Journal of ELECTRONIC MATERIALS, Vol. 43, No. 2, 2014
DOI: 10.1007/s11664-013-2906-3

Calculation of Hysteresis Losses for Ferroelectric Soft Lead Zirconate Titanate Ceramics

MAHMOUD A. HAMAD[1,2,3]

1.—Physics Department, College of Science, Al Jouf University, P.O. Box 2014, Skaka, Al Jouf, Saudi Arabia. 2.—Physics Department, Faculty of Science, Tanta University, Tanta, Egypt. 3.—e-mail: m_hamad76@yahoo.com

The phenomenological Hamad model was modified to enable retracing of the hysteresis loop of ferroelectric soft lead zirconate titanate (PZT). Comparison with experimental results revealed the modified model can retrace polarization versus electric field for different electric field amplitudes and temperatures. Hysteresis loss per unit volume per cycle for soft PZT was predicted and estimated. The results revealed that energy loss increased with decreasing temperature and with increasing electric field amplitude.

Key words: Modeling, hysteresis, polarization, soft PZT ceramics, hysteresis loss

INTRODUCTION

Ferroelectric materials are used in a broad range of applications including nonvolatile information storage, capacitors, actuators, sensors, energy harvesting, energy storage, atomic force microscopy, ultrasonic flow meters, and cosmic dust detection.[1–4] In recent years, the most successful ferroelectric material in industry has been the ceramic solid solution $Pb(Zr_{1-x}Ti_x)O_3$ (abbreviated "PZT") which has a high piezoelectric coefficient of approximately 250 pC/N for a given modified composition that is advantageous in conversion between electrical energy and mechanical energy.[5] In fact, PZT acts as the basis of most electromechanical devices in multilayer actuators, medical ultrasound, hydrophones, acoustic amplifiers, and ultrasound generators, among other applications.[5] Although hysteresis is inherent in all ferroic materials in current use, the extent and severity of these effects can often be mitigated by restricting power levels, use of appropriate power electronics, or incorporating feedback loops in transducers. In many applications, hysteresis is unavoidable and must be incorporated in models and subsequent control designs to achieve the full capabilities of materials. At a fundamental level, the presence of a hysteresis loop is a basic property characterizing ferroelectric materials. Simulation may aid the design of ferroelectric devices. For example, by simulation, it may be possible to find an optimum pulse for the readout. Observation of hysteresis characteristics will offer clues in a search for ferroelectric materials suitable for memory use if we determine which factors govern the shape of hysteresis loops. Hence, a model which can predict hysteresis curves for ferroelectrics under a variety of conditions is attractive challenge.

Theoretical modeling of complex physical systems may be conducted to improve the precision of empirical description, furnish deeper physical understanding of phenomena, or to enable prediction of physical properties.[6–15] There is growing interest in ferroelectric materials because of their applications.[16–30] In recent years, a number of models of ferroelectric ceramics have been successfully adopted, e.g. Hwang et al.,[31] Pasco and Berry,[32] Yu et al.,[33] the Landau-type model,[34] Zouari et al.,[35] and Butz et al.[36] Among hysteresis models proposed in recent years for representation of nonlinear characteristics, the Hamad model has the advantages of simplicity and ease of implementation for calculation.[37]

The Hamad model is used for magnetic materials in the form of nanocrystalline and nanowire arrays in the paramagnetic state; it is not suitable for some ferroelectric materials, especially at the end of

(Received May 14, 2013; accepted November 6, 2013; published online November 22, 2013)

nonlinear curves, so it must be modified. In this work, a modification of the Hamad model is proposed to make it more suitable for some ferroelectric materials. The proposed modification is based on an empirical approach and experimental observations. The purpose of the work discussed in this paper was to obtain a modification of the Hamad model enabling good simulation of the behavior of soft PZT. Hysteresis loss of soft PZT was predicted by use of the modified model.

ORIGINAL HAMAD MODEL

The modified Hamad model was used to model hysteresis loops for ferroelectric soft PZT. The model equations are summarized briefly below.

In the original Hamad model, hysteresis loops of magnetic nanoscale materials in demagnetization and magnetization processes were modeled as shown below.

(a) In hysteretic demagnetization, the hysteresis loop for a magnetic nanoscale material in the demagnetization process is expressed as follows:

$$M = m \cdot \left(K \cdot \log_{\mathrm{e}} \left| \frac{H + H_{\mathrm{c}}}{H_{\mathrm{c}}} \right| + M_{\mathrm{r}} \right), \quad (1)$$

where $m = \begin{cases} 1 & \text{when } H > -H_{\mathrm{c}}, \\ 0 & \text{when } H = -H_{\mathrm{c}}, \\ -1 & \text{when } H < -H_{\mathrm{c}}. \end{cases}$

$$K = \frac{M_{\mathrm{s}} - M_{\mathrm{r}}}{\log_{\mathrm{e}} \left| \frac{H_{\mathrm{s}} + H_{\mathrm{c}}}{H_{\mathrm{c}}} \right|},$$

M, H, H_c, H_{s}, M_{s}, and M_{r} are magnetization, applied magnetic field, coercivity, amplitude of applied magnetic field, saturation, and remanence, respectively.

(b) In hysteretic magnetization, the hysteretic loop for a magnetic nanoscale material in the magnetization process is expressed as follows:

$$M = n \cdot \left(K \cdot \log_{\mathrm{e}} \left| \frac{H - H_{\mathrm{c}}}{H_{\mathrm{c}}} \right| + M_{\mathrm{r}} \right), \quad (2)$$

where $n = \begin{cases} 1 & \text{when } H > H_{\mathrm{c}}, \\ 0 & \text{when } H = H_{\mathrm{c}}, \\ -1 & \text{when } H < H_{\mathrm{c}}. \end{cases}$

MODIFICATION OF THE HAMAD MODEL

To characterize the characteristics of hysteresis for output polarization and an applied voltage, a modification of the phenomenological Hamad model is proposed in this section. To retrace the polarization of soft PZT in satisfactory agreement with experimental results, it is possible to use the following modification. In this model, four variables must be identified: coercivity E_{c}, remnant polarization P_{r}, electric field amplitude E_{s}, and saturation P_{s}. After tests on modification of the model, an additional term for both the polarization equation and the depolarization equation is proposed, as described below:

1. In a hysteretic depolarization process, polarization P is related to field strength E by the formula:

$$P = m \cdot \left[\mathrm{K}\ \mathrm{Log}_{\mathrm{e}} \left| \frac{E + E_{\mathrm{c}}}{E_{\mathrm{c}}} \right| + P_{\mathrm{r}} + A \cdot e^{B \cdot (E + E_{\mathrm{s}})} \right]. \quad (3)$$

where P_{r} is the remnant polarization, E_{c} is the coercive field, and$K = \frac{(P_{\mathrm{s}} - P_{\mathrm{r}})}{\mathrm{Log}_{\mathrm{e}} \left| \frac{E_{\mathrm{s}} - E_{\mathrm{c}}}{E_{\mathrm{c}}} \right|}$.

$n = \pm 1$ or 0 (such that $n = 1$ when $E > -E_{\mathrm{c}}$, $n = -1$ when $E < -E_{\mathrm{c}}$, or $n = 0$ when $E = -E_{\mathrm{c}}$). E_{s} is amplitude of field strength. A and B are terms which can be obtained by curve fitting between the model and experimental data.

2. In a hysteretic polarization process, polarization P is related to field strength E by the formula:

$$P = n \cdot \left[\mathrm{K}\ \mathrm{Log}_{\mathrm{e}} \left| \frac{E - E_{\mathrm{c}}}{E_{\mathrm{c}}} \right| + P_{\mathrm{r}} + A \cdot e^{B \cdot (-E + E_{\mathrm{s}})} \right], \quad (4)$$

where $n = \pm 1$ or 0 (such that $n = 1$ when $E > E_{\mathrm{c}}$, $n = -1$ when $E < E_{\mathrm{c}}$, or $n = 0$ when $E = E_{\mathrm{c}}$). With this modification, it will be observed that simulation by use of this modified model furnishes data similar to experimental data, especially for the ends of hysteretic loop, compensating the deficiency of the original model when applied to a ferroelectric soft PZT material.

PREDICTION OF HYSTERESIS LOSS

Because of the time-varying electromagnetic fields in electrical machines, hysteresis loss accounts for an important proportion of total losses.[38] Because of the complex properties of ferroelectric material, engineers depend heavily on numerical simulation tools to achieve more efficient designs. The main benefits of this model are low computational cost and time whereas traditional models require more computational effort. Information about such important macroscopic properties as saturation polarization, coercivity, polarization at the remanence point, and loss density obtained by numerical integration of loop area may be acquired from polarization curves. Hysteresis loss per unit volume each cycle can be predicted from the area of a recorded hysteresis loop by use of the equation:

$$\text{Hysteresis loss} = \oint P \mathrm{d}E \quad (5)$$

where $\oint P\mathrm{d}E$is the area of the hysteresis loop. In this section, a simple method is established for predicting the energy loss in ferroelectric material undergoing cyclic polarization as shown in Fig. 1.

$$\oint P\mathrm{d}E = \text{Area of region ABCD},$$
$$= 2 \times \text{Area of region ACD},$$
$$= 2 \times [\text{Area of region ADF} - \text{Area of region CDF}],$$
$$= 2 \times \left[\int_{-E_c}^{E_s} P\mathrm{d}E - \int_{E_c}^{E_s} P\mathrm{d}E \right],$$
$$= 2 \times \int_{-E_c}^{E_s} m \cdot \left(\mathrm{K}\log_e \left| \frac{E + E_c}{E_c} \right| + P_r + A \cdot e^{\mathrm{B}\cdot(E+E_s)} \right) \mathrm{d}E,$$
$$- 2 \times \int_{E_c}^{E_s} n \cdot \left(\mathrm{K}\log_e \left| \frac{E - E_c}{E_c} \right| + P_r + A \cdot e^{\mathrm{B}\cdot(-E+E_s)} \right) \mathrm{d}E,$$
$$= 2 \times \left[K \cdot E_s \cdot \log_e \left| \frac{E_s + E_c}{E_s - E_c} \right| + 2E_c(P_r - K) + \frac{A}{B}\left(e^{2\mathrm{B}\cdot E_s} - 2 \cdot e^{\mathrm{B}\cdot(-E_c+E_s)} + 1\right) \right]. \quad (6)$$

Today, because of developments in materials science, Eq. 6 can be used by instrument designers to estimate hysteresis energy loss. It can, moreover, easily be used to estimate power loss of the AC magnetic field of biomaterials in the study of hyperthermia.

MODIFIED MODEL VERIFICATION

To illustrate and validate the modification of the Hamad model, its results were compared with some significant experimental results. The soft PZT ceramic is taken as an example. Its properties and hysteretic loss under different conditions are listed in Tables I and II.

To verify the suggested model, experimental and modeling data were compared using the data from Ref. 39. Figure 2 shows experimental data for soft PZT ceramic at T = 373 K with varying electric field

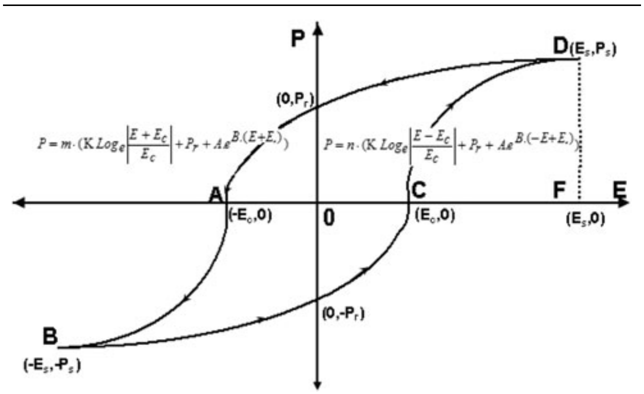

Fig. 1. Hysteresis loop: polarization P as function of ferroelectric field strength E.

Table I. Terms of the modified Hamad model for hysteresis loops of soft PZT ceramic at T = 373 K with varying electric field amplitude E_s

E_c (kV/cm)	E_s (kV/cm)	P_r (μC/cm^2)	P_s (μC/cm^2)	K (μC/cm^2)	A (μC/cm^2)	B cm/kV)	Hysteresis loss (kJ/m^3)
8.40	40	26.24	34.54	4.74	1.83	−0.09	848
8.17	35	25.28	33.35	4.85	3.34	−0.09	768
8.10	30	25.21	32.52	4.72	3.10	−0.09	771
7.64	25	23.65	30.39	4.64	3.75	−0.06	684
7.48	20	23.65	29.50	4.50	3.34	−0.09	686

Table II. Terms of the modified Hamad model for hysteresis loops of soft PZT ceramic at E_s = 40 kV/cm with varying temperature

Temperature (K)	E_c (kV/cm)	E_s (kV/cm)	P_r (μC/cm^2)	P_s (μC/cm^2)	K (μC/cm^2)	A (μC/cm^2)	B (cm/kV)	Hysteresis loss (kJ/m^3)
453	5.13	40	20.17	30.43	4.72	1.20	−0.09	390
413	6.8	40	22.75	32.47	5.04	1.73	−0.22	604
373	8.25	40	25.27	34.23	5.07	2.12	−0.09	795
333	9.3	40	29.43	37.26	4.69	1.60	−0.09	1066
293	10.76	40	33.71	39.75	3.89	1.90	−0.01	1471

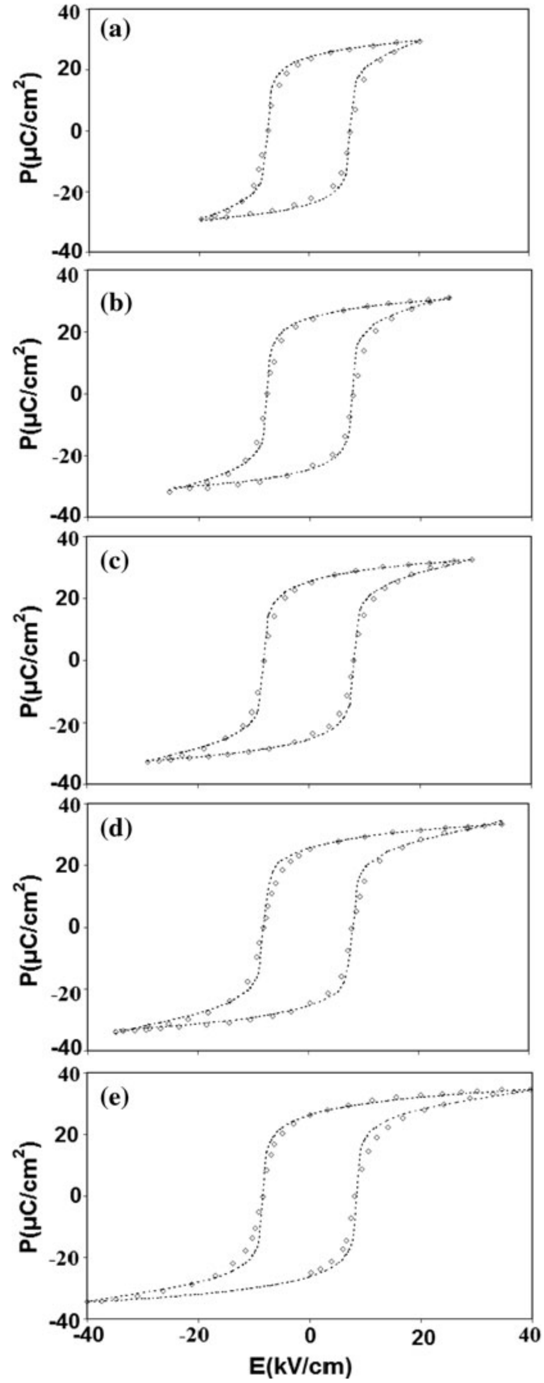

Fig. 2. Hysteresis loops of soft PZT ceramic at T = 373 K with varying electric field amplitude simulated by use of the modified Hamad model (*dashed lines*) compared with experimental results from Ref. 39 (*hollow symbols*).

amplitude and data simulated by use of the modified Hamad model. Figure 3 shows hysteresis loops for soft PZT ceramic at E_s = 40 kV/cm with varying temperature and data simulated by use of the modified Hamad model. It is clear that a good fit to the experimental data is observed, and that the

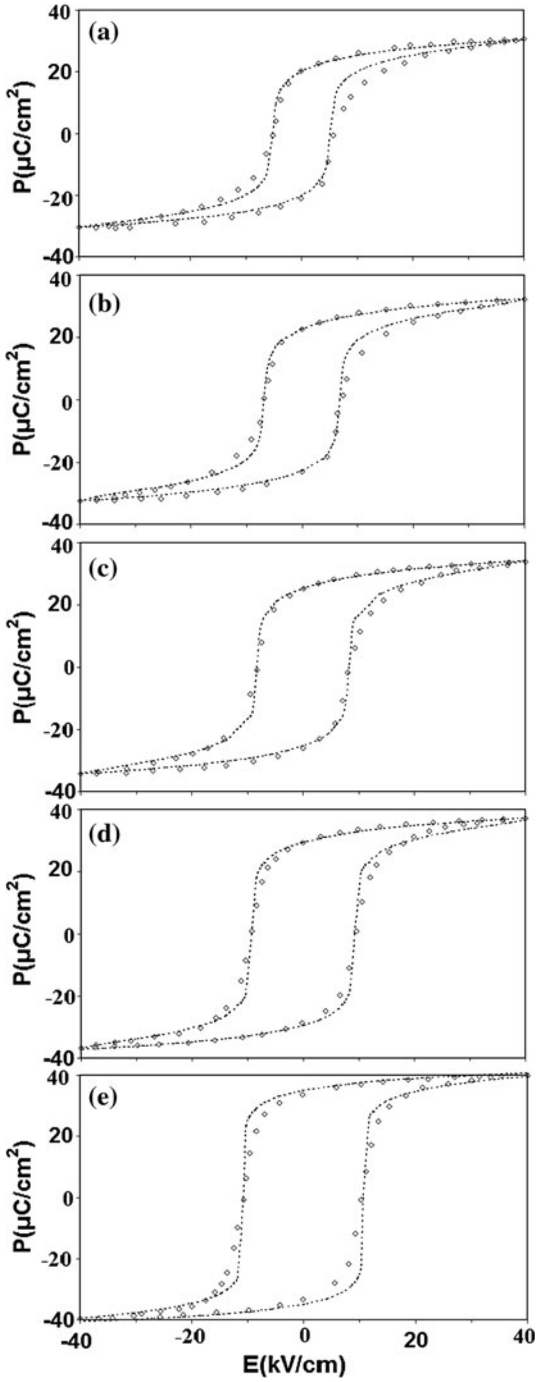

Fig. 3. Hysteresis loops of soft PZT ceramic at E_s = 40 kV/cm with varying temperature simulated by use of the modified Hamad model (*dashed lines*) compared with experimental results from Ref. 39 (*hollow symbols*).

modified model is a powerful tool for study of a wide range of ferroelectric materials.

From Table I it is apparent that hysteresis loss for soft PZT ceramic increases with increasing electric field amplitude, as a result of increasing maximum polarization and coercivity. This is generally attributed to an increase in the magnitude of polarization within a domain, a change of the direction of polarization within a domain, and a change of the relative volume of different domains as a result of displacement of domain boundaries. From Table II it is apparent that hysteresis loss per unit volume per cycle for soft PZT ceramic decreases with increasing temperature. This is because, as temperature increases, the maximum polarization decreases and the remnant polarization decreases, because fewer domain walls are available for pinning. Moreover, there is an increase in exchange interactions with increasing temperature.

It can be stated that the modified Hamad model has several advantages over other models based on free energy:

(i) few variables are required;
(ii) the behavior of the materials in the regions of interest is measured;
(iii) the procedure used does require additional computational effort for the numerical simulation;
(iv) the processing time required for simulation of hysteresis is limited;
(v) it is suitable for coupling with field computation;
(vi) it is compatible with available circuit simulation software; and
(vii) the modified model is suitable for prediction of performance for an instrument designer.

CONCLUSION

In the work discussed in this paper the Hamad model was modified to enable capture of the hysteresis behavior of soft PZT ceramic. The modified model can be used to predict hysteresis loss. Hysteresis loss per unit volume per cycle for soft PZT ceramic was predicted and estimated experimentally. Predicted and experimental results both showed that hysteresis loss per unit volume per cycle increased with decreasing temperature and increasing electric field amplitude. The modified model is suitable for analysis of electrical machines and is suitable for computer-aided design of electromagnetic devices. Moreover, the modified model is useful for feed forward and signal control in unipolar drive fields without expensive sensors such as capacitive position sensors or laser vibrometers.

REFERENCES

1. K. Jayabal, D. Srikrishna, T.S. Abhinandan, A. Arockiarajan, and S.M. Srinivasan, *Int. J. Eng. Sci.* 47, 1014 (2009).
2. B. Marinkovic, T. Kaya, and H. Koser, *J. Appl. Phys.* 109, 014904 (2011).
3. S. Takechi, T. Onishi, S. Minami, T. Miyachi, M. Fujii, N. Hasebe, K. Mori, K. Nogami, H. Ohashi, S. Sasaki, H. Shibata, T. Iwai, E. Grün, R. Srama, and N. Okada, *Adv. Space Res.* 43, 455 (2009).
4. S. Takechi, T. Miyachi, M. Fujii, N. Hasebe, K. Nogami, H. Ohashi, S. Sasaki, H. Shibata, T. Iwai, E. Grün, R. Srama, and N. Okada, *J. Atmos. Solar Terr. Phys.* 71, 191 (2009).
5. W. Duan and Z.R. Liu, *Curr. Opin. Solid State Mater. Sci.* 10, 40 (2006).
6. M.A. Hamad, *J. Supercond. Nov. Magn.* (2013). doi: 10.1007/s10948-013-2189-1.

7. M.A. Hamad, *J. Supercond. Nov. Magn.* 26, 449 (2013).
8. M.A. Hamad, *J. Therm. Anal. Calorim.* 111, 1251 (2013).
9. M.A. Hamad, *J. Supercond. Nov. Magn.* doi:10.1007/s10948-013-2244-y.
10. M.A. Hamad, *J. Adv. Ceram.* 2, 213 (2013).
11. M.A. Hamad, *J. Supercond. Nov. Magn.* (2013). doi:10.1007/s10948-013-2270-9.
12. M.A. Hamad, *J. Therm. Anal. Calorim.* (2013). doi:10.1007/s10973-013-3362-2.
13. M.A. Hamad, *J. Adv. Ceram.* 1, 290 (2012).
14. M.A. Hamad, *J. Therm. Anal. Calorim.* 113, 609 (2013).
15. M.A. Hamad, *J. Supercond. Nov. Magn.* (2013). doi:10.1007/s10948-013-2124-5.
16. M.A. Hamad, *Appl. Phys. Lett.* 102, 142908 (2013).
17. M.A. Hamad, *AIP Adv.* 3, 032115 (2013).
18. M.A. Hamad, *Philos. Magn. Lett.* 93, 346 (2013).
19. M.A. Hamad, *Phase Transit.* 86, 307 (2013).
20. M.A. Hamad, *J. Adv. Ceram.* (2013). doi:10.1007/s40145-013-0076-7.
21. M.A. Hamad, *J. Adv. Dielectr.* 3, 1350008 (2013).
22. A. Erturk, J. Hoffmann, and D.J. Inman, *Appl. Phys. Lett.* 94, 254102 (2009).
23. Y. Lin and H.A. Sodano, *J. Appl. Phys.* 106, 114108 (2009).
24. J. Sigman, G.L. Brennecka, P.G. Clem, and B.A. Tuttle, *J. Am. Ceram. Soc.* 91, 1851 (2008).
25. G.L. Brennecka and B.A. Tuttle, *J. Mater. Res.* 22, 2868 (2007).
26. T.P.C. Juan, C.L. Lin, W.-C. Shih, C.C. Yang, J.Y.M. Lee, D.C. Shye, and J.H. Lu, *J. Appl. Phys.* 105, 061625 (2009).
27. N. Satoh, K. Kobayashi, S. Watanabe, T. Fujii, T. Horiuchi, H. Yamada, and K. Matsushige, *Jpn. J. Appl. Phys.* 42, 4878 (2003).
28. M.A. Hamad, *Int. J. Thermophys.* 34, 1158 (2013).
29. S. Takechi, T. Miyachi, M. Fujii, N. Hasebe, K. Nogami, H. Ohashi, S. Sasaki, H. Shibata, T. Iwai, E. Grün, R. Srama, and N. Okada, *J. Atmos. Solar Terr. Phys.* 71, 191 (2009).
30. M.A. Hamad, *J. Comput. Electron.* 11, 344 (2012).
31. S. Hwang, C. Lynch, and R. McMeeking, *Acta Metall. Mater.* 43, 2073 (1995).
32. Y. Pasco and A. Berry, *J. Intell. Mater. Syst. Struct.* 15, 375 (2004).
33. Y. Yu, N. Naganathan, and R. Dukkipati, *Mech. Mach.* 37, 49 (2002).
34. Y.L. Wang, X.Y. Wang, L.Z. Chu, Z.C. Deng, W.H. Liang, B.T. Liu, G.S. Fu, N. Wongdamnern, T. Sareein, and R. Yimnirun, *Phys. Lett. A* 373, 4282 (2009).
35. W. Zouari, T. Ben Zineb, and T. Benjeddou, *Int. J. Solids Struct.* 48, 87 (2011).
36. A. Butz, S. Klinkel, and W. Wagner, *Proceedings Third MIT Conference on Computational Fluid and Solid Mechanics*, ed. K. Bathe (Cambridge: Massachusetts Institute of Technology, 2005) pp. 291–296.
37. M.A. Hamad, *Jpn. J. Appl. Phys.* 49, 085004 (2010).
38. H. Mamiya and B. Jeyadevan, *Magn. Magn. Mater.* 323, 1417 (2011).
39. R. Yimnirun, R. Wongmaneerung, S. Wongsaenmai, A. Ngamjarurojana, S. Ananta, and Y. Laosiritaworn, *Appl. Phys. Lett.* 90, 112906 (2007).