Theory of magnetostriction with application to TerfenolD

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A mechanism for magnetostriction in the highly magnetostrictive material Terfenol-D is explained in detail. This mechanism is based on a theory of magnetostriction [R. D. James and D. Kinderlehrer, Philos. Mag. B **68**, 237 (1993)] that is particularly suited to predictions of the macroscopic behavior of materials that exhibit large magnetostriction. Some experiments that test these predictions are proposed.

I. INTRODUCTION

A theory of magnetostriction that is adapted to the description of large magnetostriction is given by the authors in Ref. 1. The theory is a micromagnetic in nature and follows the pattern established by Brown.[2] The focus is on an exact specification of the potential wells of the anisotropy energy.

For large specimens such as those encountered in usual actuator applications, the presence of complicated domain structures has hindered the usefulness of micromagnetic theories that seek to incorporate magnetostriction. In recent years new methods of calculating energy minimizers for this situation have emerged, partly as a result of advances in the analysis of the microstructure of martensite and partly as a result of related developments on the design of optimal composites. Using these ideas, the authors[1] found compatible energy minimizing domain structures for the theory specialized to a growth-twinned specimen of $Tb_xDy_{1-x}Fe_2$, $x \approx 0.3$ (Terfenol-D).

The patterns found in Ref. 1 consist of laminates which meet at the growth twin boundary; see, e.g., Fig. 1. Each layer of the laminate has a constant deformation gradient and a substructure of magnetic domains. The motivation for looking at laminates as energy minimizers arose from the observations of Lord *et al.*[3] Analyzing all possible compatible energy-minimizing laminates that meet at the growth twin boundary, we found five distinct energy minimizing microstructures, pictured in Fig. 1, one of which agreed with the photomicrograph of Lord *et al.* Subsequently, all of the other four patterns were observed (Lord[4]), with good agreement of the geometry and even (in the cases available) of the magnetic substructure.

Each of these structures is kinematically compatible and energy minimizing for all values of the volume fraction γ. As γ changes from 0 to 1 the sample experiences a macroscopic strain in the direction $[-211]$, the typical axial direction of a Terfenol-D rod; however, the amount of strain differs for the different laminates. It might then be inferred that, after a few cycles, the rod would be likely to choose the domain structure that yields the maximum magnetostrictive strain, because there is certainly an energy barrier to changing the whole pattern.

A more refined prediction suggests that something different might occur. There are actually two kinds of laminated microstructures predicted by the theory. In the first kind, analogous to "semicoherent" interfaces in the literature on

martensite, the laminates are separated by a transition layer, which of course contains some energy. The value of this energy is proportional to the twin spacing, so it can be reduced as close to zero as desired by refining the twins. The second kind of laminate has no transition layer; it is "exactly compatible." The property of being exactly compatible versus approximately compatible is predicted by the theory; it is not just based on microscopic examination. With exchange energy present, it is clear that exactly compatible laminates are energetically preferred over those which have a transition layer.

Exactly compatible laminates have another interesting feature: The volume fraction γ can change with position along the growth twin boundary, giving them some additional freedom to meet remote conditions.

When we went back and checked the different laminates, we found that the laminate which achieves maximum magnetostrictive strain is not exactly compatible. This suggests that there may possibly be a drastic change in the domain pattern, and as explained herein, this would be favored by compressive stress. These predictions are explained in more detail below. They suggest some interesting experiments, now in progress (Tickle[5]).

These considerations apply to a specimen with parallel growth twins, such as one that is obtained by float-zone processing. Bridgman-grown specimens contain a significant number of grain boundaries and sometimes several different growth twin systems, which surely would rule out simple laminated energy minimizers.

II. THEORY OF TERFENOL-D

We give a brief description of the theory specialized to Terfenol-D. The basic unknown functions of the theory are the deformation $y(x)$ and magnetization $m(y)$. These are minimizers or relative minimizers of the total free energy

$$E(\mathbf{y},\mathbf{m}) = \int_\Omega \varphi\{\nabla \mathbf{y}(\mathbf{x}), \mathbf{m}[\mathbf{y}(\mathbf{x})]\}d\mathbf{x} + \tfrac{1}{2}\int_{R^3}|\nabla u(\mathbf{z})|^2 \, d\mathbf{z}. \tag{1}$$

Here Ω is the reference configuration of the specimen, φ is the anisotropy energy, and the magnetostatic potential u is determined from $\mathbf{m}$ by solving the magnetostatic equation

$$\text{div}(-\nabla u + \mathbf{m}) = 0 \quad \text{on } R^3. \tag{2}$$

In solving this equation it is understood that $\mathbf{m}=0$ outside of $\mathbf{y}(\Omega)$, the deformed configuration. The theory is geometri-

cally exact, to allow it to apply for arbitrarily large deformations. In fact, the elasticity could be linearized, and then the theory would be similar to that of Clark,[6] but it is no more difficult to calculate the energy minimizers for the geometrically exact theory. The energy (1) is appropriate to a single-crystal specimen. Supposing that Ω is divided in two by a growth twin on the plane $x \cdot m_1 = 0$, so that $\Omega = \Omega_1 \cup \Omega_2$ with $\Omega_1 = \Omega \cap (x \cdot m_1 > 0)$ and $\Omega_2 = \Omega \cap (x \cdot m_1 < 0)$, then the appropriate expression for the energy is

$$E_{gt}(\mathbf{y}, \mathbf{m}) = \int_{\Omega_1} \varphi\{\nabla \mathbf{y}(\mathbf{x}), \mathbf{m}[\mathbf{y}(\mathbf{x})]\} d\mathbf{x}$$

$$+ \int_{\Omega_2} \varphi\{\nabla \mathbf{y}(\mathbf{x}) \mathbf{R}_0, \mathbf{m}[\mathbf{y}(\mathbf{x})]\} d\mathbf{x}$$

$$+ \frac{1}{2} \int_{R^3} |\nabla u(\mathbf{z})|^2 \, d\mathbf{z}, \tag{3}$$

where $\mathbf{R}_0$ is a 180° rotation about $\mathbf{m}_1$. Here $\mathbf{m}_1$ is proportional to (111) and expression (3) takes into account the observed crystallography of the type-I growth twins of Terfenol-D. When minimizing Eq. (3), compatibility at the growth twin boundary is automatically taken into account by the assumption that $\mathbf{y}(\mathbf{x})$ is continuous on Ω. Finally, expressions (2) and (3) are appropriate for no applied field or loads.

The key feature about the anisotropy energy φ is its potential-well structure. For Terfenol-D it is assumed to have minima on the set

$$\{\mathbf{RU}_1, \pm \mathbf{Rm}_1\} \cup \{\mathbf{RU}_2, \pm \mathbf{Rm}_2\}$$

$$\{\mathbf{RU}_3, \pm \mathbf{Rm}_3\} \cup (\mathbf{RU}_4, \pm \mathbf{Rm}_4), \tag{4}$$

where $\mathbf{m}_1 = \alpha[111]$, $\mathbf{m}_2 = \alpha[-111]$, $\mathbf{m}_3 = \alpha[1-11]$, $\mathbf{m}_4 = \alpha[11-1]$, $\mathbf{U}_i = \eta_1 \mathbf{1} + (\eta_2 - \eta_1)\hat{\mathbf{m}}_i \otimes \hat{\mathbf{m}}_i$, $\hat{\mathbf{m}}_i = \mathbf{m}_i/|\mathbf{m}_i|$, $i = 1,...,4$, and $\mathbf{R}$ is an arbitrary proper 3×3 rotation matrix. The scalar constant of proportionality α is $1/\sqrt{3}$ of the saturation magnetization, $\eta_2 - 1$ is the saturation strain along [111], and $\eta_1 - 1$ is the saturation strain along $[1-10]$. The notation $\mathbf{a} \otimes \mathbf{b}$ denotes the matrix with components $a_i b_j$ and $\mathbf{1}$ is the identity matrix. Thus, the linear transformation $\mathbf{U}_1$ transforms a cube (aligned with the cubic axes in Ω) by stretching its [111] diagonal with a strain $\eta_2 - 1$ while contracting every direction perpendicular to [111] with a strain $\eta_1 - 1$. This can be seen by applying $\mathbf{U}_1$ to unit vectors in the various directions. We shall say that the specimen is in variant i at a point $\mathbf{x} \in \Omega_1$ if $[\nabla \mathbf{y}(\mathbf{x}), \mathbf{m}(\mathbf{x})]$ has the form $\{\mathbf{QU}_i, \pm \mathbf{Qm}_i\}$ for some rotation matrix $\mathbf{Q}$. A different notation is appropriate for the region Ω_2; that is, because of the presence of $\mathbf{R}_0$ in Eq. (3), the potential wells are modified there. Hence, we say that the specimen is in variant i' at a point $\mathbf{x} \in \Omega_2$ if $[\nabla \mathbf{y}(\mathbf{x}), \mathbf{m}(\mathbf{x})]$ has the form $\{\mathbf{QR}_0 \mathbf{U}_i \mathbf{R}_0, \pm \mathbf{QR}_0 \mathbf{m}_i\}$ for some rotation matrix $\mathbf{Q}$.

III. ENERGY-MINIMIZING COMPATIBLE DOMAIN STRUCTURES

The energy minimizers computed in Ref. 1 consist of laminates above and below the growth twin boundary. All possible compatible laminates were considered; that is, it was assumed that $\nabla \mathbf{y}$ oscillated between two values on Ω_1 and two different values on Ω_2, with possibly a transition layer in between. To be energy minimizing, these values nec-
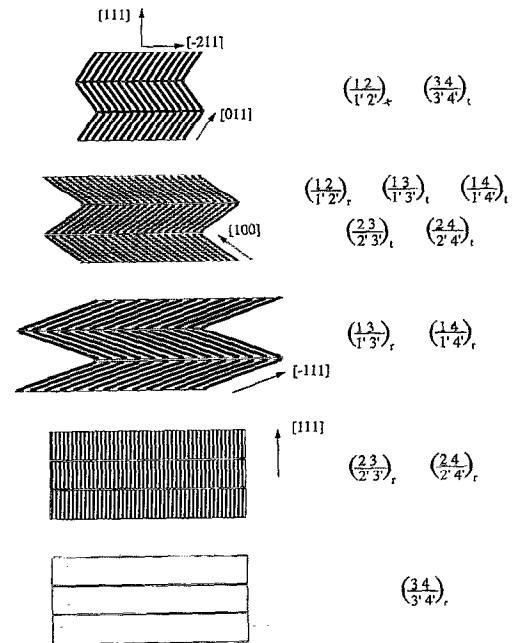

FIG. 1. Appearance of the predicted laminates on the (0-11) plane with light and dark denoting the individual layers within a laminate. The magnetic substructure is not shown.

essarily have to come from the potential wells (4), the energy in the transition layer has to be reducible to zero by refining the layers, and the substructure of magnetic domains has to be arranged to make the field energy arbitrarily small (see Ref. 1 for the details of how this was done).

To summarize the results of these calculations, we use the notation

$$\left(\frac{ij}{k'l'}\right)_{\text{subscript}} \tag{5}$$

to denote a minimizer which uses variants i and j on Ω_1 and k' and l' on Ω_2. The subscript can take the value t ("twin") or r ("reciprocal twin") which refers to which of the two kinematically compatible interfaces ({100} or {110}, respectively) is used in making the laminate; this choice also affects the rotations involved. Using this notation, all compatible variants are shown in Fig. 1. This figure shows only the domains of distortion [as would be revealed by differential interference contrast (DIC) microscopy, for example], not the magnetic substructure. Energy minimization gives one additional restriction: The volume fraction above and below the growth twin boundary is necessarily the same.

As mentioned in Sec. I, some of these laminates are exactly compatible. Exact compatibility is most easily explained by Fig. 2. An approximately compatible laminate has the property that the energy in the transition layer can be reduced to zero by refining the layers. Exchange energy imposes a limit to how much the laminates can be refined, and the ultimate fineness is determined by a compromise between the energy in the transition layer and the total interfacial energy. Exactly compatible laminates have no transition layer and, therefore, can be coarse or fine, and the tendency would be toward coarseness since the only energy present is exchange.
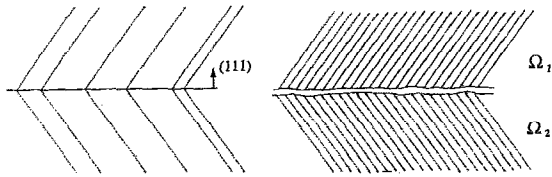
FIG. 2. Exactly compatible configuration (left-hand side) and approximately compatible configuration (right-hand side) laminates.

The exactly compatible laminates are[1]

$$\frac{23}{3'3'} \quad \frac{34}{3'4'} \quad \frac{24}{2'4'}. \tag{6}$$

IV. MECHANISM OF MAGNETOSTRICTION

If we imagine a unit line segment drawn on the specimen in the [−211] direction above the Curie point (i.e., in the reference configuration), this line will become a zig-zag line when evaluated for any of these laminates. It will never "break," as we have assumed kinematic compatibility [i.e., the continuity of $\mathbf{y(x)}$]. For such a line that is long compared to the layer width, the length of the line depends only on the volume fraction γ and the choice of the laminate. A line in the [−211] direction will have the same length whether it is placed above or below the growth twin boundary, by compatibility. It is easy to calculate the maximum and minimum lengths of such lines, as γ goes from 0 to 1, for all the laminates, and the result is given in Table I. (The right-hand-side column is an evaluation of these lengths for material constants η_1 and η_2 appropriate for Terfenol-D, obtained by using surface relief measurements of Al-Jiboory and Lord.[7]) It is seen from Table I that the maximum strain $\Delta l/l$ is obtained using the variants 1 2; however, from Eq. (6) these variants do not achieve exact compatibility. Another look at Table I shows that the reason that the variants 1 2 give the

maximum magnetostrictive strain is that they give both the shortest (0.9992) and the longest (1.001 33) length; but, in fact, the variant pairs 2 3 and 2 4, both of which are exactly compatible, have the same maximal length (1.001 33) as 1 2 but their minimum length is longer.

Computations with applied field and load using the present theory are in progress,[8] but are not yet available; however, as far as the effect of small or moderate loads and fields is concerned, it is likely that they will reveal that compressive stress will favor variants yielding the shortest length, while applied fields in the direction [−211] will favor variants yielding the longest length (This comes from a closer examination of the magnetic subdomain structure.)

Putting these assumptions together with the idea that exactly compatible variants are preferred, we can arrive at a proposal for what might happen. With no compressive stress or applied field, variants 2 3, 2 4, or perhaps 3 4 would be preferred. Still in the absence of stress but with a [−211] field applied, 2 3 and 2 4 are then preferred, even up to large fields, because they give maximum extension and they are exactly compatible; however, upon application of compressive stress, there is clearly a preference for 1 2 (or 1 3 or 1 4, which, however, would not seem to survive cycling the stress, because they achieve only modest maximal lengths).

V. PROPOSED EXPERIMENTS

This proposal suggests some relatively simple experiments. Since the domains of distortion discussed above are observed by making use of surface relief, it is important to polish the specimen in the single-domain state. Failure to do this will result in the possibility of seeing false domain boundaries at places where leveled domain boundaries existed during polishing. Since it is difficult to polish above the Curie temperature, it is reasonable to use stress to do this. According to the results of the theory, a compressive stress favors the single-domain state consisting of variant 1, and a reasonably large compressive stress is advised, to overcome barriers associated with the inevitable defects. A fixture has been built to do this.

With domains of distortion revealed, the experiment consists of observing the effects of applied field and compressive stress on the domain pattern and macroscopic magnetostriction, as indicated in the last paragraph of Sec. IV. The switch from variants 2 3 to 1 2 should be accompanied by a significant change of both the geometry and the fineness.

TABLE I. The minimum and maximum macroscopic lengths of a line oriented along [−211] which in the reference configuration had unit length.

Variants	Length	Length with $\eta_1 = 0.9992$, $\eta_2 = 1.0016$
1 2	min η_1	0.9992
	max$[\eta_1^2 + \frac{8}{9}(\eta_2^2 - \eta_1^2)]^{1/2}$	1.001 33
1 3	min η_1	0.9992
	max$[\eta_1^2 + \frac{2}{9}(\eta_2^2 - \eta_1^2)]^{1/2}$	0.999 73
1 4	min η_1	0.9992
	max$[\eta_1^2 + \frac{2}{9}(\eta_2^2 - \eta_1^2)]^{1/2}$	0.999 73
2 3	min$[\eta_1^2 + \frac{2}{9}(\eta_2^2 - \eta_1^2)]^{1/2}$	0.9997
	max$[\eta_1^2 + \frac{8}{9}(\eta_2^2 - \eta_1^2)]^{1/2}$	1.001 33
3 4	min$\left(\eta_1^2 + \frac{2}{9}(\eta_2^2 - \eta_1^2) - \frac{(\eta_1^2 - \eta_2^2)^2}{3(2\eta_2^2 + \eta_1^2)}\right)^{1/2}$	0.999 47
	max$[\eta_1^2 + \frac{2}{9}(\eta_2^2 - \eta_1^2)]^{1/2}$	0.999 73
(3 4)$_r$	min=max=$[\eta_1^2 + \frac{2}{9}(\eta_2^2 - \eta_1^2)]^{1/2}$	0.999 73
2 4	min$[\eta_1^2 + \frac{2}{9}(\eta_2^2 - \eta_1^2)]^{1/2}$	0.999 73
	max$[\eta_1^2 + \frac{8}{9}(\eta_2^2 - \eta_1^2)]^{1/2}$	1.001 33

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