Co layers at high field (1-10 kOe). When the field strength is further increased to 30 kOe, a distinct

hysteresis again appears in the torque curves, as shown in Figure 24(g-h). This is associated with rotation of the negative exchange coupled Tb moments with respect to the Fe and Co ions within the multilayer. Finally, Figure 24(i) shows that a field of 70 kOe is sufficient to rotate the entire multilayer. Figure 4(a-i) shows the torque curve for the same multilayer at 10 K. Figure 25 shows the torque curves for the same multilayer at 10 K. In contrast to the torque behavior at 298 K in Figure 24, the torque curves in Figure 25 shows that even at fields as high as 1 kOe, the Tb-Fe and Fe-Co layers exhibit unidirectional anisotropy, representing a marked stiffening of exchange between the adjacent layers. A significant hysteresis appears in the torque curves at fields higher than 1 kOe and persists at fields as high as 70 kOe. Also note that the exerted torque at 10 K is more than an order of magnitude higher than that measured at 298 K.

Figure 24. Torque curves as a function of field for TbFe (7 nm)/FeCo (10 nm) multilayers with 25 bilayers at 298 K. See text for explanation.

Figure 25. Torque curves as a function of field for TbFe (7 nm)/FeCo (10 nm) multilayers with 25 bilayers at 10 K. See text for explanation.

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2. “A method to study temperature and stress induced magnetic transitions”, Harsh Deep Chopra and Matthew R. Sullivan, *Review of Scientific Instruments* Vol. **76**, 013910-1 to 013910-6 (2005). (6 pages)
3. “Temperature and field dependent evolution of micromagnetic structure in ferromagnetic shape memory alloys”, Matthew R. Sullivan and Harsh Deep Chopra, *Physical Review B* Vol. **70**, 094427-1 to 094427-8 (2004). (8 pages)
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Invited Talks/Lectures acknowledging grant:

1. "Polymagnets", Department of Materials Science and Engineering, Virginia Tech., Blacksburg, VA, April-14-15, 2005
2. "Magnetic Mosaics in Crystalline Tiles – A New Perspective on Ferromagnetic Shape Memory Alloys - Abstract", **Harsh Deep Chopra**, Session: Active Materials-2, *41st Technical Meeting of the Society of Engineering Science (SES)*, Lincoln, Nebraska, October 11, 2004.
3. "A new perspective on ferromagnetic shape memory alloys - Abstract", **Harsh Deep Chopra**, Session: Materials and Devices for Smart Systems, Boston, *MRS Fall'03 Meeting*, December 5, 2003.
4. "Magnetic mosaics in crystalline tiles – A New Perspective on Ferromagnetic Shape Memory Alloys", **Harsh Deep Chopra**, Symposium on Active Materials, *Society of Industrial and Applied Mathematics Conference on Mathematical Aspects of Materials Science*, Los Angeles, May 25, 2003.

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