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Doru C. Lupascu

Fatigue in Ferroelectric Ceramics and Related Issues

With 80 Figures

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To Kathrin, my parents, and my brother

Preface

Ferroelectric devices are nowadays widely found in advanced technology as well as everyday household machinery. The broad range of physical properties in this class of materials has allowed for the evolution of devices ranging from piezoelectric components and optical devices to ferroelectric non-volatile memories in microelectronics. Examples extend from simple gas igniters, sonic and ultrasonic sensors and actuators to the more advanced ultrasonic motors, high precision positioning devices, optical switches and recently permanent memories for computer applications. Many more applications are anticipated. Particularly the fact that many parts are at the edge of becoming large numbered consumer products necessitates a much higher reliability of the devices. Two aspects are relevant with respect to this, an adequate device design to ensure a minimum loading of the ferroelectric material on the one hand, and optimized material properties for high reliability at maximum performance on the other. Unfortunately, ferroelectric materials are mostly brittle both as single crystals as well as polycrystalline material, and in the worst case even water soluble crystals. Particularly tensile mechanical loading will often lead to an immediate failure of the device. On longer terms, cyclic loading will deteriorate the performance of the material. Multiple loading will either lead to cyclic crack growth or the modification of the material properties at the mesoscopic and microscopic level. During the last fifty years of material development it has become clear that effects on a wide range of time and size scales down to the atomic level are relevant for the properties of ferroelectrics. In these same ranges of times and sizes the sources of fatigue have to be identified. The present text is an attempt to assign the fatigue phenomenon in bulk ferroelectric ceramics to its microscopic, mesoscopic, and macroscopic sources.

Darmstadt, October 2003 *Doru Constantin Lupascu*

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Symbols and Abbreviations

a.c.	Alternating current
AE	Acoustic emissions
AFM	Atomic force microscopy
BHP	Barkhausen pulses
BiT	Bismuth titanate ($Bi_4Ti_3O_12$)
BLT	Lanthanum substituted bismuth titanate ($Bi_{3.25}La_{0.75}Ti_3O_{12}$)
BNdT	Bismuth titanate ($Bi_4Ti_3O_{12}$)
$Bi_{4-x}Nd_xTi_3O_{12}$ (BNdT) BTO	Bismuth titanate ($Bi_4Ti_3O_{12}$)
d.c.	Direct current
EELS	Electron energy loss spectroscopy
EPR	Electron paramagnetic resonance
ESR	Electron spin resonance (=EPR)
FEM	Finite element modeling
GMO	Gadolinium molybdate ($Gd_2(MoO_4)_3$)
HRTM	High resolution transmission electron microscopy
LSCO	Lanthanum strontium cobaltdate $(La, Sr)CoO_3$
PBN	Lead-barium-niobate ($Pb_xBa_{1-x}Nb_2O_3$)
PLZT	Lanthanum doped lead-zirconate-titanate ($La_yPb_{1-y}(Zr_xTi_{1-x})O_3$)
PMN	Lead magnesium niobate ($Pb(Mg_{1/3}Nb_{2/3})O_3$)
p_{O_2}	Oxygen partial pressure
PT	Lead-titanate ($PbTiO_3$)
PZN	Lead-zink-niobate ($Pb(Zn_{1/3}Nb_{2/3})O_3$)
PZT	Lead-zirconate-titanate ($Pb(Zr_xTi_{1-x})O_3$)
RTA	Rapid thermal annealing
SBN	Strontium barium niobate ($Sr_{1-x}Ba_xNb_2O_6$)
SBT	Strontium bismuth tantalate ($SrBi_2Ta_2O_9$)
SEM	Scanning electron microscopy
$\Sigma 3$	Crystallographic coincidence lattice twin of period 3

SIMS	Scanning ion mass spectrometry
TEM	Transmission electron microscope
UV	Ultraviolet
$V_O^{\bullet\bullet}$	Twice positively charged oxygen vacancy
XRD	X-ray diffraction
XPS	X-ray excited photoelectron spectroscopy
YBCO	Yttrium barium copper oxide ($YBa_2Cu_3O_7$)
$\otimes$	Dyadic or outer product (Malvern, 1969)
$^{\bullet}$, $'$, $^{\times}$	Positively, negatively, and un-charged ion with respect to lattice position in an ionic crystal according to Kröger and Vink (1956)