Macroscopic behaviour of magnetic shape-memory polycrystals and polymer composites

May 16, 2006

S. Conti[a*], M. Lenz[b], M. Rumpf[b]

[a] *Fachbereich Mathematik, Universität Duisburg-Essen, Lotharstr. 65, 47057 Duisburg, Germany*
[b] *Institut für Numerische Simulation, Universität Bonn, Nussallee 15, 53115 Bonn, Germany*

Single crystals of magnetic shape-memory materials display a large spontaneous deformation in response to an applied magnetic field. In polycrystalline material samples the effect is frequently inhibited by incompatibilities at grain boundaries. This motivates technological interest in textured polycrystals and composites of single-crystal magnetic shape-memory particles embedded in a soft polymer matrix. We use a continuum model based on elasticity and micromagnetism to study the induced macroscopic material behavior in dependence of such mesoscopic structures via numerical simulation in two dimensions.

Keywords: Magnetic shape-memory, composites, polycrystals, homogenization

1   Introduction

Ferromagnetic shape-memory materials exhibit comparably large strains in response to an applied magnetic field. For single crystals one can achieve strains of order of magnitude 10% [1, 2, 3, 4]. In the search for applicable devices, polycrystal samples have been explored, mainly due to their much easier production. In polycrystals the effectivity drops significantly, as a consequence of the rigidity of interacting grains. The same difficulty is present in shape-memory materials, such as InTl or NiTi. In many cases, the shape-memory effect disappears due to blocking at grain boundaries, this typically being the case when a small number of variants is present. In magnetic shape-memory (MSM) materials the number of variants is typically very low. In particular, NiMnGa has a cubic-to-tetragonal transformation, with only three variants. Furthermore, compatibility conditions for the magnetization at grain boundaries are also relevant. The role of texturing has been, to some extent, investigated experimentally [5, 6], showing that orienting the grains has a strong impact on the macroscopic MSM properties. Here we investigate the role of misalignment numerically and quantify its impact on the effective spontaneous strain.

A recently-proposed alternative for shape memory devices is to embed small single-crystal shape-memory particles in a soft polymer matrix [7, 8]. This approach gives a large freedom in the material development, which includes the type of polymer, the density of particles, their shape, and their orientation [9]. In the case of classical magnetostrictive materials, such as Terfenol-D, experiments have shown that using elongated particles, and orienting them via a bias field during solidification of the polymer, leads to a much larger magnetostriction of the composite [10, 11]. We exemplarily demonstrate the potential of this approach and show that a significant percentage of the single-crystal spontaneous strain can be realized in polymer composites.

2   Micromagnetic-elastic model

We briefly present our model in a geometrically linear setting; for more details and a full nonlinear discussion see [12]. We work on a domain $\Omega \subset \mathbb{R}^2$ occupied by a polycrystal, which consists of grains ω_i, each of which is a single crystal. In the case of polymer composites, the ω_i denote the particles in a polymer matrix $\Omega \setminus \omega$. In both cases, $\omega = \cup \omega_i$.

Kinematics. Let $u : \Omega \to \mathbb{R}^2$ be the elastic displacement, $p : \omega \to \{1, 2\}$ be the phase index, which is supposed to be constant on each grain (each particle, in the case of composites), and $M : \mathbb{R}^2 \to \mathbb{R}^2$ the magnetization. Notice that v and p are defined on the reference configuration, whereas M is defined on the deformed configuration.

Elasticity. We assume the elastic deformation $v(x) = x + u(x)$ to be injective on Ω. We use linear elasticity throughout. The particles have two energy-minimizing phases, which are distinguished by the phase parameter p, and are elastically anisotropic.

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[*]Corresponding author: Sergio Conti, Tel. +49-(0)203-379-2696; Fax +49-(0)203-379-2117; Email sergio.conti@uni-due.de
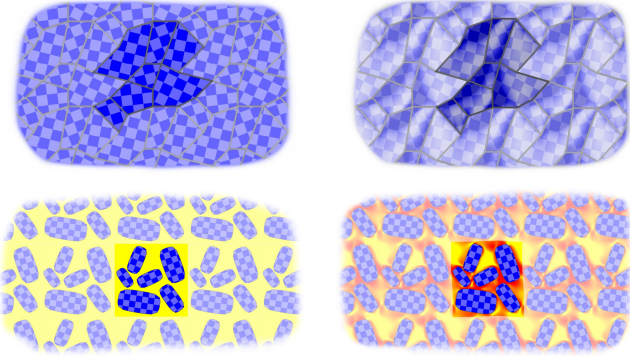
Figure 1: Top line: Reference configuration (left) and deformed configuration (right) for a periodic polycrystal, containing grains with seven different orientations. Bottom line: the same for a composite. For illustrative purposes, a finite portion of a periodic sample is plotted, with the computational cell in the center. The checkerboard pattern in the grains (particles) illustrates the lattice orientation (it is not related to a numerical mesh), and the shading (coloring in the case of polymer composite) shows the elastic energy density.

Their elastic energy is

$$E_{\text{elast}} = \int_\omega W((\nabla v(x))Q(x) - \varepsilon_{p(x)})\, dx\,. \qquad (2.1)$$

Here $Q : \omega \to SO(2)$ represents the crystal lattice orientation in the reference configuration, ε_p is the eigenstrain of phase p, say,

$$\varepsilon_1 = \begin{pmatrix} -\varepsilon_0 & 0 \\ 0 & \varepsilon_0 \end{pmatrix}, \qquad \varepsilon_2 = \begin{pmatrix} \varepsilon_0 & 0 \\ 0 & -\varepsilon_0 \end{pmatrix}, \quad (2.2)$$

and the energy density $W(F)$ penalizes deviations from a preferred, phase-dependent, strain. We parametrize it with the cubic elastic constants C_{11}, C_{12} and C_{44}. Notice that we only linearize the deformation, and that the lattice rotation is inserted explicitly and nonlinearly through $Q(x)$.

In composites we additionally have the elastic energy of the polymer matrix, which is assumed to be an isotropic, linearly elastic material.

Micromagnetism. Let $M = M_s m$ be the magnetization, where M_s denotes the saturation magnetization. The vector field $m : \mathbb{R}^2 \to \mathbb{R}^2$ has unit length on the deformed domain $v(\omega)$, i.e., on the grains (particles), and vanishes elsewhere. The magnetic energy

is given by the coupling to the external field H_{ext}, the demagnetization term, and the phase-dependent anisotropy term:

$$\begin{aligned} E_m &= \int_{\mathbb{R}^2} \frac{1}{2} \frac{M_s^2}{\mu_0} |H_d|^2 - \frac{M_s}{\mu_0} H_{\text{ext}} \cdot m\, dy \\ &\quad + K_u \int_{v(\omega)} \varphi_{p(v^{-1}(y))} \left((R_{\nabla v \circ v^{-1}} Q(y))^T m \right)\, dy\,. \end{aligned}$$

The field $H_d : \mathbb{R}^2 \to \mathbb{R}^2$ is the projection of m onto curl-free fields. Furthermore, $\varphi_2(m) = \frac{m_1^2}{|m|^2}$, $\varphi_1(m) = \frac{m_2^2}{|m|^2}$ are the two magnetic anisotropy functions reflecting the phase-dependent easy axis, and K_u is the uniaxial anisotropy constant. The matrix $R_{\nabla v(x)} \in SO(2)$ is the rotation associated with the elastic deformation v at a reference point x. Explicitly, we consider $R \approx \text{Id} + \frac{1}{2}(\nabla v - (\nabla v)^T)$.

Homogenization and numerical methods. In the spirit of the theory of homogenization [13], we study periodic configurations, where each periodic cell contains a small number of particles, as illustrated in Figure 1. We use boundary elements [14] to express both the elastic and the magnetic problem in the full space in terms of the deformation and the magnetization on the boundary of each grain (particle). Details are given in [12].

Material parameters. We use parameters for NiMnGa, precisely: $\frac{M_s}{\mu_0} \simeq 0.50\frac{\text{MPa}}{\text{T}}$ [15], $K_u \simeq 0.13$ MPa [4], $\varepsilon_0 \sim 0.058$, $C_{11} = 160$ GPa, $C_{44} = 40$ GPa, $C_{11} - C_{12} = 4$ GPa [16] For the polymer we take $\lambda \sim 20$ GPa, $\mu \sim 1$ GPa.

3 Polycrystals

We investigate magnetostriction in a periodic polycrystal. For better comparison, we keep the grain geometry fixed, and vary only the lattice orientation in the grains. This means that the ω_i are fixed, and the Q_i are varied. Figure 2 shows the results for three different choices of the orientation, ranging from almost oriented to completely random. Whereas an orientation mismatch of about $2°$ does not substantially reduce the magnetostriction with respect to a single crystal, already with an average mismatch of $8°$ the effect is reduced by almost 30%. In the unstructured case only one-quarter of the single-crystal spontaneous strain survives.
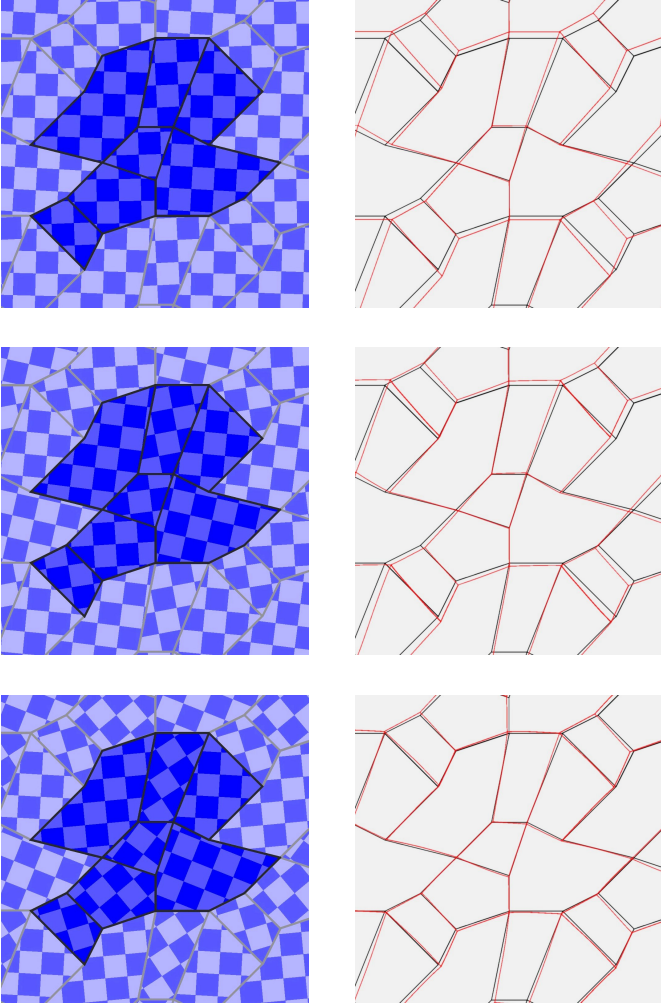
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Figure 2: Spontaneous deformation of a polycrystal with different texturing. Only one unit cell is plotted. With fixed grain geometry, we vary the amount of misorientation. The average orientation mismatch at grain boundaries is 2° (first line) and 8° (second line). The third line depicts a configuration with completely random lattice orientation (i.e. average mismatch close to 22.5°). The first column shows the reference configuration, the second one the deformed state at the spontaneous deformation, with an applied horizontal external field $H_{ext} = 1T$. The spontaneous strain is 5.6%, 4.2%, and 1.6% in the three cases. For comparison, the value for a single crystal is 5.8%.

4 Composites

As a second case we consider polymer composites. Again, we keep the shape and size of the particles
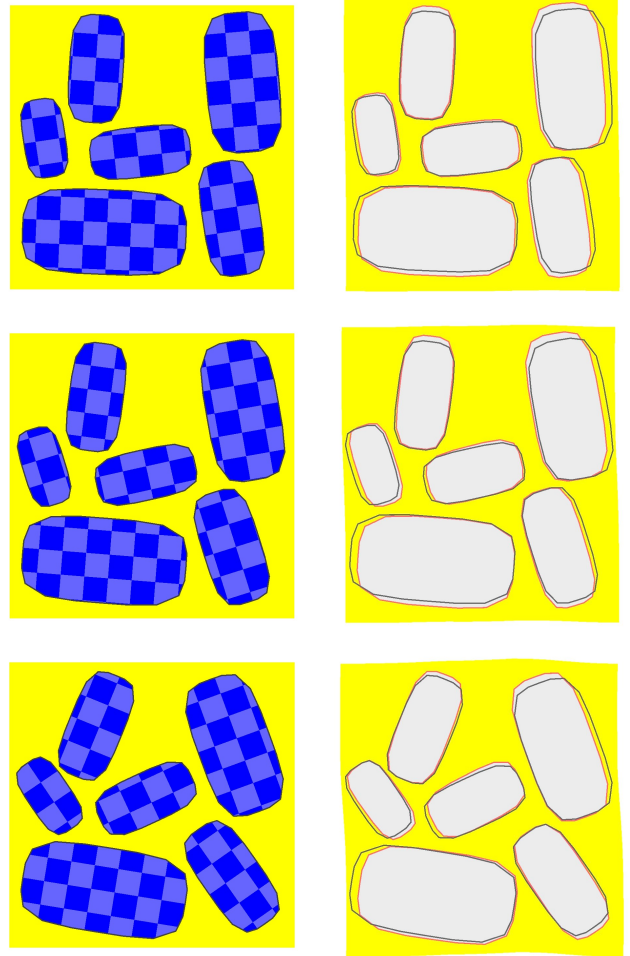
Figure 3: Spontaneous deformation of a composite with different texturing. The volume fraction of the particles is about 50%. Only one unit cell is plotted. With fixed particle geometry, we vary the amount of misorientation between the particles. The average orientation mismatch is 2° (first line), 8° (second line), and completely random (third line). Again, the first column shows the reference configuration, the second one the deformed state at the spontaneous deformation, with an applied horizontal external field $H_{ext} = 1T$. The spontaneous strain is 3.7%, 3.9% and 2.9% in the three cases, compared to the 5.8% in case of a single crystal.

fixed, and vary their orientation (and consequently their spatial arrangement). We assume that the volume fraction of the active material is 50%. Figure 3 shows that the dependence of the macroscopic magnetostriction on the orientation is much smaller

than for polycrystals. In particular, misorientations of 2° and 8° give almost exactly the same magnetostriction, which in turn is comparable to the one of the polycrystal at 8° misorientation. (Indeed, in this range the spontaneous strain also depends significantly on the details of the geometry. For a symmetric arrangement of particles one actually observes monotonicity of the spontaneous strain with respect to the lattice alignment. Here, we refer to Fig. 12 in [12].) Even in the non-oriented case the composite recovers almost one-half of the single-crystal magnetostriction.

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