AIP Journal of Applied Physics

Dynamic magnetoelastic properties of epoxy-bonded Sm0.88Nd0.12Fe1.93 pseudo-1-3 negative magnetostrictive particulate composite

Fang Yang, Chung Ming Leung, Siu Wing Or, Wei Liu, Zhidong Zhang et al.

Citation: J. Appl. Phys. **111**, 07A940 (2012); doi: 10.1063/1.3679045
View online: http://dx.doi.org/10.1063/1.3679045
View Table of Contents: http://jap.aip.org/resource/1/JAPIAU/v111/i7
Published by the American Institute of Physics.

Dynamic magnetoelastic properties of epoxy-bonded $Sm_{0.88}Nd_{0.12}Fe_{1.93}$ pseudo-1-3 negative magnetostrictive particulate composite

Fang Yang,[1] Chung Ming Leung,[1] Siu Wing Or,[1,a)] Wei Liu,[2] Zhidong Zhang,[2] and Yuan-Feng Duan[3]

[1]*Department of Electrical Engineering, The Hong Kong Polytechnic University, Hung Hom, Kowloon, Hong Kong*

[2]*Shenyang National Laboratory for Materials Science, Institute of Metal Research and International Centre for Materials Physics, Chinese Academy of Sciences, Shenyang 110016, People's Republic of China*

[3]*College of Civil Engineering and Architecture, Zhejiang University, Hangzhou, 310058, People's Republic of China*

(Presented 31 October 2011; received 23 September 2011; accepted 1 December 2011; published online 9 March 2012)

Pseudo-1-3 negative magnetostrictive particulate composite is prepared by embedding and aligning light rare earth (Sm and Nd)-based negative magnetostrictive $Sm_{0.88}Nd_{0.12}Fe_{1.93}$ particles with randomly distributed sizes of 10–180 μm in a passive epoxy matrix using a particle volume fraction of 0.5. The dynamic magnetoelastic properties of the composite are investigated as a function of both magnetic bias field and frequency under a constant magnetic drive field. The dynamic relative permeability (μ_{r33}) exhibits a flat frequency response with no observable dispersion at all bias field levels, except for the fundamental shape resonance range of 40–50 kHz. The free (μ^T_{r33}) and clamped (μ^S_{r33}) relative permeabilities attain their maximum values at low bias field levels of $\leq$10 kA/m because of the relatively easy 180° domain-wall motion. The elastic modulus at constant magnetic field strength (E^H_3) and that at constant magnetic flux density (E^B_3) show a maximum *negative*-ΔE effect, accompanying a maximum dynamic strain coefficient (d_{33}) of -2 nm/A, at about 100 kA/m due to the maximum motion of non-180° domain walls. © *2012 American Institute of Physics*. [doi:10.1063/1.3679045]

Pseudobinary $RR'Fe_2$ Laves alloys (R, R′ ≡ rare earths, where R ≠ R′) have received considerable research interest over the past decade due to their potentially high magnetoelastic properties and technologically important applications in sonar and actuation devices.[1] Among various kinds of $RR'Fe_2$ Laves alloys available today, $Tb_{0.3}Dy_{0.7}Fe_{1.92}$ (Terfenol-D) and $Sm_{0.88}Dy_{0.12}Fe_{1.93}$ (Samfenol-D) have been recognized as key positive and negative magnetostrictive alloys, respectively, because of their giant positive and negative magnetostrictions with low magnetocrystalline anisotropy at room temperature.[2,3] By fabricating polymer-bonded particulate composites, not only the eddy-current losses and mechanical brittleness intrinsic in the alloys can be mitigated, but also the shape novelty, property tailorability, and material cost can be improved.[4–9]

While Terfenol-D, Samfenol-D, and their composites are useful, the inclusion of certain amounts of scanty and expensive heavy rare earths Tb and Dy in these alloys has called for alternatives which are constituted by relatively abundant and cost-effective light rare earths in more recent years. On the side of the positive magnetostrictive alloys, several good light rare earth-substituted candidates have been synthesized by substituting light rare earth Pr or Nd for Tb and/or Dy in the Terfenol-D composition.[10–12] On the side of the negative magnetostrictive alloys, few reports have been made even though $Sm_{0.88}Nd_{0.12}Fe_{1.93}$ alloy has showed promise at room temperature.[13]

We have succeeded in synthesizing $Sm_{1-x}Nd_xFe_{1.55}$ ($0 \leq x \leq 0.56$) Laves alloys,[14] preparing epoxy-bonded $Sm_{1-x}Nd_xFe_{1.55}$ ($0 \leq x \leq 0.56$) pseudo-1–3 composites,[15] and reporting their quasistatic magnetoelastic properties.[14,15] In this paper, we further prepare epoxy-bonded $Sm_{0.88}Nd_{0.12}Fe_{1.93}$ pseudo-1–3 composite with 0.5 particle volume fraction and study its dynamic magnetoelastic properties as a function of both magnetic bias field and frequency under a constant magnetic drive field in order to obtain an improved physical insight into such an emerging magnetostrictive material system for enabling device applications.

Polycrystalline $Sm_{0.88}Nd_{0.12}Fe_{1.93}$ alloy was synthesized by an arc-melting process in a high-purity argon atmosphere.[14] The constituent metals used in the arc-melting process were 99.9% pure Sm, 99.9% pure Nd, and 99.8% pure Fe. An extra 3 wt. % Sm was added to compensate the volatilization of Sm for its high vapor pressure during the arc-melting process. The ingot was sealed in a quartz tube for homogenization at 600 °C for 7 days in a high-purity argon atmosphere. The homogenized ingot was ground into particles with randomly distributed sizes of 10–180 μm. X-ray diffractrometry with CuKα radiation was applied to analyze the phase structure of the particles at room temperature in a Riguku D/max-2500pc diffractrometer equipped with a graphite monochromator. The x-ray diffraction results confirmed the presence of a single cubic Laves phase in the as-prepared alloy.

To fabricate an epoxy-bonded $Sm_{0.88}Nd_{0.12}Fe_{1.93}$ pseudo-1–3 composite, $Sm_{0.88}Nd_{0.12}Fe_{1.93}$ particles and Araldite LY5210/HY2954 epoxy were homogeneously mixed in a paper cup using a particle volume fraction of 0.5. The mixed

a)Author to whom correspondence should be addressed. Electronic mail: eeswor@polyu.edu.hk.

slurry was transferred into a bronze mold with a rectangular cavity of 6×6 mm^2 cross-section and 20 mm length and degassed under vacuum for 30 min to eliminate air bubbles. The mold was sealed and placed between a pair of Nd–Fe–B permanent magnets to experience a uniform magnetic field of about 180 kA/m along the length direction of the mold. This caused the enclosed particles to align with the magnetic flux lines, producing particulate chains similar to pseudo-fiber composites or, in general, a pseudo-1–3 particulate composite.[5,9] With the particles being lengthwise-aligned in the mold, the entire mold-magnet assembly was placed in a temperature-controlled chamber at 80 °C for 10 h to ensure a proper cure of the epoxy for achieving a certainly high degree of cross-linking as well as to impart an average axial residual compressive stress of about 3 MPa to the particulate chains through the thermal shrinkage of the epoxy in the composite.[16] This built-in residual compressive stress was shown to be effective in creating a preferred non-180° domain state in the as-prepared composite although no external stress was used.[5,6] The effect is similar to the case of applying an external prestress to assert an initial non-180° domain state in monolithic Terfenol-D.[1] After demolding, the particle volume fraction of the composite was confirmed to be 0.5 based on Archimedes′ principle and rule-of-mixture formulation for density.[15]

An in-house automated magnetostrictive measurement system was used to measure the dynamic magnetoelastic properties of the composite in the length direction at room temperature and with zero external stress bias.[5,6] The composite to be measured was placed in the middle of a pair of Helmholtz coils, and the whole sample-coil assembly was situated in the pole gap of a C-shaped, water-cooled electromagnet (Myltem PEM-8005 K), to receive the stimulus of an ac magnetic drive field (H_3) and a dc magnetic bias field (H_{Bias}) in the length direction of the composite, respectively. By sweeping a sinusoidal H_3 of amplitude 80 A/m over a prescribed frequency (f) range of 1–70 kHz at a rate of 26 step/s, the corresponding magnetic flux density (B_3) and dynamic strain (S_3) at discrete frequency intervals of 25 Hz/step were measured under various H_{Bias} of 10–240 kA/m. H_3 and H_{Bias} were monitored *in situ* by a pick-up coil and a Hall probe connected to a Gaussmeter (F.W. Bell 5080), respectively. B_3 and S_3 were acquired using a search coil wrapped around the composite and a strain gauge attached to the center of the composite, respectively. All qualities were gathered using a data acquisition unit (Nation Instruments BNC-2110 and NI-PCI6132) under the control of a computer with a Labview program. The f dependence of dynamic relative permeability (μ_{r33}) was determined, together with the H_{Bias} dependence of elastic modules at constant magnetic field strength (E_3^H), elastic module at constant magnetic flux density (E_3^B), and dynamic strain coefficient (d_{33}). Details can be found elsewhere.[5,6,17]

Figure 1 shows the f dependence of μ_{r33} of the $Sm_{0.88}Nd_{0.12}Fe_{1.93}$ composite at various H_{Bias}. It is seen that μ_{r33} is essentially flat over the measured f range of 1–70 kHz with no observable frequency dispersion effect for all H_{Bias} levels of 10–240 kA/m, apart from the fundamental longitudinal-mode resonance-induced variations at about 45 kHz. The observation indicates that the eddy-current losses are insignificant in the composite for f up to 70 kHz.[5] This suggests potential applications of the composite at high frequencies.

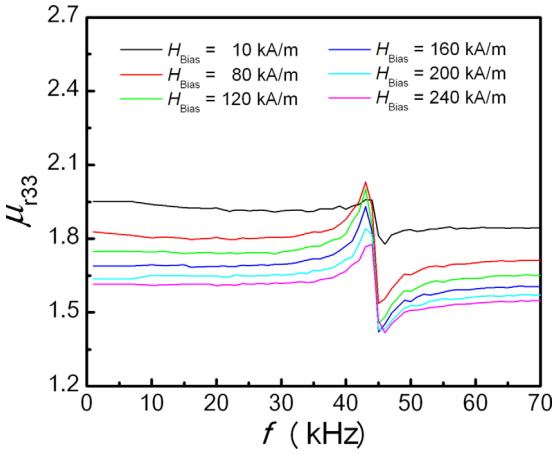

FIG. 1. (Color online) f dependence of μ_{r33}.

Figure 2 plots the dependence of μ_{r33}^T and μ_{r33}^S on H_{Bias} for the $Sm_{0.88}Nd_{0.12}Fe_{1.93}$ composite. μ_{r33}^T and μ_{r33}^S denote the relative permeability at constant stress (i.e., free relative permeability) and that at constant strain (i.e., clamped relative permeability), respectively. The values of μ_{r33}^T and μ_{r33}^S are extracted from the measured μ_{r33} spectrum in Fig. 1 with the corresponding μ_{r33} values at 1 and 70 kHz, respectively. It is found that the two permeabilities attain their highest values at low H_{Bias} levels of $\leq$10 kA/m and their values decrease with an increase in H_{Bias}. In fact, the permeabilities are primarily determined by domain-wall motion at the beginning of the technical magnetization process.[1,5,6,15,17] Accordingly, they are maximized at rather low H_{Bias} levels of $\leq$10 kA/m due to the relatively easy motion of 180° domain walls. As H_{Bias} is increased beyond 10 kA/m, the reduced motion of 180° domain walls competes with the increased motion of non-180° domain walls, giving rise to an obvious decrease in their values.

Figure 3 illustrates the variation of E_3^H and E_3^B on H_{Bias} for the $Sm_{0.88}Nd_{0.12}Fe_{1.93}$ composite. It is clear that both moduli decrease initially with increasing H_{Bias} from 10 to 100 kA/m, leading to a *negative*-ΔE effect with a maximization near $H_{\mathrm{Bias}} = 100$ kA/m. At elevated H_{Bias} levels of 100–240 kA/m, both moduli increase with increasing H_{Bias}. Due to the fact that residual compressive stress has been preset in our

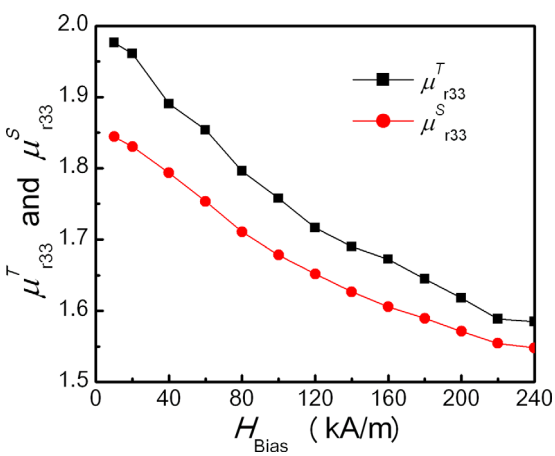

FIG. 2. (Color online) Dependence of μ_{r33}^T and μ_{r33}^S on H_{Bias}.

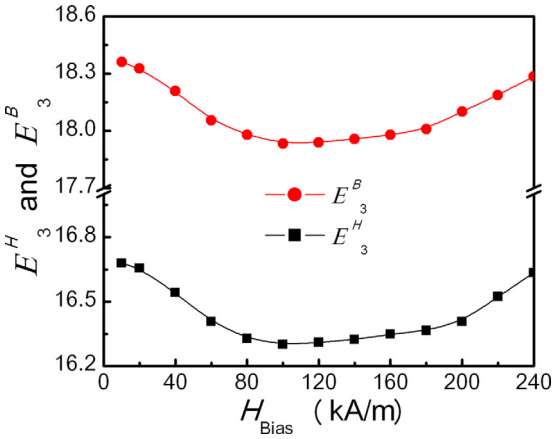

FIG. 3. (Color online) Variation of E_3^H and E_3^B on H_{Bias}.

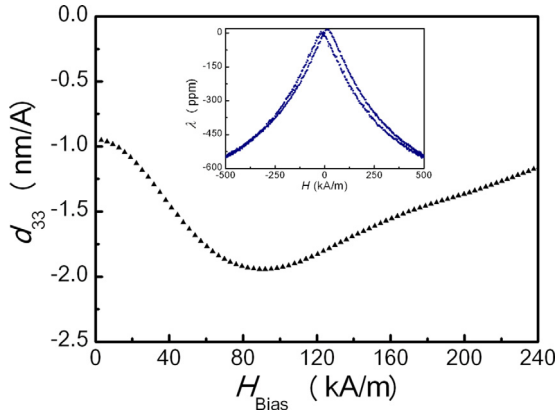

FIG. 4. (Color online) d_{33} at 1 kHz as a function of H_{Bias}. The inset shows the quasistatic λ–H curve measured at 0.05 Hz and with zero external stress bias.

composite through thermal curing of the epoxy phase during composite fabrication, this residual compressive stress essentially exerts on the embedded $Sm_{0.88}Nd_{0.12}Fe_{1.93}$ particles to create a preferred non-180° domain-wall state in the as-prepared composite.[5,6,16] Since non-180° domain-wall motion changes both magnetization (M_3) and strain (S_3) while 180° domain-wall motion changes only M_3,[1,5,6,17] the initial decrease in moduli with increasing H_{Bias} in Fig. 3 is mainly attributed to non-180° domain-wall motion. As H_{Bias} is increased to about 100 kA/m where maximum *negative*-ΔE effect occurs, the compliance associated with the increased deformation contribution from this non-180° domain-wall motion reaches a maximum, resulting in a minimum in stiffness. Above 100 kA/m, the moduli increase with increasing H_{Bias} because constraining of non-180° domain-wall motion by H_{Bias} tends to stiffen the composite.

Figure 4 shows d_{33} at 1 kHz as a function of H_{Bias} for the $Sm_{0.88}Nd_{0.12}Fe_{1.93}$ composite. It is noted that d_{33} carries a negative sign, indicating the presence of a negative magnetostrictive effect (i.e., a magnetic field-induced negative strain) in the composite. The value of d_{33} increases initially up to the maximum of about -2 nm/A near 100 kA/m and then decreases with increasing H_{Bias}. This is a result of increasing and maximizing S_3 contribution from the non-180° domain-wall motion and then domain saturation with limited S_3 contribution from the motion of the available non-180° domain walls. In particular, the presence of maximum d_{33} near $H_{\text{Bias}} = 100$ kA/m is confirmed by the quasistatic magnetostriction–magnetic field (λ–H) curve measured at a low frequency of 0.05 Hz and with zero external stress bias in the inset of Fig. 4. It is obvious that the composite is biased in the center of the "burst region" of the λ–H curve where non-180° domain wall motion is maximum.[1,5,6,17] The results agree with the minimization of the two moduli and the maximization of *negative*-ΔE near 100 kA/m (Fig. 3). It is noted that the d_{33} values reported in Fig. 4 are not necessarily the largest available values of our $Sm_{0.88}Nd_{0.12}Fe_{1.93}$ composite because the d_{33} and λ measurements were done under a mechanically free condition. The use of an additional mechanical stress would preset more initial non-180° domains and thus may produce a larger S_3 output for a given H_3 value, i.e., a larger d_{33}.[1,4,16]

We have fabricated epoxy-bonded $Sm_{0.88}Nd_{0.12}Fe_{1.93}$ pseudo-1–3 negative magnetostrictive composite with 0.5 particle volume fraction and investigated its dynamic magnetoelastic properties as a function of both H_{Bias} and f under an H_3. It has been found that the composite generally possesses insignificant eddy-current losses and its magnetoelastic properties depend greatly on H_{Bias}. The magnetoelastic properties of the composite at low H_{Bias} levels of ≤ 10 kA/m are mainly influenced by the relatively easy 180° domain-wall motion. The maximization of *negative*-ΔE effect and d_{33} near $H_{\text{Bias}} = 100$ kA/m is a result of maximizing non-180° domain-wall motion. The present study provides an improved physical insight into such an emerging light rare earth-based negative magnetostrictive material system for device applications.

This work was supported by the Research Grants Council of the HKSAR Government (PolyU 5266/08E), The Hong Kong Polytechnic University (G-U741), the National Natural Science Foundation of China (50831006, 50571098, and 50908202), and the National Basic Research Program (2010CB934603) of the Ministry of Science and Technology of China.

[1]G. Engdahl, *Handbook of Giant Magnetostrictive Materials* (Academic, San Diego, CA, 2002).
[2]A. E. Clark, AIP Conf. Proc. **18**, 1015 (1974).
[3]R. Abbundi, A. E. Clark, and O. D. McMasters, J. Appl. Phys. **53**, 2664 (1982).
[4]N. Nersessian, S. W. Or, and G. P. Carman, J. Magn. Magn. Mater. **263**, 101 (2003).
[5]S. W. Or, N. Nersessian, and G. P. Carman, IEEE Trans. Magn. **40**, 71 (2004).
[6]S. W. Or, T. L. Li, and H. L. W. Chan, J. Appl. Phys. **97**, 10M308 (2005).
[7]L. Sandlund, M. Fahlander, T. Cedell, A. E. Clark, and J. B. Restorff, J. Appl. Phys. **75**, 5656 (1994).
[8]T. A. Duenas and G. P. Carman, J. Appl. Phys. **87**, 4696 (2000).
[9]T. A. Duenas and G. P. Carman, J. Appl. Phys. **90**, 2433 (2001).
[10]X. K. Lv, S. W. Or, W. Liu, X. H. Liu, and Z. D. Zhang, J. Alloys Compd. **476**, 24 (2009).
[11]W. J. Ren, Z. D. Zhang, X. P. Song, X. G. Zhao, and X. M. Jin, Appl. Phys. Lett. **82**, 2664 (2003).
[12]Y. G. Shi, S. L. Tang, Y. J. Huang, L. Y. Lv, and Y. W. Du, Appl. Phys. Lett. **90**, 142515 (2007).
[13]V. H. Babu, G. Markandeyulu, and A. Subrahmanyam, Appl. Phys. Lett. **90**, 252513 (2007).
[14]F. Yang, W. Liu, X. K. Lv, B. Li, S. Q. Li, J. Li, and Z. D. Zhang, J. Magn. Magn. Mater. **322**, 2095 (2010).
[15]F. Yang, C. M. Leung, S. W. Or, W. Liu, X. K. Lv, and Z. D. Zhang, J. Alloys Compd. **509**, 4954 (2011).
[16]T. A. Duenas, "Giant magnetostrictive composites," Ph.D. thesis, University of California, Los Angeles, 2001.
[17]S. W. Or, N. Nersessian, G. P. McKnight, and G. P. Carman, J. Appl. Phys. **93**, 8510 (2003).