

### Andrea Raphaela Engert (Autor) Experimental studies on crack growth in ferroelectric ceramics



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# Chapter 1 Introduction

Ferroelectric ceramics are used, amongst other applications, as electro-mechanical transducers, converting mechanical forces into an electric potential (direct piezoelectric effect) or vice versa (inverse piezoelectric effect). They offer advantages such as nanometer precision in positioning, large force generation and fast response time (in the us-range) in actuator applications (electro-mechanical transducer), high sensitivity as force or displacement sensors (mechano-electrical transducer), and operation in a high frequency range as ultrasonic transmitters or receivers.<sup>1,2</sup> During their lifetime, ferroelectric ceramics must be able to operate under long-term cyclic electro-mechanical loading. While they continue to find increasing use, questions regarding lifetime and reliability are still investigated. Thereby the prediction of crack growth under electromechanical loading is difficult due to the complex material behavior of ferroelectric ceramics. This work seeks to add to the understanding of fracture under combined quasi-static mechanical and constant or cyclic electrical loading. In the following a short introduction and literature review is given on ferroelectric ceramics and on the fracture mechanics of ferroelectric ceramics. Thereby crack growth under mechanical loading as well as under direct current (DC) and alternating current (AC) electrical loading is addressed. Afterwards the motivation and the contents of this work are introduced in Chapter 2. In Chapter 3 the samples and the two experimental methods used in this work to generate the experimental data are described. The experimental results are presented in Chapter 4 and 5. These two chapters can be understood independently from each other. While the first one presents a macroscopic analysis of the mechanical loads required for crack advance, the second one presents a microscopic study of the dielectric properties of the crack. The work is concluded with a summary, which also contains a short outlook on further experiments.

<sup>&</sup>lt;sup>1</sup> Jaffe, Cook & Jaffe (1971), p. 275

<sup>&</sup>lt;sup>2</sup> Haertling (1999), p. 812

Parts of this work, mainly presented in Chapter 4, were developed within the diploma thesis of Dipl.-Ing. Matthias Mecklenburg.<sup>3</sup> The conceptual ideas for the measurements were formed in discussions with Prof. Dr. rer. nat. Gerold A. Schneider, Dr. rer. nat. Hans Jelitto and Dr.-Ing. Peter Neumeister. All FE calculations, which were used to evaluate the data, were provided by Dr.-Ing. Peter Neumeister. The contents of this chapter have been published to a great extent in the journal article "Influence of small cyclic and DC electrical loads on the fracture toughness of ferroelectric ceramics" in the Journal of the European Ceramic Society, 31 (2011) 531-540, Copyright 2010 by Elsevier.<sup>4</sup> The material from this journal article is reused in this thesis with permission.

<sup>&</sup>lt;sup>3</sup> Mecklenburg (2009), p. 31-95

<sup>&</sup>lt;sup>4</sup> Engert, Neumeister, Mecklenburg *et al.* (2011)

## Chapter 2 Fundamentals

### 2.1 Ferroelectric ceramics

According to Telle ceramics can be described as non-metallic, inorganic materials with a crystalline content of at least 30 %.<sup>5</sup> Ferroelectric ceramics are polycrystalline ceramics. A broad overview about ferroelectric ceramics is given e.g. in the textbook of Jaffe.<sup>6</sup> Essential information is also found in textbooks of physics<sup>7</sup> or solid state physics.<sup>8,9</sup> A review paper by Haertling summarizes the recent developments in the field of ferroelectric ceramics.<sup>10</sup> This Chapter 2.1 introduces general dielectric properties of materials and summarizes the important micro- and macroscopic properties of ferroelectric ceramics for this work.

### 2.1.1 Polarization

The dipole moment p pointing from a negative charge -q to a positive charge q separated by a distance I is described as:<sup>11</sup>

$$\boldsymbol{p} = q\boldsymbol{l} \qquad \text{or} \qquad \boldsymbol{p}_i = q\boldsymbol{l}_i \tag{2.1}$$

The subscript i names the vector components, here of p and l, on the corresponding Cartesian axes x, y, and z.

Dielectric materials are non-conducting polarizable materials. Whereas nonpolar dielectric materials acquire an induced dipole moment in an electric field, which disappears if the perturbing field is removed, polar materials can have their existing dipole moments temporarily modified or permanently oriented by an applied field. According to these two

<sup>&</sup>lt;sup>5</sup> Telle (2007), p. 2

<sup>&</sup>lt;sup>6</sup> Jaffe, Cook & Jaffe (1971), complete book

<sup>&</sup>lt;sup>7</sup> Meschede (2010), p. 334-339

<sup>&</sup>lt;sup>8</sup> Ashcroft & Mermin (1976), p. 554-557

<sup>&</sup>lt;sup>9</sup> Kittel (2005), p. 455-482

<sup>&</sup>lt;sup>10</sup> Haertling (1999)

<sup>&</sup>lt;sup>11</sup> Meschede (2010), p. 330

properties, the total polarizability can be separated into two parts: into displacement polarization (displacement of the electron shell relative to a nucleus (electronic dipole) or displacement of charged ions with respect to other charged ions (ionic dipole)) and orientation of already existing permanent electric dipole moments in an applied electric field.<sup>12</sup> An infinite dielectric crystal of identical cells, each with a dipole moment  $p_0$  of a unit cell, has a dipole moment of  $p_0$  times the number of cells in the crystal, and thus a polarization P of

$$\boldsymbol{P} = \frac{\boldsymbol{p}_0}{\boldsymbol{\nu}_0} \quad \text{or} \quad \boldsymbol{P}_i = \frac{(\boldsymbol{p}_0)_i}{\boldsymbol{\nu}_0} \tag{2.2}$$

throughout the crystal, with  $v_0$  being the volume of crystal unit cell.<sup>13</sup>



Figure 2.1 (a) Schematic drawing of a dielectric slab with a polarization P (blue solid line) in a capacitor of area A and separation d. The free charge Q and bound charge  $q_{\text{bound}}$ , the electric field E (red solid line) and the electric displacement D (black solid line) are also shown. (b) Electric displacement field lines (black solid lines) and (c) electric field lines E = U/d (red solid lines) between the plates of a capacitor filled with a dielectric. Part of the electric field lines (red dotted lines), originating from charge on the electrodes, are compensated by a depolarization field  $E_{\text{depol}} = P/\varepsilon_0$  (blue dotted lines), originating from polarization charge.

If a voltage U is applied to a pair of parallel conducting plates, with a distance d between them, a positive charge  $Q^+$  accumulates at the positive and a negative charge Q accumu-

<sup>&</sup>lt;sup>12</sup> Meschede (2010), p. 335-336

<sup>&</sup>lt;sup>13</sup> Ashcroft & Mermin (1976), p. 539

lates at the other plate. These charges give rise to an electric field  $E = U/d = Q/(\varepsilon_0 A)$ between the plates, pointing from the positive to the negative plate. Hereby A is the area of the capacitor and  $\varepsilon_0 = 8.85 \cdot 10^{-12} \text{ F/m}$  is the vacuum permittivity. The ratio between the amount of charge stored on each plate and the applied voltage is described by the capacity  $C^{14}$ .

$$C = \frac{Q}{U} = \frac{\varepsilon_0 A}{d} \,. \tag{2.3}$$

If a linear, homogeneous dielectric slab of area A and height d is inserted between this pair of parallel conducting plates (Figure 2.1(a)), it is polarized. Hereby the macroscopic polarization depends on the applied electric field according to:

$$\boldsymbol{P} = \boldsymbol{\chi} \boldsymbol{\varepsilon}_0 \boldsymbol{E} \qquad \text{or} \qquad \boldsymbol{P}_i = \boldsymbol{\chi}_{ij} \boldsymbol{\varepsilon}_0 \boldsymbol{E}_j \tag{2.4}$$

where  $\chi$  is the tensor of the electric susceptibility of an anisotropic material in generalization of the isotropic case.<sup>15</sup> In the interior of the dielectric slab, the polarization is homogeneous. At the surface where the polarization decreases to zero, the same acts as the source of a depolarizing field (blue dotted lines in Figure 2.1(b)). The depolarization field, induced by this "bound" surface polarization charge, is directed opposite to the applied electric field (red lines) and therefore tends to reduce the strength of which. If the capacitor is connected to a power-supply, applying a constant electric load, the vacuum "free" surface charge density  $Q_0/A$  on the plates is increased by a charge equal to the polarization P to maintain the strength of the electric field between the plates at U/d. The capacitance of a capacitor is therefore higher if, instead of air, a dielectric fills the space between the plates:

$$C = \frac{Q}{U} = \frac{\varepsilon_0 \varepsilon_r A}{d} = \frac{\varepsilon A}{d}, \qquad (2.5)$$

where  $\boldsymbol{\varepsilon}_{r} = 1 + \boldsymbol{\chi}$  is the relative dielectric constant of the material and  $\boldsymbol{\varepsilon} = \varepsilon_{0} \boldsymbol{\varepsilon}_{r}$  describes the absolute dielectric constant of the material.<sup>16</sup>

D is defined by Gauss's law, which describes the relationship between an electric field and the generating electric charges. It states that the electric flux  $\Theta_{E,A}$  through any closed surface is proportional to the enclosed net electric charge, including both free  $q_{\text{free}}$  and bound (polarization) charges  $q_{\text{bound}}$ :

<sup>&</sup>lt;sup>16</sup> Meschede (2010), p. 334



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<sup>&</sup>lt;sup>14</sup> Meschede (2010), p. 328

<sup>&</sup>lt;sup>15</sup> Meschede (2010), p. 335