AIP Journal of Applied Physics

Domain wall release in “hard” piezoelectric under continuous large amplitude ac excitation

Yongkang Gao, Kenji Uchino, and Dwight Viehland

Citation: Journal of Applied Physics **101**, 114110 (2007); doi: 10.1063/1.2745437
View online: http://dx.doi.org/10.1063/1.2745437
View Table of Contents: http://scitation.aip.org/content/aip/journal/jap/101/11?ver=pdfcov
Published by the AIP Publishing

Articles you may be interested in

Effective piezoelectric response of twin walls in ferroelectrics
J. Appl. Phys. **113**, 187222 (2013); 10.1063/1.4801988

De-aging of Fe-doped lead-zirconate-titanate ceramics by electric field cycling: 180°- vs. non-180° domain wall processes
J. Appl. Phys. **112**, 034103 (2012); 10.1063/1.4739721

Evidence of temperature dependent domain wall dynamics in hard lead zirconate titanate piezoceramics
J. Appl. Phys. **112**, 014113 (2012); 10.1063/1.4736582

Evaluation of domain wall motion in bipolar fatigued lead-zirconate-titanate: A study on reversible and irreversible contributions
J. Appl. Phys. **107**, 104119 (2010); 10.1063/1.3386461

Compression of piezoelectric ceramic at constant electric field: Energy absorption through non-180° domain-wall motion
J. Appl. Phys. **92**, 1504 (2002); 10.1063/1.1489498

MIT LINCOLN LABORATORY CAREERS

Discover the satisfaction of innovation and service to the nation

- Space Control
- Air & Missile Defense
- Communications Systems & Cyber Security
- Intelligence, Surveillance and Reconnaissance Systems
- Advanced Electronics
- Tactical Systems
- Homeland Protection
- Air Traffic Control

LINCOLN LABORATORY
MASSACHUSETTS INSTITUTE OF TECHNOLOGY

LEARN MORE

[This article is copyrighted as indicated in the article. Reuse of AIP content is subject to the terms at: http://scitation.aip.org/termsconditions. Downloaded to] IP: 128.173.126.47 On: Thu, 07 May 2015 17:50:27

JOURNAL OF APPLIED PHYSICS **101**, 114110 (2007)

Domain wall release in "hard" piezoelectric under continuous large amplitude ac excitation

Yongkang Gao[a)]
Hitchiner Manufacturing, Inc., 127 Old Wilton Road, Milford, New Hampshire 03055

Kenji Uchino
International Center for Actuators and Transducers, Materials Research Laboratory, Pennsylvania State University, University Park, Pennsylvania 16802

Dwight Viehland
Department of Materials Science and Engineering, Virginia Tech, Blacksburg, Virginia 24060

(Received 10 September 2006; accepted 28 April 2007; published online 15 June 2007)

Systematic investigation of large amplitude ac excitation effects on the polarization–electric-field (P-E) and strain–electric-field (ε-E) characteristics of both "hard" and "soft" $Pb(Zr,Ti)O_3$ ceramics was performed. Upon ac excitation, a gradual disappearance of the shift on P-E response along the E axis (E_{int}) was observed for the hard piezoelectric; however, no such change was observed for the soft one. With increasing ac excitation cycle number, for a hard piezoelectric the internal dipolar field and mechanical quality factor Q_m decrease by following an exponential time law, whereas the electromechanical and dielectric properties increase with significant deviation from the time law. It is proposed that additional domain release mechanism may exist for hard piezoelectric, besides the decrease of internal dipolar field upon continuous ac excitation. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2745437]

I. INTRODUCTION

High power piezoelectric materials offer increased energy density; thus, a reduction in the size of ultrasonic motors, piezoelectric transformers, and high power sonar can be achieved. For these electromechanical transduction applications, large ac electric fields need to be applied, which can be relatively high. The investigation of piezoelectric property change under continuous ac field excitation is a very important issue for both practical application and theoretical analysis of domain dynamics.

The internal dipolar field (E_{int}) is shown as a shift of polarization–electric-field (P-E) hysteresis loop along the E axis (electric field) without applying external dc bias, which was found to be associated with acceptor dipole defect clusters.[1–5] Aging of the physical properties in lower-valent iron-modified $Pb(Zr,Ti)O_3$ (PZT) was found to follow logarithmiclike responses in time domain and can be attributed to acceptor dipolar defects.[5–7]

Direct observation of a submicroscopic bending of domain walls between pinning sites was reported.[8–10] Electron microscopy investigations of K-modified PZT have shown the development of "wavy" domains from normal micron sized domains when increasing K concentration. Such wavy domains were attributed to the domain wall pinning effect.

In previous paper,[11] we reported a significant increase of Q_m with time t for "hard" PZTs, whereas no such change was observed for the "soft" ones during the same time period. Correspondingly, a pronounced increase in E_{int} was observed to follow $\ln(t)$. This observation could not be explained by traditional domain wall pinning model as Q_m developed during a relatively long time period (several hours after poling), which is very different from domain wall pinning process.

Application of continuous ac electric field on a fully aged Fe-modified PZT resulted in a gradual decrease in E_{int} and corresponding gradual increase in the dielectric constant K. The "fresh" state was gradually recovered.[12] So far, only few investigations were made on materials property changes during internal dipolar field relaxation process. Very few papers related the large signal parameter (P-E hysteresis shift) to small signal materials property changes. In this paper, systematic investigation of P-E hysteresis and related materials property changes upon continuous ac excitation will be conducted. A mechanism analysis of the piezoelectric property changes during this process will be performed. The experimental results suggested that besides internal dipolar field relaxation, additional domain wall release mechanism exists upon continuous large amplitude ac excitation in hard piezoelectric.

II. EXPERIMENTAL PROCEDURE

In this study the following compositions were investigated:

(1) $0.9Pb(Zr_{0.52}Ti_{0.48})O_3-0.1Pb(Sb_{0.067}Mn_{0.033})O_3-0.2$ at. % RE (Yb, Nd, La),
(2) commercialized hard PZT (APC 841, APC International Ltd.), and
(3) commercialized soft PZT (Type D, Taiheiyo Cement Corporation).

The samples were cut into $40\times7\times1$ mm^3 plates. The

---

[a)]Electronic mail: gaomary@yahoo.com

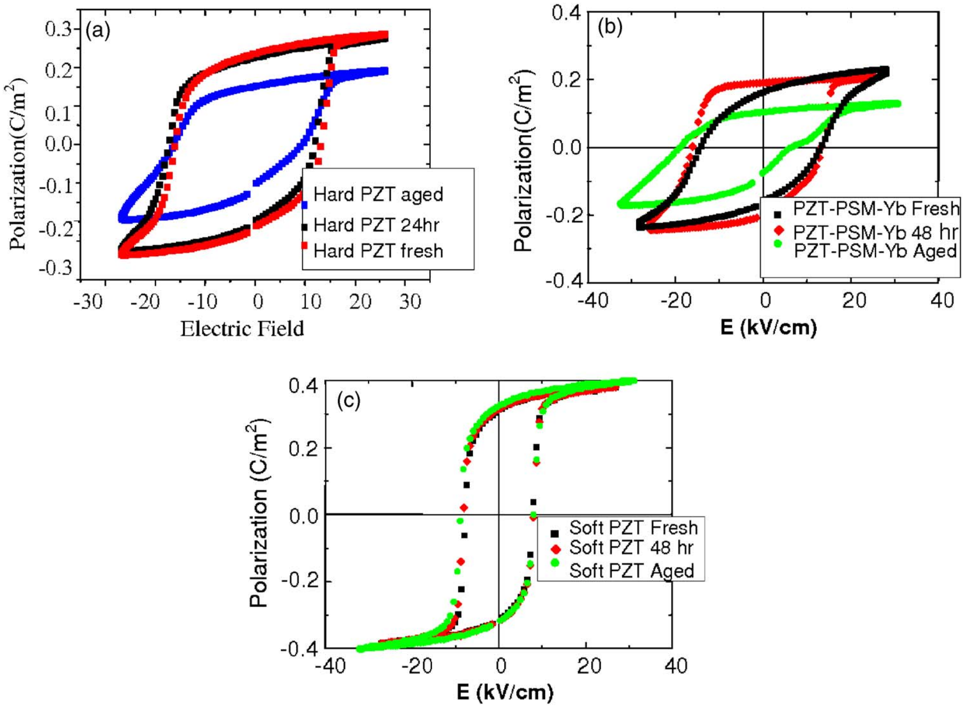

FIG. 1. (Color online) *P-E* response of (a) commercialized "hard" PZT, (b) RE doped PZT-PSM system, and (c) commercialized "soft" PZT during aging process (Ref. 11).

poling process was conducted by immersing samples in silicon oil at 130 °C under an electric field of 30 kV/cm for 15 min.

The electromechanical properties were measured by using a standard impedance analyzer. Spectra were obtained under a constant low-level voltage drive by using an HP4194 impedance analyzer. From the resonance f_r (i.e., the admittance maximum) and the antiresonance f_a (i.e., the admittance minimum) frequencies, the piezoelectric constant, the elastic compliance, the mechanical quality factor, and the electromechanical coupling coefficient were determined according to IEEE standard.[13] For a rectangular piezoceramic plate, for example, the transverse coupling coefficient k_{31} can be calculated as $k_{31}^2/(1-k_{31}^2)=(\pi^2/4)(\Delta f/f_r)$, where $\Delta f=f_a-f_r$. The mechanical quality factor Q_m was calculated as $Q_m=f_r/f_2-f_1$, where f_r is the resonance frequency and f_2/f_1 are the frequencies 3 dB down from the maximum admittance.

The *P-E* and ε-*E* curves (strain versus electric field) were measured by using a modified Sawyer-Tower circuit in conjunction with a linear variable differential transducer. The maximum amplitude of the applied ac electric field was as high as 30 kV/cm. The frequency was 1 Hz (the time period for each cycle was 1 s). For high field measurement, samples were immersed in the insulating liquid of Galden HT-200 to prevent arcing.

III. EXPERIMENTAL RESULTS

A. *P-E* and ε-*E* characteristics

Previously we observed the development of E_{int} with time for hard piezoelectric ceramics. Figures 1(a)–1(c) show the *P-E* response of commercialized hard PZT, rare earth modified $\text{Pb(Zr,Ti)O}_3-\text{Pb(Sb,Mn)O}_3$ (PZT-PSM-RE), and soft PZT during aging process,[11] respectively. The measurements were conducted at 10 min, 48 h, and one month after poling. At 10 min, *P-E* response was a symmetric loop for all of hard piezoelectric specimens, indicating that no significant E_{int} was developed yet; however, after 48 h, a slight shift of the *P-E* response along the *E* axis was found; whereas after one month, an E_{int} field as large as 10 kV/cm was shown in the *P-E* loop of PZT-PSM-RE. This is the highest E_{int} observed among all of the investigated specimens. Contrary to hard piezoelectric, no such shift of *P-E* response along the *E* axis was observed for soft PZTs, as shown in Fig. 1(c), indicating that no E_{int} was developed during the entire aging process.

In this paper, systematic investigation was conducted on the effect of continuous ac excitation on the *P-E* and ε-*E* responses of both hard and soft PZTs. Under continuous ac excitation, a relaxation of E_{int} was found for hard-type piezoelectric ceramics, while no such change was observed for soft PZTs. Figure 2(a) shows the *P-E* response of an aged commercialized hard PZT under continuous 30 kV/cm ac field. At the first cycle $N=1$, the *P-E* response of the aged hard PZT showed a shift along the *E* axis, with a remnant polarization $P_r\sim 0.15$ C/m^2; however, with increasing ac excitation cycle number *N*, *P-E* loop gradually became symmetric and finally E_{int} disappeared. At $N=360$, P_r increased to ~ 0.28 C/m^2, which is close to but slightly higher than that of the "freshly" poled sample, as shown in Fig. 1(a).

Similar tendency was observed for PZT-PSM-RE. Figure 2(b) shows the *P-E* relaxation process for PZT-PSM-RE. The aged specimen had an asymmetric *P-E* loop and a significant shift along the *E* axis; however, with increasing cycle number, the *P-E* response gradually became symmetric. At $N=1$, P_r was ~ 0.18 C/m^2; at $N=360$, P_r increased to ~ 0.35 C/m^2. This result indicates that with increasing ac excitation cycle number, the domain stabilizing force decreases and the domain becomes switchable again.

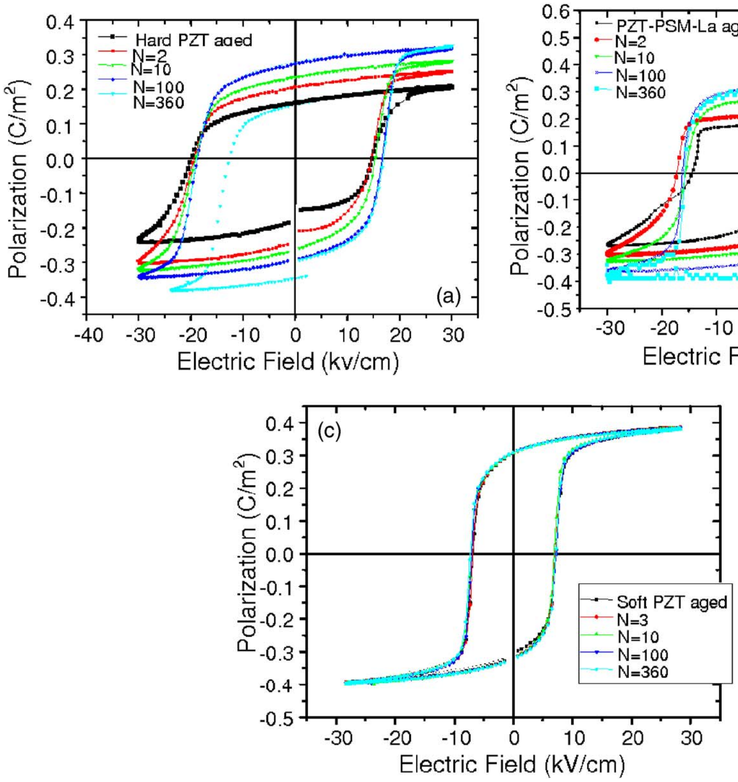

FIG. 2. (Color online) *P-E* hysteresis relaxation of (a) commercialized "hard" PZT, (b) RE doped PZT-PSM system, and (c) commercialized "soft" PZT.

Contrary to hard-type PZTs, soft PZT did not show *P-E* relaxation under continuous ac excitation. No such change of *P-E* response was observed during ac excitation process, as shown in Fig. 2(c). It is clear that with or without E_{int} is the root difference between hard and soft PZTs.

Figure 3 summarizes P_r versus cycle number N for both commercialized hard PZT and PZT-PSM-RE. For both materials, P_r increased with increasing N. Continuous ac excitation resulted in the decrease of domain stabilizing force E_{int} or domain wall releasing from pinning sites, which may contribute to the increase of switchable polarization.

The value of E_{int} can be determined by differential changes of the *P-E* response between consecutive measurements. Naturally, this method has the limitation that noise is introduced by numerical differentiation. Figure 4 shows the calculated E_{int} as a function of N. If we define time t as the product of time period for each cycle (1 s for this study) and cycle number N, the decrease of E_{int} with time can be well fitted to the exponential relation

$$E_{\text{int}}(t) = E_{\text{int}}(0)\exp(-t/\tau), \tag{1}$$

where $E_{\text{int}}(0)$ is the internal dipolar field in the aged condition and τ is a characteristic relaxation time. We found this equation valid only within limited ranges of temperature T and field strength E, as τ depends on both T and E.

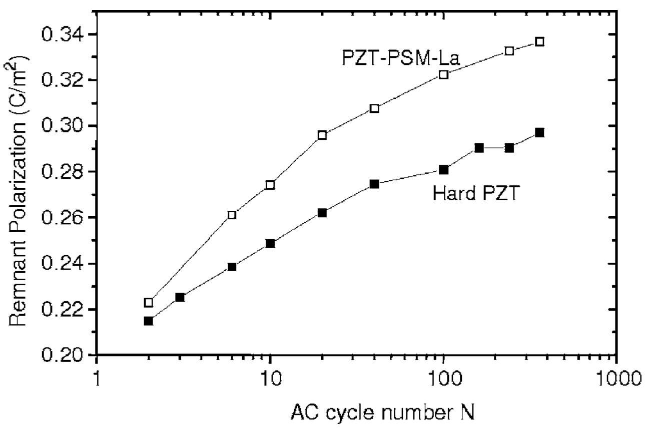

FIG. 3. The remanent polarization P_r increase as a function of ac cycle number N for commercialized "hard" PZT and RE doped PZT-PSM ceramics.

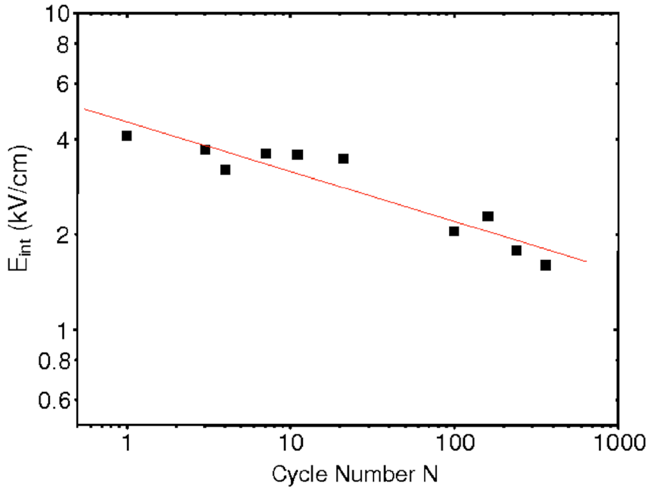

FIG. 4. (Color online) Calculated internal dipolar field as a function of ac cycle number N for commercial "hard" PZTs.

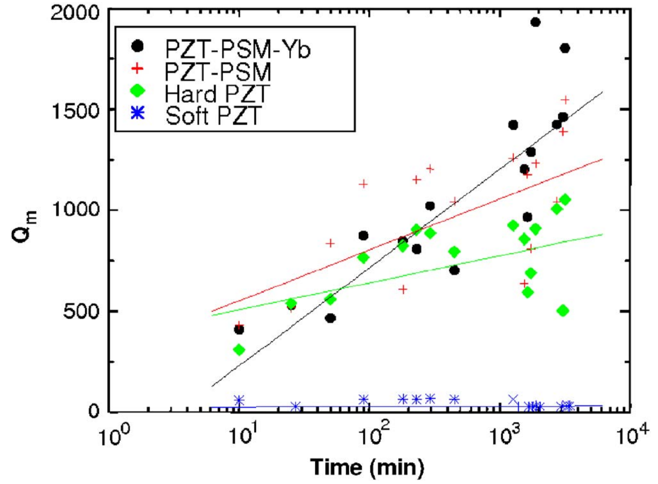

FIG. 5. (Color online) Q_m vs time for commercialized "hard" PZTs, PZT-PSM-RE, and "soft" PZTs within 48 h (2880 min) after poling (Ref. 11).

B. Resultant property changes due to relaxation of E_{int}

Previously we observed a significant increase of the mechanical quality factor Q_m as a function of time after poling.[11] Figure 5 shows the Q_m versus time for commercialized hard PZTs, PZT-PSM-RE, and soft PZTs within 48 h (2880 min) after poling. The initial Q_m value of PZT-PSM-RE was only 400 at 10 min after poling; however, at 2880 min after poling, Q_m increased up to 1600. The Q_m value increased significantly for hard piezoelectrics and the increasing rate seems to be related to the degree of "hardness:" the harder the materials are, the higher the Q_m increasing rate is. However, for soft PZT no Q_m change was observed within the same time frame. The pronounced increase of both E_{int} and Q_m was observed to follow a logarithm time relationship $\ln(t)$, which is relatively slow and significantly different from exponential relationship as E_{int} relaxes during ac excitation process, as shown in Fig. 4 and Eq. (1). The development of E_{int} occurs very gradually because oxygen vacancy diffusion takes relatively long time.[11] The developed internal dipolar fields clamp polarization switching, via a conjugate coupling to P. The slow development of Q_m and E_{int} at room temperature is contradictory to the traditional domain wall pinning model as at room temperature pinning sites should not have enough mobility to diffuse to domain boundaries, when Q_m and E_{int} develop.

The deaging process caused by ac excitation is much faster than aging process. The oxygen diffusion rate is very slow compared to external ac cycle rate. The oxygen vacancy-acceptor defect dipoles do not have enough time to reorient to the direction parallel to spontaneous polarization during each ac cycle; therefore E_{int} decreased at a much higher rate upon ac excitation as compared to the process when E_{int} develops.

Internal dipolar fields E_{int} play an important role in domain stabilization. The reduced E_{int} will result in an enhancement in the physical properties and a decrease in the mechanical quality factor.

Figures 6(a)–6(c) show how electromechanical coupling coefficient k_{31} changes during internal dipolar field relaxation process for a commercialized hard PZT, PZT-PSM-RE, and commercialized soft PZT, respectively. With increasing time t (or ac cycle number N), k_{31} can be seen to increase for both commercialized hard PZT and PZT-PSM-RE. The k_{31} dependence on t (or N) deviated from the exponential time law as shown in Eq. (1), which E_{int} follows during the relaxation process. This result suggests that additional domain

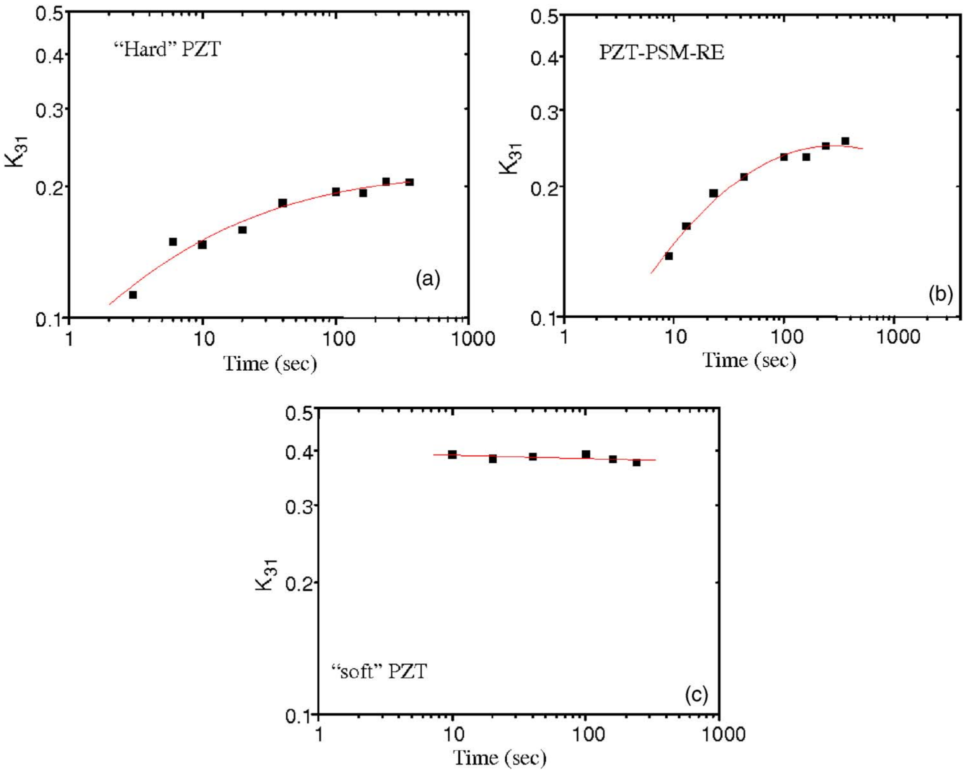

FIG. 6. (Color online) Electromechanical coupling coefficient k_{31} value change during internal dipolar field relaxation process for (a) commercialized "hard" PZT, (b) RE doped PZT-PSM, and (c) commercialized "soft" PZT.

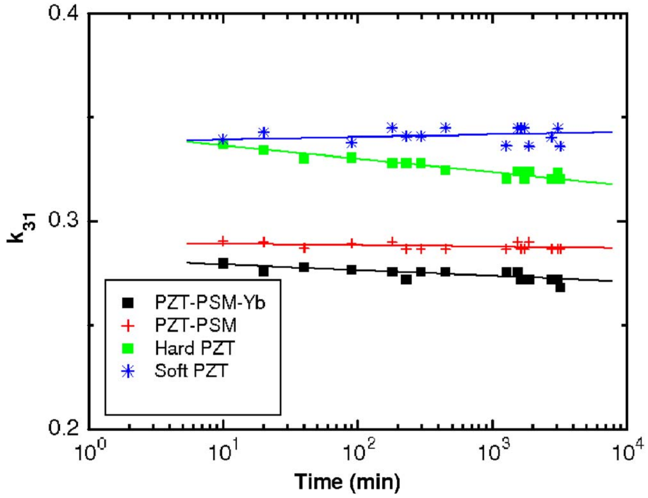

FIG. 7. (Color online) Electromechanical coupling coefficient k_{31} value change during aging with the development of internal dipolar field E_{int} for commercialized "hard" PZT, RE doped PZT-PSM, and commercialized "soft" PZT.

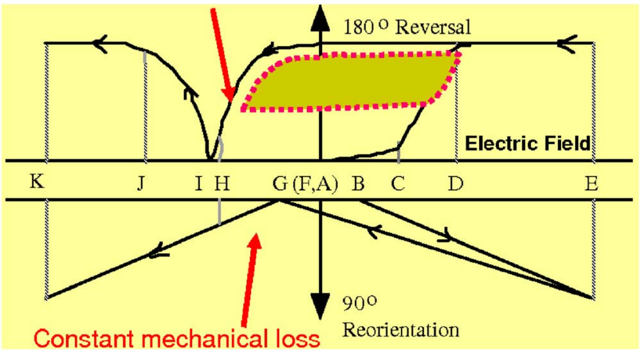

FIG. 8. (Color online) Uchida-Ichida model (Refs. 14 and 15) was applied to understand how E_{int} affects Q_m and other electromechanical properties of "hard" piezoelectric.

release mechanism besides the decrease of E_{int} may contribute to the electromechanical performance of hard piezoelectric upon continuous ac excitation. In addition, it is important to notice that the increase of k_{31} was quite significant for hard piezoelectric ceramics upon ac excitation, whereas no such change was observed for soft PZTs. At $t=360$ s ($N=360$), k_{31} increased by 0.12 for PZT-PSM-RE; however, k_{31} decreased only by 0.01 during the aging process, as shown in Fig. 7.[11] It is very clear that there is a significant difference of electromechanical property changes between aging and deaging processes. Besides the decrease of E_{int}, additional domain release mechanism may exist which contribute to the significant k_{31} increase during ac excitation process. Uchida-Ichida model[14,15] was applied to understand how E_{int} affects Q_m and other electromechanical properties of hard piezoelectric, as shown in Fig. 8. E_{int} has a similar effect as external positive dc bias on domain dynamics. Electrical field has only gradual influence on 90° domain switching, whereas above certain field level, it could significantly affect 180° domain reversal.[15] It was believed that mechanical loss is mainly associated with 90° domain switching whereas dielectric loss is associated with 180° domain reversal.[16] At resonance heat generation is dominantly contributed by intensive mechanical loss. Considering that intensive mechanical loss consists of extensive dielectric loss;[16] above a certain field level extensive dielectric loss increases drastically (as shown in Fig. 8); thus the mechanical quality factor Q_m (inverse value of mechanical loss) should be a field sensitive parameter, whereas k_{31} should not be very sensitive to electric field. The effects of E_{int} on Q_m and other electromechanical properties during aging process after poling supported the

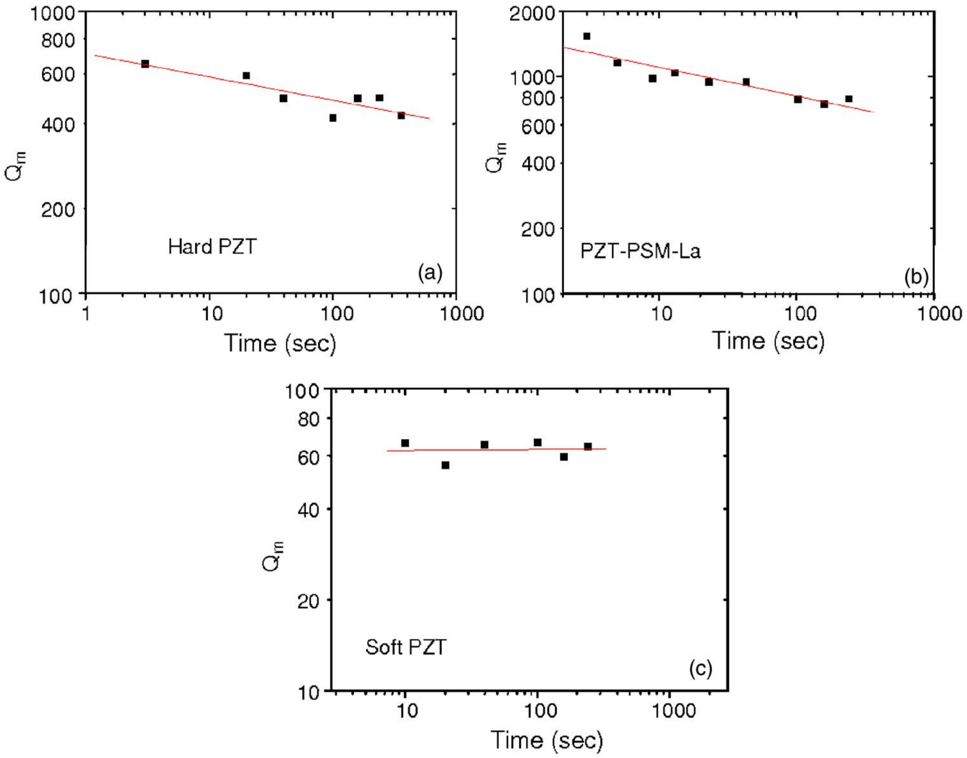

FIG. 9. (Color online) Mechanical quality factor Q_m value change during internal dipolar field relaxation process for (a) commercialized "hard" PZT, (b) RE doped PZT-PSM, and (c) commercialized "soft" PZT.

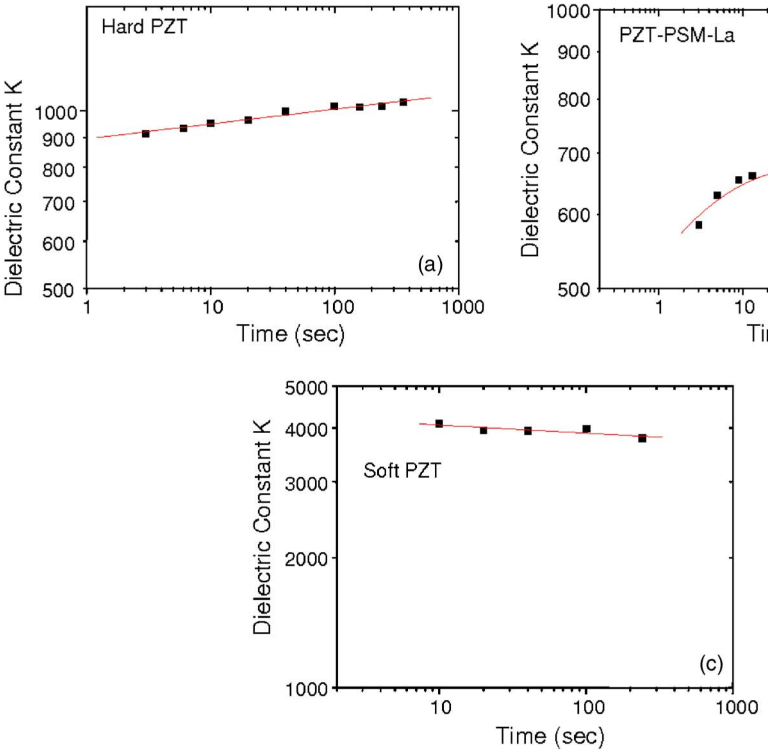

FIG. 10. (Color online) Dielectric constant K value change during internal dipolar field relaxation process for (a) commercialized "hard" PZT, (b) RE doped PZT-PSM, and (c) commercialized "soft" PZT.

conjecture that E_{int} has a significant impact on Q_m but not on k_{31}.[11] The possible explanation for the significant k_{31} increase upon large amplitude ac excitation could be attributed to the depinning of defect clusters from domain boundaries, in addition to E_{int} decrease. Internal dipolar field could be formed by reorienting the dipolar defects via oxygen diffusion and the defect clusters do not necessarily need to be accumulated around the domain boundaries. However, to form domain wall pinning, defect complexes need to physically diffuse into domain boundaries and pin the walls. Internal dipolar field and domain wall pinning effects could be somewhere overlapping but different mechanisms contribute to domain dynamics.

Figures 9(a)–9(c) show Q_m as a function of time for commercialized hard PZT, PZT-PSM-RE, and commercialized soft PZT upon continuous ac excitation, respectively. The Q_m can be seen to decrease with time for hard piezoelectric ceramics [Figs. 9(a) and 9(b)] by following an exponential relation $\exp(-t/\tau)$ as shown in Eq. (1), whereas no such change was observed for soft PZT [Fig. 9(c)]. This observation is consistent with the conjecture that E_{int} has a large impact on Q_m but not on electromechanical properties. The pronounced decrease of mechanical quality factor for hard piezoelectric ceramics during ac excitation accompanies the increased energy transduction from electrical to mechanical forms, as k_{31} increases. More mechanical energy is transformed from electrical energy, but a higher fraction of energy is dissipated as heat. Electrically induced shape changes which occur on domain release are inherently dissipative. The changes in Q_m have a large impact on the application of high power transducer materials, as mechanical loss factor contributes dominantly to the heat generation under resonance drive. The decrease of Q_m and E_{int} during ac excitation limits possible driving life of high power operations.

Figures 10(a)–10(c) show the dielectric constant K as a function of time for a commercialized hard PZT, PZT-PSM-RE, and commercialized soft PZT, respectively. K was observed to increase with time upon ac excitation for "hard" piezoelectric [Figs. 10(a) and 10(b)] however, a slight decrease of K was observed for soft PZT [Fig. 10(c)], which may be caused by microcrack generated during large amplitude ac excitation. A significant deviation from exponential time relationship was observed for PZT-PSM-RE specimens. K increased first and then decreased with further increasing ac excitation cycle number. PZT-PSM modified with various rare earth elements were investigated; however, similar tendency was observed for each specimen. The mechanism is not clear yet. Contrary to hard PZTs, no increase of dielectric constant, electromechanical properties, and decrease of Q_m were observed for soft PZT upon continuous ac excitation.

IV. SUMMARY

Systematic investigation of P-E, ε-E, and related electromechanical property change of both "hard" and "soft" piezoelectrics upon continuous large amplitude ac excitation was performed. E_{int} decreased with increasing ac cycle number, by following an exponential time relationship $\exp(-t/\tau)$. The decrease of E_{int} during ac excitation much faster than E_{int} develops by following $\ln(t)$ during aging process. The slow increase of Q_m and E_{int} during aging is contradictory to the traditional domain wall pinning model. The relaxation of E_{int} was shown to result in an increased transient coupling coefficient k_{31}. However, the increase of energy transduction from electrical to mechanical forms is accompanied by significantly increased mechanical energy dissipation as heat. For hard piezoelectric, k_{31} increased significantly upon continuous ac excitation, with a large deviation from the exponential time law, whereas E_{int} follows $\exp(-t/\tau)$ during relaxation. This observation indicates that additional domain release mechanism which contributes to enhanced electromechanical response exists besides the decrease of E_{int} upon ac excitation.

[1] H. H. Wieder, J. Appl. Phys. **30**, 1010 (1959).
[2] U. Robels, J. H. Calderwood, and G. Arlt, J. Appl. Phys. **77**, 4002 (1995).
[3] E. T. Keve, K. L. Bye, P. W. Whipps, and A. S. Annis, Ferroelectrics **3**, 39 (1971).
[4] K. Carl and K. H. Hardtl, Ferroelectrics **17**, 473 (1978).
[5] U. Robels and G. Arlt, J. Appl. Phys. **73**, 3454 (1993).
[6] U. Robels, R. Lohkamper, and G. Arlt, Proceedings of the ISAF'90, Urbana, IL, 1990 (unpublished).
[7] U. Robels, R. Lohkamper, and G. Arlt, IEEE Ultrasonic Symposium, 1991 (unpublished), p. 44.
[8] T. J. Yang, V. Gopalan, P. J. Swart, and U. Mohideen, Phys. Rev. Lett. **82**, 4106 (1999).
[9] Q. Tan, J. F. Li, and D. Viehland, Philos. Mag. B **76**, 59 (1997).
[10] Q. Tan, J. Li, and D. Viehland, Appl. Phys. Lett. **75**, 418 (1999).
[11] Y. Gao, K. Uchino, and D. Viehland, Jpn. J. Appl. Phys., Part 1 **45**, 12 (1996).
[12] K. Carl and K. H. Hardt, Ferroelectrics **17**, 473 (1978).
[13] IRE Standard (1957).
[14] Y. Gao, K. Uchino, and D. Viehland, J. Appl. Phys. **101**, 054109 (2007).
[15] N. Uchida and T. Ikeda, Jpn. J. Appl. Phys. **4**, 867 (1965).
[16] K. Uchino, IEEE Trans. Ultrason. Ferroelectr. Freq. Control **48**, 307 (2001).