Enhanced differential magnetostrictive response in annealed Terfenol-D

N. Galloway  
*University of Hull*

M. P. Schulze  
*University of Hull*

R. D. Greenough  
*University of Hull*

David C. Jiles  
*Iowa State University, dcjiles@iastate.edu*

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Abstract
The field and pressure dependencies of the magnetostriction of Tb$_{0.316}$Dy$_{0.684}$Fe$_{1.982}$ have been measured in a grain-oriented rod after thermally annealing for 1 day at 850 °C and for 4 days at 950 °C in an argon atmosphere. The results of the heat treatment are a fivefold increase in the strain coefficient $d_{33} (=d\lambda/dH)$ and a 100% increase in the maximum strain ($\lambda$). There was also an increase in the $\lambda$-vs-$H$ hysteresis. Under compressive uniaxial stress there was virtually no bulk change in magnetostrictive strain until the field exceeded a critical value which depended on the applied stress, for instance ~12 kA/m under a stress of 6 MPa.

Keywords
Annealing, Atmospheric pressure, Heat treatments, Magnetic hysteresis, Magnetostriction

Disciplines
Electromagnetics and Photonics | Engineering Physics

Comments
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N. Galloway, M. P. Schulze, and R. D. Greenough

Department of Applied Physics, University of Hull, Hull HU6 7RX, United Kingdom

D. C. Jiles

Ames Laboratory, Iowa State University, Ames, Iowa 50011

The field and pressure dependencies of the magnetostriction of \( \text{Tb}_{0.316}\text{Dy}_{0.684}\text{Fe}_{1.982} \) have been measured in a grain-oriented rod after thermally annealing for 1 day at 850 °C and for 4 days at 950 °C in an argon atmosphere. The results of the heat treatment are a fivefold increase in the strain coefficient \( d_{33} = \frac{d\lambda}{dH} \) and a 100% increase in the maximum strain (\( \lambda \)). There was also an increase in the \( \lambda \)-vs-\( H \) hysteresis. Under compressive uniaxial stress there was virtually no bulk change in magnetostrictive strain until the field exceeded a critical value which depended on the applied stress, for instance \( \sim 12 \text{ kA/m} \) under a stress of 6 MPa.

In the grain-oriented pseudobinary compound \( \text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.95} \) (also known as Terfenol-D), large magnetostrictions (\( \sim 1600 \text{ ppm} \)) are generated. The magneto-crystalline anisotropy in this material is controlled by the Tb:Dy ratio, and the small negative anisotropy constant \( (K_r \approx -60 \text{ kJ/m}^3) \) results in energy minima along the eight (111) directions, which are therefore the preferred directions for the magnetic moments to orient along. On applying a uniaxial compressive stress along the [112] axis, the free-energy surfaces distort due to the magnetoelastic coupling, generating well-defined local energy minima along the [111] and [111] directions which lie at \( 90^\circ \) to the [112] axis. This causes a redistribution of moments in favor of these two particular directions.

According to the anisotropic rotation model, a magnetic field applied along the [112] axis causes a "jump" of moments from the [111] and [111] directions, towards the [111] direction, which lies only \( 19.5^\circ \) from the [112] axis. This specific moment reorientation causes the burst effect, a sharp increase in strain with applied field \( (d_{33} > 100 \text{ nm/A}) \). The field at which this occurs depends on the magnitude of the compressive stress. This strain burst was not seen in \( \text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.95} \) and the purpose of the present work was to identify any composition or structural factors which regulate the magnetoelastic performance of Terfenol-D, especially the occurrence of the burst effect.

A rod of float-zoned Terfenol-D, 90 mm long and 7.6 mm in diameter, had a targeted composition of \( \text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9} \). After annealing, it was shown to have the composition \( \text{Tb}_{0.316}\text{Dy}_{0.684}\text{Fe}_{1.982} \) by chemical analysis. The specimen was sectioned normal to the cylinder axis and one half was held in an argon atmosphere and heated to 850 °C for 1 day and then at 950 °C for 4 days. After each anneal, a thin oxide layer was removed by grinding so that the final diameter was 7 mm. Magnetization and magnetostriction measurements were made in a 0.6-m-long solenoid which generated fields of up to 140 kA/m. Compressive uniaxial stresses up to 30 MPa were applied using a spring-loaded stressing mechanism. Values of \( d_{33} \) were obtained by differentiating quasi-static \( \lambda-H \) curves. Magnetostriction data were obtained using resistive strain gauges and \( B-H \) curves were measured with an integrating flux magnetometer. The sample was demagnetized before each measurement in order to bring it to a known reference magnetic state, and after measuring the initial magnetization and magnetostriction curves the field was cycled in the range \( -140 \leq H < 140 \text{ kA/m} \).

Figure 1 compares the \( \lambda-H \) curves at 0 and 6 MPa before and after annealing. The main features are that after annealing, the burst effect appears, the maximum strain measured at 100 kA/m increases, and there is an increase in the hysteresis. A maximum fivefold increase in \( d_{33} \) associated with the burst effect is seen with an applied pressure of \( \sim 6 \text{ MPa} \) (Fig. 2). During the initial magnetization in the low-field region, the onset of magnetostriction is delayed until, for instance, a field of \( H \sim 22 \text{ kA/m} \) is applied with a uniaxial compressive pressure of \( \sigma \sim 6 \text{ MPa} \) (Fig. 3). This delay is also seen after field cycling (Fig. 1), but in the former case the strain is virtually zero.

The most immediate explanation for these effects may be a change in anisotropy due to the compositional or structural changes induced during the anneal. However, previous data obtained by Verhoeven et al. show that with the same annealing procedures used in the present study, \( \lambda \) (measured at 2.5 kOe or 200 kA/m), \( \lambda_m \) (the bulk change in strain generated during the burst effect), and \( d\lambda/dH_{\text{max}} \) (the maximum slope in \( \lambda \) vs \( H \)) all increase as the iron:rare earth (RE) ratio increases (Fig. 4). As the (RE)Fe\(_2\) stoichiometric ratio is approached, there is a gradual reduc-
tion in the area of the RE-rich phase near the grain boundaries.

This strong correlation between the enhanced magnetostrictive performance and increase in Fe:RE ratio for iron content in the range 1.90–2.00 has been reported previously, albeit in polycrystalline cast samples. It is concluded that the magnetostrictive properties are regulated primarily by the iron content. As this reaches ~1.982–1.985, the material is nearly single phase and there is then maximum magnetoelastic interaction between the iron and rare-earth ions in the Laves phase structure. The small difference between the iron content and the ideal stoichiometric value of 2 is attributable to an inability to exactly control the loss of rare-earth material during fabrication. The present sample, after annealing, possesses this critical Fe:RE ratio and therefore exhibits an enhanced magnetostrictive performance.

The other notable compositional feature is the high Tb:Dy ratio in the present sample (0.316:0.684 compared with the minimum anisotropy ratio 0.27:0.73 or the target ratio 0.3:0.7), and in all the samples used by Verhoeven in previous work where the Tb fractions were in the range 0.314±0.004. This increases the magnetocrystalline anisotropy compared, for example, with Tb0.27Dy0.73Fe1.95 where the anisotropies of the Tb and Dy are compensated. This makes the energy minima along the (111) axes deeper and hence constraints the magnetization vectors more strongly along these axes.

Prior to the application of a magnetic field, the magnetic moments will be fairly evenly distributed among the (111) axes, but with the application of compressive uniaxial pressure along the grain growth [112] axis, the populations of the [111] and [111] axes are enhanced, which in turn generates the large change in bulk magnetostriction when a magnetic field is applied along the [112] axis. The present sample, after annealing, satisfied the requirement for the burst effect to occur, namely that sufficient magnetocrystalline anisotropy should be present ($K_1 < 0$) prior to the application of a compressive stress along the [112] axis in order to maintain the magnetization vectors along the [111] and [111] axes when a small magnetic field is applied along the [112] axis.

The delay in the appearance of magnetostriction at low fields is due to the applied stress. Although the magnetization can develop via 180° domain processes, the magnetostriction depends on non-180° domain processes which, because of the associated change in strain, require more energy to activate.

The only observation which cannot satisfactorily be explained by the Fe:RE or Tb:Dy ratios is the increase in $\lambda - H$ hysteresis that appears with the burst effect. Data from the present sample show that it is independent of applied pressure. The increase in hysteresis with annealing cannot be associated with domain-wall pinning by the RE-rich phase because this phase is reduced or eliminated by annealing. It seems likely that the hysteresis is related to the energy surfaces in the anisotropic rotation model which are distorted by magnetoelastic energy that accompanies the anisotropic magnetostriction. Further work is in

**FIG. 2.** The field and stress dependence of the magnetostrictive coefficient $d_{33}$ after heat treatment.

**FIG. 3.** Magnetostriction strains as a function of applied field at 0, 6, and 18 MPa developed during the initial magnetization.

**FIG. 4.** Data from Verhoeven (Ref. 6) for the various of $\lambda$, $\lambda_{an}$, $d_{33}$, and hysteresis as a function of iron:rare-earth ratio.
progress to identify the origin of the increase in hysteresis.

The enhanced magnetostriction observed in the present sample after annealing was due primarily to an increase in the iron-to-rare earth ratio, which brought the chemical composition close to the stoichiometric value of (RE)Fe₂.

The appearance of the burst effect, which is the almost discontinuous change in magnetostriction \( \lambda \) with field \( H \), depends on both the iron-to-rare earth ratio and the terbium-to-dysprosium ratio. The stoichiometric composition leads to a homogeneous microstructure, while the terbium-to-dysprosium ratio provides magneto-crystalline anisotropy, which ensures that the magnetic moments of the electrons align along the family of \( \langle 111 \rangle \) axes in the material. When a compressive stress is applied along the \( [112] \) axis, the two axes \( [\overline{1}11] \) and \( [1\overline{1}1] \), which are perpendicular to the stress axis, become energetically favored as a result of the magnetoelastic coupling.

Without the preferential alignment of moments along the \( \langle 111 \rangle \) axes, as a result of magneto-crystalline anisotropy, it would be much more difficult to arrange the reorientation of magnetic moments along the directions perpendicular to the applied field. This could perhaps be achieved by the application of a much higher compressive stress, but in that case the rotation of moments into the field direction would be continuous as a function of magnetic field \( H \). In other words, the rapid, almost discontinuous switching of the moments, known as the burst effect, increases with anisotropy. Consequently, the burst effect has been found to occur in the Tb:Dy=0.3:0.7 composition, which has higher anisotropy, but not in the Tb:Dy=0.27:0.73, low anisotropy composition.

No other mechanism is required to explain the burst effect. It is not a first-order transition, as has been confirmed through an examination of the dependence of \( \lambda \) on magnetization \( M \), which is continuous during even the larger of strain bursts (\( \Delta \lambda \approx 1000 \text{ ppm} \)), with a \( d \) coefficient of 300 nm⁻¹.

The hysteresis which accompanies the burst effect cannot adequately be explained by the presence of a RE-rich phase. Heat treatment used in the present and previous work caused a reduction in the area of the rare-earth-rich phase and any domain-wall pinning by that phase is expected to diminish when stoichiometric \((\text{RE})\text{Fe}_2\) composition is approached. It is desirable that the origins of the hysteresis are identified because, for example, it imposes greater demands on a control system that has to achieve precise and repeatable displacements from an actuator. Since this is likely to be a major application of the material, the reduction of hysteresis in magnetostriction is highly desirable.

In conclusion, the changes in \( d_{113} \) and \( \lambda \) are attributed to an increase in magnetoelastic coupling, which is consistent with a measured change in composition during the anneal. Specifically, there was an increase in the iron-to-rare earth ratio as a result of the anneal. With the application of compressive stress, magnetic moments are redistributed among the \( \langle 111 \rangle \) easy axes, enhancing the population of the \( [\overline{1}11] \) and \( [1\overline{1}1] \) axes prior to the application of the field along the \( [112] \) rod axis that generates the strain burst or Ostenson effect. The delay in the onset of this strain is considered to be due to the extra anisotropy arising from the applied stress.

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\[5\] J. E. Ostenson (private communication).

