Abstract

The phase-field method has been successfully employed during the past ten years to simulate a wide variety of microstructural evolution in materials. Phase-field models describe the microstructure of a material by using a set of field variables whose evolution is governed by thermodynamic functionals and kinetic continuum equations. A phase-field model to describe martensitic and magnetic domain features in ferromagnetic shape memory alloys is presented. The purpose of this model is to serve as a predictive tool to guide ferromagnetic shape memory alloy design. Free energy functionals are based on the phase-field microelasticity and micromagnetic theories; they account for energy contributions from composition, temperature, martensite variant boundaries, elastic strain, applied stress, magnetocrystalline anisotropy, magnetic domain walls, magnetostatic potential, and applied magnetic fields. The time-dependent Ginzburg-Landau and Landau-Lifshitz kinetic continuum equations are employed to track the microstructural and magnetic responses in ferromagnetic shape memory alloys to applied temperature, stress, and/or magnetic fields. The model successfully predicts the expected microstructural responses to these applied fields.

A PHASE-FIELD COMPUTER MODEL OF MICROSTRUCTURE EVOLUTION IN A FERROMAGNETIC SHAPE MEMORY ALLOY

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I. Introduction

The shape memory effect is a special property possessed by certain metallic alloys that allows them to recover their original shape after a large deformation by way of a reversible martensitic phase transformation. The martensitic phase transformation is a diffusionless, cooperative shear-like motion of atoms from a (usually) high symmetry phase to a lower symmetry phase.[1] The shape memory effect is present only in alloys that have a high temperature phase (generally called austenite) that can reversibly and martensitically transform to a low temperature phase (called martensite). The reversibility of the structural change from austenite to martensite allows the martensite, upon heating, to revert back to the austenite in its original orientation, thus recovering the original shape. Because of their unique properties, shape memory alloys can be utilized as "multi-functional" materials in structural, sensing, and actuating applications.

The low symmetry martensite phase can form multiple correspondence variants, i.e. regions of the martensite that have the same crystal structure but different orientations with respect to the austenite. Particular arrangements of these correspondence variants, which are twin related, allow for the phase transformation strain to be self-accommodating, resulting in a low localized strain energy and no net deformation in the material.

A. Ferromagnetic Shape Memory Effect

Large strains can be generated in alloys that are ferromagnetic and exhibit the shape memory effect by the application of a magnetic field. These alloys are called ferromagnetic shape memory alloys. An applied magnetic field is not only an additional means to control the shape memory effect, but is also attractive because it is a much faster method of actuation than the more traditional method of heating and cooling a shape memory alloy.

To be a useful ferromagnetic shape memory alloy, the martensite phase must have high uniaxial magnetocrystalline anisotropy. Magnetocrystalline anisotropy simply means that the magnetic properties of a crystal are dependent on the direction in which they are measured.[2] In Ni_2MnGa, the most widely studied ferromagnetic shape memory alloy, the martensite phase has a tetragonal crystal structure, which contracts along one axis and expands along the other two orthogonal axes when transforming from the cubic austenite phase.[3] The magnetic easy axis, the direction along which the magnetization tends to align, is the contracted axis in the martensite. Recall that the microstructure of the martensite in Ni_2MnGa consists of many variants that have their axes oriented in different directions. The magnetocrystalline anisotropy causes the magnetization vector of each variant to prefer to be aligned with its contracted axis.

When a magnetic field is applied to a crystal in a direction that is different from its easy magnetic axis, the magnetization direction can be rotated to align with the direction of the applied magnetic field. However, the energy required to rotate the magnetization against the anisotropy force is relatively high, and it can be characterized by the magnetic anisotropy constant, K, which is a material-specific constant that describes the strength of its magnetic anisotropy. The applied magnetic field also produces a driving force for variant boundary motion, called the Zeeman energy difference, across the twin boundaries between variants.[4] The Zeeman energy difference creates a pressure on the twin boundaries, and since their mobility in ferromagnetic shape memory alloys is very high, the energy required to move them is lower than the magnetocrystalline anisotropy energy associated with rotating the magnetization vector within a martensite variant. As a result, twin boundaries can move through the material to increase the volume fraction of the favorably oriented martensite variants and decrease the volume fraction of the unfavorably oriented variants, which lowers the total energy of the system.

Figure 1 is a schematic diagram that illustrates this process. The regions between the twin boundaries represent different variants of martensite in a ferromagnetic shape memory alloy. The small arrows represent the direction of magnetization in those martensite variants. When different magnitudes of a magnetic field are applied, the twin boundaries move to increase the volume fraction of the favorably aligned variants, which results in an overall strain of the alloy.

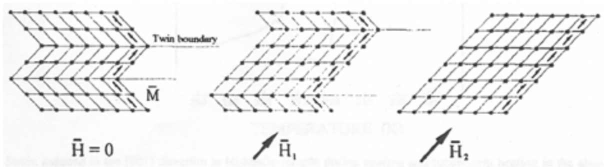


Figure 1: Schematic drawing of the ferromagnetic shape memory effect[5]

B. Phase-Field Microstructural Models

Over the past ten years, a computational method for modeling microstructural evolution, called the phase-field method, has emerged as a useful tool for studying many different microstructural evolution processes. These processes include solidification, solid-state phase transformations, coarsening and grain growth, and dislocation dynamics. For a recent review of phase-field modeling efforts, see a paper by L.Q. Chen.[6]

In conventional approaches to modeling microstructural evolution, the interfaces between regions in a microstructure are considered to be sharp interfaces. As the microstructure changes, the positions of the sharp interface must be tracked. This approach works well for simple one-dimensional systems, but three-dimensional microstructures with complex morphologies render it unfeasible. The phase-field method approximates these interfaces as diffuse interfaces, i.e. the change across them is steep but still continuous. As a result, a continuum equation can be used to describe the microstructural evolution, and the interfaces no longer must be tracked.

A phase-field model describes a microstructure, i.e. the structural or compositional domains contained in it, as well as the interfaces between them, by using a set of field variables. A free energy description of the microstructural evolution, based on the physical mechanisms assumed to be involved in the process, is defined. Then, without any initial assumptions, the field variables are allowed to evolve to an equilibrium state, based on the mathematical free energy functional. Phase-field models assume that all the thermodynamic and kinetic coefficients can be related to microscopic parameters.

C. Research Objective

The objective of this work is to develop a ferromagnetic shape memory alloy phase-field model that can be used as a predictive alloy design tool. Utilizing a phase-field model as a design tool first requires identifying trends in the material properties of previously characterized ferromagnetic shape memory alloys. Then, the trends in material properties can be extrapolated to uncharted alloy compositions, and "virtual experiments" can be conducted to identify favorable compositions that warrant further investigation. The use of a predictive model can save both time and money associated with alloy production and characterization.

II. Description of Ferromagnetic Shape Memory Alloy Phase-Field Model

Phase-field models that describe the martensitic evolution, based on the microelasticity theory developed by Khachaturyan,[7] have been developed by several researchers.[8,9,10,11] Additionally, magnetic domain evolution in materials has been modeled by Kazaryan, et. al.[12] and Jin, et. al.[13] The only previous work to couple these two phenomena to model ferromagnetic shape memory alloys has been done by Koyama and Onodera.[14] However, their work only accounts for the magnetocrystalline anisotropy in the martensite phase, while omitting other magnetic energy contributions that more accurately describe the magnetic structure of the alloy.

The novel aspect of our current work is the coupling of the martensitic and magnetic evolution in ferromagnetic shape memory alloys by utilizing a more comprehensive magnetic energy description than Koyama and Onodera have considered.

A. Martensite Phase-Field Model

The free energy description (called the free energy functional) used to model the martensite transformation is:

$$G = G_{ch} + G_{interface} + G_{elastic} + G_{\sigma app} \qquad (1)$$

where G is the total free energy of the system, G_{ch} is the chemical free energy, $G_{interface}$ is the twin boundary energy between martensite variants, $G_{elastic}$ is the elastic energy generated by the martensite variants, and $G_{\sigma app}$ is the free energy contribution from an applied stress. All of these free energy terms are functions of the field variables that describe the martensite variants.

Equation (1) describes the thermodynamic driving force of martensite evolution, while the time-dependent Ginzburg-Landau equation describes the time evolution of the transformation. The time-dependent Ginzburg-Landau equation is:

$$\frac{\partial \eta_p(r,t)}{\partial t} = -L_{pq} \frac{\delta G}{\delta \eta_q(r,t)} + \xi(r,t) \qquad (2)$$

where η_p are the field variables (one for each martensite variant), r and t are position and time, L_{pq} is a matrix of kinetic coefficients, $\frac{\delta G}{\delta \eta_q(r,t)}$ is the variational derivative of the free energy with respect to the field variables, and $\xi(r,t)$ is a Gaussian noise term. In essence, the time-dependent Ginzburg-Landau equation dictates that the rate of evolution of the field variables is a linear function with respect to the thermodynamic driving forces for the martensitic transformation.

A two-dimensional simulation of the martensite transformation from a cubic (square in 2D) austenite to a tetragonal (rectangle in 2D) martensite in a single crystal is shown in Figure 2 .

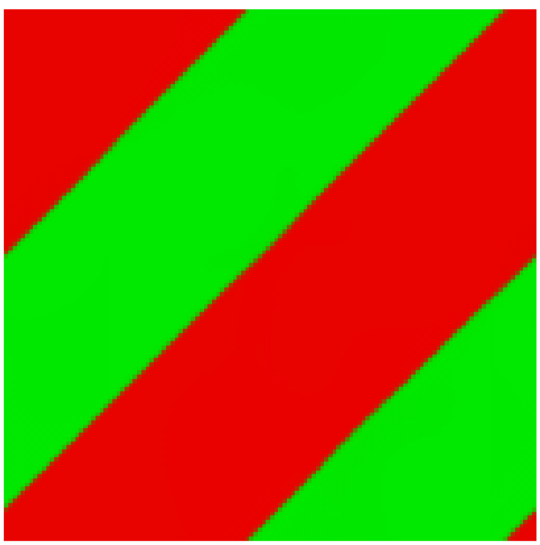


Figure 2: Equilibrium martensite configuration generated with a 2D phase-field model. Red regions are martensite variants with the short axis in the horizontal direction, and green regions have short axis along the vertical direction.

The two martensite variants assume a configuration with 45º twin boundaries and a 1:1 volume fraction ratio to minimize the elastic strain energy associated with the martensitic transformation from the austenite phase.

The martensite phase-field model can also be extended to model the martensitic phase transformation in a polycrystalline material, as shown in Figure 3.

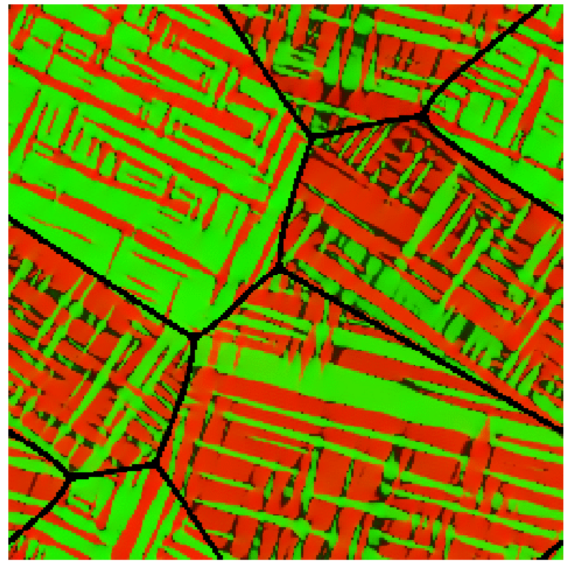


Figure 3: Martensite configuration in a polycrystalline material. Black lines represent crystalline grain boundaries.

In the case of a polycrystal, the equilibrium volume fraction ratio of martensite variants is still near 1:1, but the angles of the twin boundaries vary depending on the orientation of the austenite within in each grain. In the polycrystal, the elastic strain energy is minimized only by the existence of small regions of retained austenite phase, which are the dark areas between the martensite variants.

B. Magnetic Phase-Field Model

The free energy description used to model the magnetic domain evolution in a material is:

$$E = E_{anis} + E_{exch} + E_{ms} + E_{app} \qquad (3)$$

where E is the total energy, E_{anis} is the magnetocrystalline anisotropy energy, E_{exch} is the magnetic exchange energy (a short-range interaction that encourages magnetic moments to be closely aligned with one another), E_{ms} is the magnetostatic energy (a measure of the long range interaction of magnetic moments in the system), and E_{app} is the energy due to an external applied magnetic field.

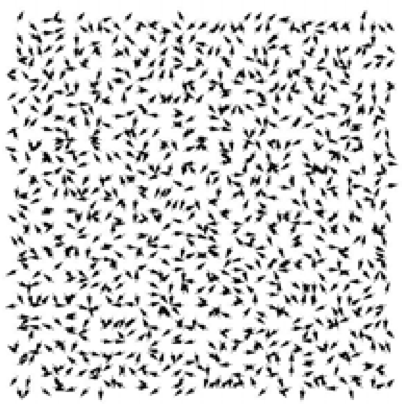
The time evolution of magnetic domain formation is described by the Landau-Lifshitz equation:

$$\frac{\partial M(r,t)}{\partial t} = -\frac{\lambda}{M_s}[M(r) \times (M(r) \times H_{eff}(r))] \qquad (4)$$

where *M(r)* is the magnetization vector, M_s is the saturation magnetization, $H_{eff}(r)$ is the net magnetic field at point *r*, and λ is a positive constant that characterizes the damping of magnetization. The effective field, $H_{eff}(r)$, can be represented as the variational derivative of the total energy of the system with respect to magnetization, $\frac{-\delta E}{\delta M(r)}$.

The initial condition for the magnetic evolution in the austenite is an array of randomly aligned magnetization vectors (Figure 4a). From this starting condition, the model predicts the time evolution of the rotation of magnetization vectors until they reach an equilibrium state (Figure 4b) wherein particular regions (domains) of the material possess magnetization vectors aligned in the same direction. The driving force for magnetic domain formation is the minimization of the magnetostatic energy, i.e. the material tends to minimize magnetic dipole formation within itself, as well as external fields caused by an overall net magnetization.

a)



b)

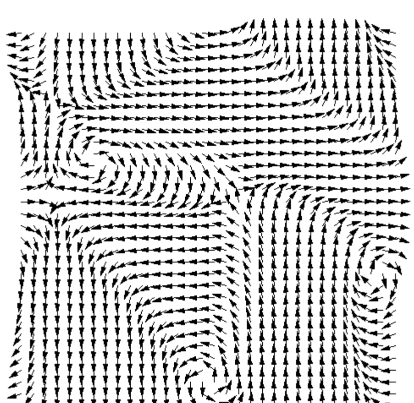


Figure 4: Magnetic domain evolution in austenite; (a) initial configuration, and (b) equilibrium magnetic domain configuration.

C. Coupled Martensitic and Magnetic Domain Evolution

To couple the martensitic structural evolution to the magnetic domain evolution in ferromagnetic shape memory alloys, we have introduced a magnetic anisotropy energy term that contains field variables for both the martensite and the magnetization.

$$E_{ani} = \int K_{aus}(m_1 m_2) + K_{mart}\left[1 - \left(\eta_1{}^2 m_1{}^2 + \eta_2{}^2 m_2{}^2\right)\right] d^2 r \qquad (5)$$

where K_{aus} is the magnetocrystalline anisotropy constant in the austenite, K_{mart} is the magnetocrystalline anisotropy constant in the martensite, m_1 and m_2 are the field variables representing the magnetization vector components in the x- and y-directions, respectively, and η_1 and η_2 are the field variables for the two martensite variants.

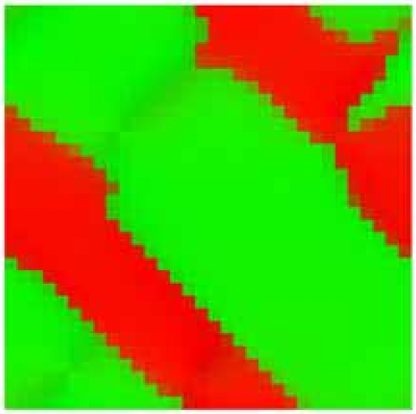
The E_{ani} expression shown above is incorporated into both the martensite free energy functional and the magnetic free energy functional. So, the martensite evolution depends on the magnetic domain structure while, simultaneously, the magnetic domain structure depends on the martensite variant morphology.

III. Coupled Martensitic and Magnetic Evolution in Ni_2MnGa Ferromagnetic Shape Memory Alloy

Material properties for Ni_2MnGa, gathered from various literature sources, were inputted into the ferromagnetic shape memory alloy phase-field model. Simulations were executed to demonstrate the coupled martensitic and magnetic evolution.

Figure 5a shows the martensitic microstructure and Figure 5b shows the corresponding magnetic domain structure in Ni_2MnGa. The twin boundaries between martensite variants no longer assume a 45º orientation, a result of the energy contribution from the magnetocrystalline anisotropy. The magnetic domains that exist within the martensite variants are strongly aligned along the magnetic easy axes, due to the large magnetocrystalline anisotropy constant, K_{mart}, in Ni_2MnGa martensite.

a)



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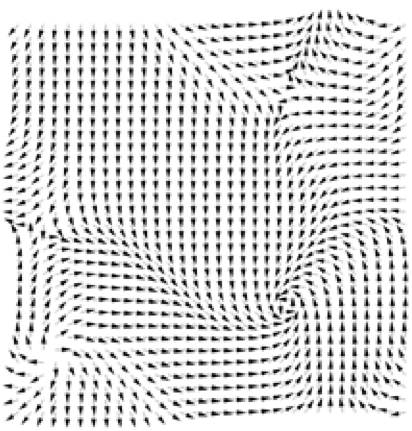


Figure 5: (a) Martensite and (b) magnetic domain structure in Ni_2MnGa ferromagnetic shape memory alloy

The ferromagnetic shape memory effect can be induced by introducing an applied magnetic field to the phase-field model while the material is in the martensitic state. The configuration in Figure 5 is used as the starting condition. With increasing magnitude of the applied magnetic field, the volume fraction of the martensite variant 1 (red) increases, while the volume fraction of martensite variant 2 (green) decreases (see Figure 6). The volume fraction change occurs via twin boundary motion, favoring variant 1 because its magnetic easy axis lies in the same direction as the applied magnetic field. While the volume fraction of variant 1 increases, the magnetization vectors align in the direction of the applied field to minimize the combined magnetic and martensitic energy contributions. Figure 7 shows the martensitic and magnetic domain structures after a reduced magnetic field of H=1.5 is applied in the horizontal direction.

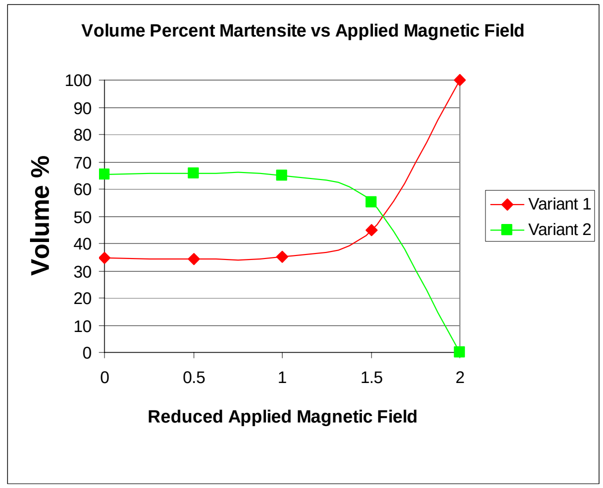
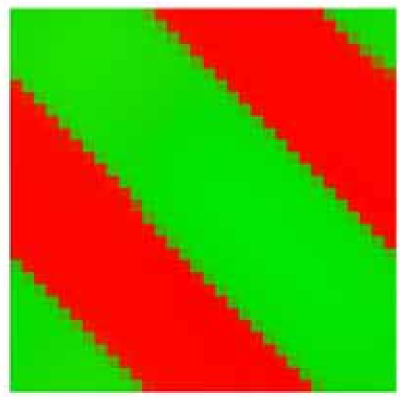


Figure 6: Volume percent martensite vs. reduced applied magnetic field in the horizontal direction

a)



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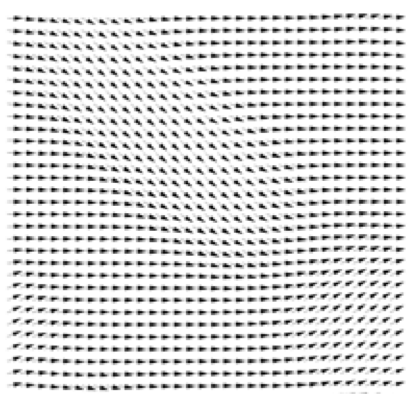

Figure 7: (a) Martensite and (b) magnetic domain structure subjected to an applied magnetic field (H=1.5)

When a reduced applied magnetic field of H=2 is applied, the full transformation to variant 1 is completed, and all of the magnetization vectors align along the easy axis of variant 1.

IV. Conclusions

A two-dimensional phase-field model of coupled martensitic and magnetic domain evolution in a ferromagnetic shape memory alloy has been developed. Using material properties for the ferromagnetic shape memory alloy, Ni_2MnGa, the model results qualitatively demonstrate the expected coupling between martensite variants and magnetic domains, i.e. the ferromagnetic shape memory effect. Extension of the model to three dimensions is needed to make an assessment of the validity of the phase-field model as an alloy design tool.

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