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# THE APPLICATION OF NONLINEAR DYNAMICS IN THE STUDY OF FERROELECTRIC MATERIALS[1]

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It is well known that the structural phase transitions in ferroelectric materials are connected with strong nonlinear properties. So we can expect all features of nonlinear dynamical systems such as period–doubling cascades and chaos in a dynamical system that contains ferroelectric materials. Therefore we can apply nonlinear dynamics to these ferroelectric materials and we are doing it in two directions: (i) We study the structural phase transitions by analyzing the large signal behaviour with the means of nonlinear dynamics. (ii) We control the chaotic behaviour of the system with the method proposed by Ott, Grebogi and Yorke.

## 1. Introduction

The structural phase transitions in ferroelectric materials are connected with strong nonlinear properties [1]. So we should expect all features of nonlinear dynamical systems as period–doubling and deterministic chaos in these materials. The aim of the presented paper is to show that the application of analysis methods of dissipative chaotic systems is a useful tool for the study of structural phase transitions and for the study of the large signal behaviour of ferroelectric materials. The investigation of the large signal behaviour is stimulated by the shift in the ferroelectrics research and development to thin films with the aim of miniaturization.

So we are enabled to apply the ideas of nonlinear dynamics to our system. Besides the scientific aspect of applying these ideas to a real dynamical system many of those ideas are becoming relevant. One of these ideas is to control the irregular chaotic motion. We applied one controlling method to our system successfully.

The paper tries to connect the topics shown in Figure 1 in the following way: In the first part we describe our system and after that we are going on to model it. This yields an ordinary differential equation with a set of free model–parameters that are to

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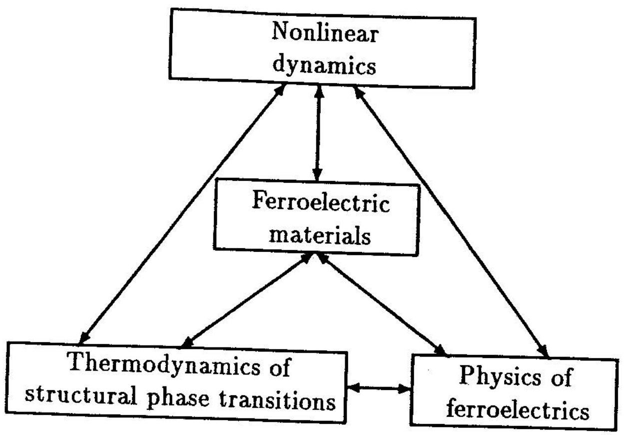


Fig. 1. Schematic illustration of the relation between the different topics in this paper.

be supposed to describe the ferroelectric material. The next step is to interprete these parameters physically. After that we determine the free parameters and describe with the help of them different ferroelectric materials. In the last part then we will show in which way we can control the irregular and chaotic regions of the dielectric nonlinear series–resonance circuit.

## 2. The dielectric nonlinear series–resonance circuit

### 2.1 Experimental Set–Up

The system we deal with is the dielectric nonlinear series–resonance circuit. Although quite simple, it is well suited for investigating the dynamical large signal behaviour of a wide range of ferroelectric systems, such as single crystals, thin films and ferroelectric liquid crystals[2], [3]. This circuit is built of a linear air coil and a capacitance filled with the ferroelectric material to be investigated, as shown in Figure 2.

The system as a whole is excited by a sinusoidal voltage. Because of the nonlinearities of the capacitance the circuit is an experimental realization of a nonlinear oscillator. The circuit has three external parameters to control its properties: Frequency and amplitude of the driving voltage and the temperature that fix the dielectric properties of the capacitor.

For reasons of measurement two further components are attached to the resonance circuit introduced above. Using the linear capacitance $C_m$ and the linear resistor $R_m$ makes it is possible to record signals proportional to the dielectric displacement $D$ on the specimen and the current density $j = dD/dt$ with time $t$ through the system



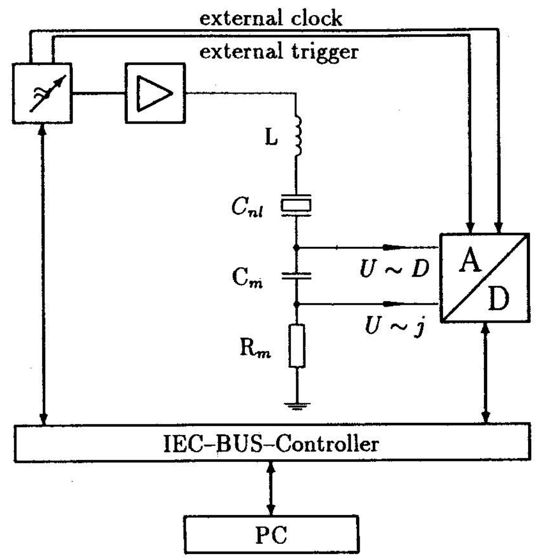


Fig. 2. Block diagram of the experimental setup.

respectively (see Figure 2). In order to be sure that nearly all the applied voltage $U_{ext} = U_0 \cos(\omega t)$ drops at the dynamical system $L$(air coil) and $C_{nl}$ (ferroelectric specimen) the unequality $R_m << 1/(\omega C_m) << 1/(\omega C_{nl})$ holds. The setup shown in Figure 2 yields a representation of the three–dimensional phase space flow in terms of $D$ (generalized location), $j$ (generalized momentum) and $t$ (time). With the help of a thermostatic or cyrostatic device (omitted in Figure 2) the temperature of the specimen can be placed into the interesting ferroelectric region that depends on the material.

The setup shown in Figure 2. makes it possible to measure:

**Time–series** for recording the complete phase–space,

**Poincaré–sections** as a stroboscopic view into the phase–space and

**Bifurcation–diagrams** for recording the qualitative behaviour of the dielectric nonlinear series–resonance circuit in the case of varying one external parameter.

So we meet all experimental requirements to apply the methods of nonlinear dynamics.

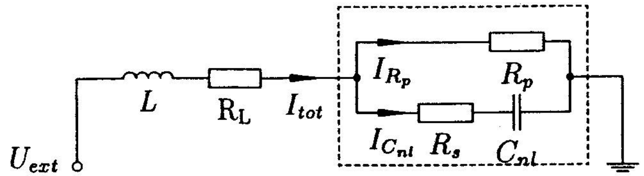


Fig. 3. The dielectric nonlinear series–resonance circuit with a substitute-network (dashed) for the ferroelectric capacitor.

## 2.2 The model–equation

The dielectric nonlinear series–resonance circuit can be described by a an ordinary differential equation. This model–equation can be yielded by exploiting two properties of the system:

(i) Its behaviour is macroscopic reproducible. That means that the external control parameters mentioned in the section above really control the dynamics of the system [2].

(ii) The dynamics has an underlying symmetry. If the driving voltage holds $U_{\text{ext}}(t) = -U_{\text{ext}}(t + T/2)$ with $T$ as its period the dielectric displacement $D$ either satisfies the same symmetry or there are two attractors $A_1$ and $A_2$ and it is valid $D_{A_1}(t + T/2) = -D_{A_2}(t)$ [2], [3].

From feature (i) it can be concluded that the ferroelectric capacitance can be described by a macroscopic model. Therefore the ferroelectric capacitor can be replaced by a substitute–network as it is shown in Figure 3 There $C_{nl}$ represents the dielectric nonlinear properties of the ferroelectric material, $R_p$ stands for the DC–conductivity of the sample and $R_s$ for losses that are caused by the current running through the capacitor. These losses may be due to motion of domain–walls or polarization reversal. Furthermore, the electric field $E_{C_{nl}}$ along the nonlinear capacity $C_{nl}$ must be homogeneous. Note that $R_L$ models the linear losses of the inductivity.

The substitute–network in Figure 3. can be analyzed with Kirchhoff's laws. With $F$ as the Area of the sample, $h$ as its width and $E_{C_{nl}}$ as the electric field over it the result of this analysis is

$$U_{\text{ext}} = LF(1 + \frac{R_s}{R_p})\ddot{D} + \frac{Lh}{R_p}\frac{dE_{C_{nl}}}{dt} + \left[R_L(1 + \frac{R_s}{R_p}) + R_s\right] F\dot{D} + (1 + \frac{R_s}{R_p})hE_{C_{nl}} \,. \quad (1)$$

Eq. (1) can be rewritten in a simplier form because $R_s/R_p << 1$ holds:

$$U_{\text{ext}} = LF\ddot{D} + \frac{Lh}{R_p}\frac{dE_{C_{nl}}}{dt} + [R_L + R_s]\, F\dot{D} + hE_{C_{nl}} \,. \quad (2)$$



The work that is left is to model the electric field in an appropriate way so that Eq. (1) models the circuit in a proper way. Because $E_{C_{nl}}$ models the electric field over the capacitor this issue can be solved by regarding the properties of these materials.

### 2.3. Nonlinear nature of the electric field

The nonlinear properties of the series resonance circuit are determined by the nonlinear dielectric properties of the ferroelectric crystal.

The ferroelectric phase transition of second-order in these materials at the critical temperature $\Theta_C$ can be described by the thermodynamic potential [1]

$$G = G_0 + \frac{\alpha_2}{2}D^2 + \frac{\alpha_4}{4}D_2^4 \,. \quad (3)$$

The basic assumption for the coefficients $\alpha_2$ and $\alpha_4$ are according to Landau's theory $\alpha_2 = \tilde{\alpha}_2(\Theta - \Theta_C)$ and $\alpha_4 = \text{const} > 0$. The coefficient $\tilde{\alpha}_2$ is a positive constant. Note the following three cases are to be distinguished:

Paraelectric material ($\Theta > \Theta_C$): At temperatures above the phase transition the coefficient $\alpha_2$ is positive. The potential has only one minimum and the electric field strength at the sample is calculated as

$$E_{C_{nl}} = \frac{\partial G}{\partial D} = \alpha_2 D + \alpha_4 D^3 \quad \text{with} \quad \alpha_2 > 0 \,. \quad (4)$$

Inserting relation (4) into equation (2) yields a generalized form of the so–called Duffing equation [4].

$$U_{\text{ext}} = LF\ddot{D} + \frac{Lh}{R_p}(\alpha_2 + 3\alpha_4 D^2)\dot{D} + [R_L + R_s]\, F\dot{D} + h(\alpha_2 D + \alpha_4 D^3) \,. \quad (5)$$

Ferroelectric material ($\Theta < \Theta_C$) with small driving voltage: At temperatures below the phase transition the coefficient $\alpha_2$ is negative. The thermodynamic potential becomes double–well. The equilibrium value of the dielectric displacement

$$D_{\text{sp}} = \pm\sqrt{\frac{-\alpha_2}{\alpha_4}} \quad (6)$$

may be derived from the equlibrium condition of the potential. Exciting the series–resonance circuit with small amplitudes $U_0$, the dielectric displacement vibrates around the equilibrium values $D_{\text{sp}}$ of the crystal without an external field. There is no polarization reversal.

Ferroelectric material ($\Theta < \Theta_C$) with large driving voltage: If the amplitude of the driving voltage is increased, polarization reversal in the ferroelectric material may occur. This process is connected with the so–called dielectric hysteresis. The nonlinear relation between electric field strength $E_{C_{nl}}$ and dielectric displacement $D$ can be written as

$$E_{C_{nl}} = \alpha_2 D_2 + \alpha_4 D_2^3 \quad \text{with} \quad \alpha_2 < 0 \,. \qquad (7)$$

At high amplitudes of the driving voltage the oscillator "feels" the whole potential and therefore the relevant differential equation is obtained with expression (7) in expression (2). The result is the generalized Duffing equation in a double–well potential

$$U_{\mathrm{ext}} = LF\ddot{D} + \frac{Lh}{R_p}(-|\alpha_2| + 3\alpha_4 D^2)\dot{D} + [R_L + R_s]F\dot{D} - h|\alpha_2|D + h\alpha_4 D^3 \,. \quad (8)$$

## 3. Results and discussion

The model–equation we derived from a macroscopic view to our system contains a set of free parameters $\{R_s, R_p, \alpha_2, \alpha_4\}$. Determining these four parameters from experimentally recorded time–series is the way to study the phase transition. We reached our goal within three steps:

1 Periodic phase portraits of the series–resonance circuit have been recorded at different temperatures above and below the phase transition.

2 By a method we developed recently [5] a set of modelling parameters is determined numerically from the recorded time–series.

3 The equations (5) and (8) are solved by numerical means. If for the set of values of the modelling parameters can be observed a simulated phase portrait that is very similar to the experimentally recorded one then this set of parameters will be accepted as valid for describing the system.

To make the term "very similar" more objective we applied two criteria that had to be fullfilled for stating the success of our approximation:

(i) The experimental phase portrait and the simulated one must be bounded by the same values for $D$ and $\dot{D}$.

(ii) Both curves must have the same internal structure.

In practice that simulating method is quite effective. It can be carried out at a personal computer and the time of calculating the model–parameters from the experimental time–series is approximately equal to the time that is required to measure it (including all IEEE–bus–transfers).

An example of the quality of the result we get is shown in Fig. 4.We analyzed time–series before and after a 1T–2T–bifurcation for betainarsenate [6] as ferroelectric



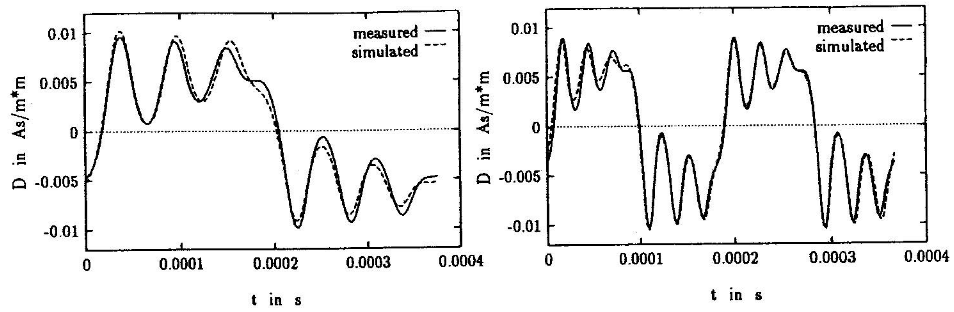


Fig. 4. Comparison between measured and simulated time–series for betainarsenate before (left) and after (right) a 1T–2T–bifurcation.

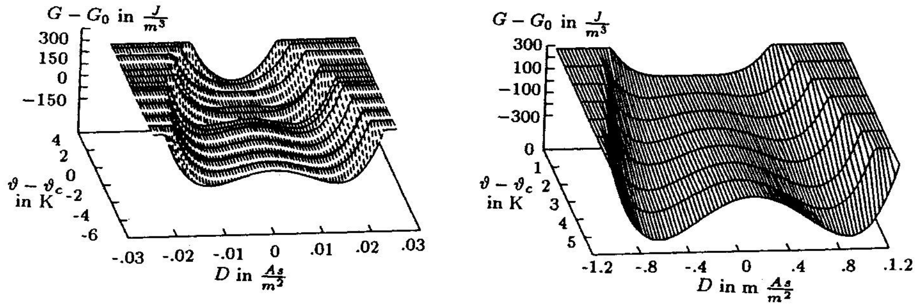


Fig. 5. Effective thermodynamic potential of a betainarsenate–crystal (left) and a ferroelectric liquid crystal (right).

material. Note, that the simulated and experimentally recorded time–series show the same qualitative properties so that the result is accepted.

In the next step the coefficients $\alpha_2$ and $\alpha_4$ may be added up according to Eq. (3). This is done and shown in Figure 5. for two different materials: Betainearsenate and a ferroelectric liquid crystal. The thermodynamic potential $G$ behaves for both materials as expected in Sec. 2.3. Note, that the energy that is represented by the potential of the ferroelectric liquid crystal is about 10 times greater than the those of betainarsenate.

We want to stress that it is not necessary to restrict to a linear ansatz of $R_s$ and $R_p$ as it was done for Eq. (1). Higher terms may be used, but then it must obeyed that the symmetry–condition (ii) mentioned in Sec. 2.2. holds for that ansatz.

## 4. Controlling the dielectric nonlinear series–resonance circuit

### 4.1 Controlling method

We want to use the method proposed by Ott, Grebogy and Yorke (OGY) [7]. The advantages of this method are that all required parameters of control can be got only by measuring of the system without using any kind of model and only very small perturbations are necessary for reaching a successful control.

This method uses the specific properties of the unstable periodic saddle orbits embedded in the chaotic attractor. The basic idea of the control method is to change a parameter (which can be varied from outside of the system) in a small range and to force the system to the stable direction of one of these saddle points.

The value of this perturbation should be selected in such way, that the system approaches that orbit in the next time if it is found next of this unstable periodic orbit. The system meets the requirements, if it depends on a parameter, which is variable in a small interval and we have an iterative map (Poincaré–section) with unstable periodic orbits. The formula introduced by OGY is the following equation:

$$\partial a = -\frac{\lambda_u}{\vec{f_u} \cdot \vec{w}} \vec{f_u} \cdot \partial \vec{\xi_n} \, . \qquad (9)$$

Here the vector $\vec{f_u}$ is the unstable contravariant basis vector and it gives the direction of the unstable manifold of the fixed point. The parameter $\lambda_u$ is the corresponding eigenvalue, the vector $\vec{w}$ comes from the linearisation of the Poincaré–map and $\partial\vec{\xi_n}$ is the difference between the fixed point $\vec{\xi_F}$ and the momentary system state $\vec{\xi_n}$.

The dielectric nonlinear series–resonance circuit has a 2-dimensional Poincaré–section $\{D, j\}$, so Eq. (9) can be rewritten as

$$\partial a = k_1 - (k_2 D + k_3 j) \, . \qquad (10)$$

This equation shows, that the formula confines to two multiplications and two additions/subtractions, if the parameters are adequate summarized. We chose the driving amplitude of the series resonance circuit as an external parameter of perturbation.

In practice we apply Eq. (10) to the dielectric nonlinear series–resonance circuit by an analog computer that we designed for that purpose. The block–diagram of this analog–computer is shown in Figure 6.

One A/D–converter is used for recording the Poincaré–section. The controlling computer, that is in fact a 386–PC, determines then the fixed point $\vec{\xi_F}$ and the values of $k_1$, $k_2$ and $k_3$. The other A/D–converter then converts the numerical values of these parameters into small voltages. Two multipliers and one adder perform then the calculation of Eq. (10). This process is performed continously, however, the value of the control–parameter is only required in the very moment of obtaining a point in the Poincaré–section. That is why we need a sample–and–hold–unit, triggered by the driving generator.

We require a very small parameter for the perturbation. Unfortunately, often it is the case that this parameter is not small enough. Especially, this case will occur if the state of the system is far from the fixed point $\vec{x}_F$. Therefore a window–discriminator is necessary. Its output controls the amplifier in order to modulate the driving voltage of the dielectric nonlinear series–resonance circuit. No modulation will take place if the perturbation is set to zero or too big.



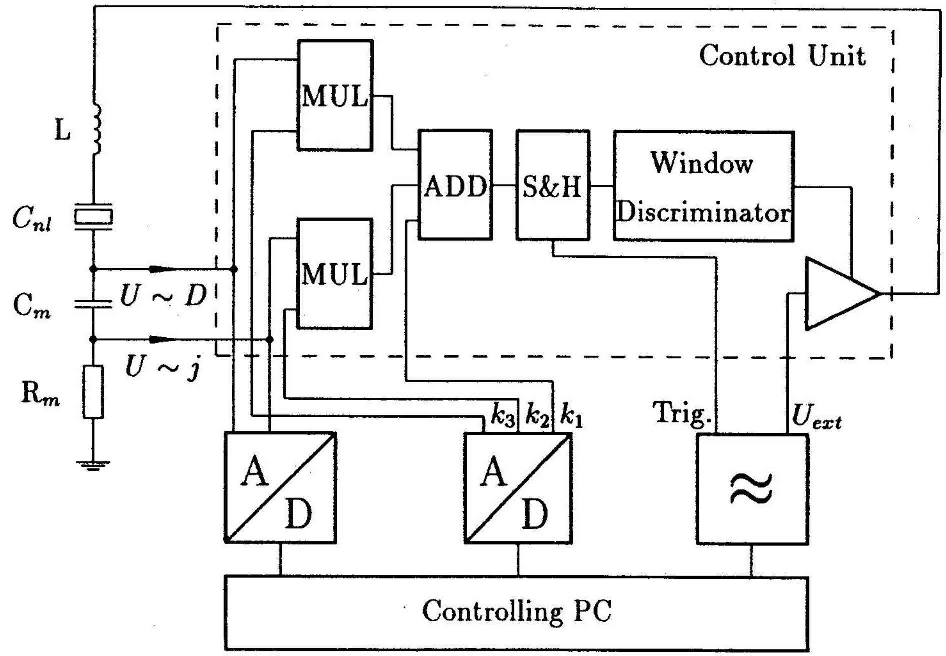


Fig. 6. Block–diagramm of the analogue–computer that controls the dielectric nonlinear series–resonance circuit.

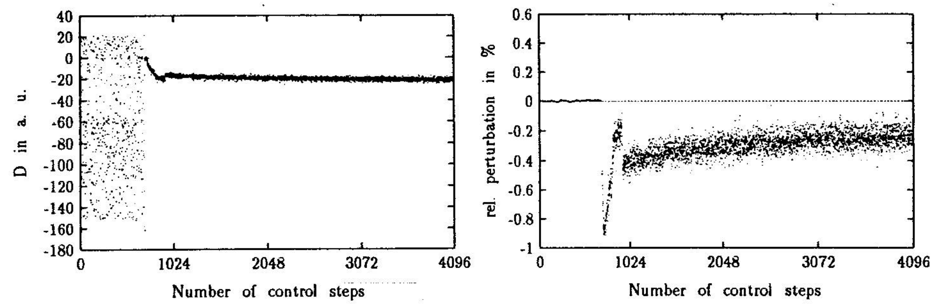


Fig. 7.Results of a control–process. Note, that the control is switched on after 750 steps.

## 4.2 Results of controlling the dielectric nonlinear series–resonance circuit

The results of the control are shown in Figure 7. There are shown the control perturbation in percent of driving amplitude and the dielectric displacement vs. the control steps. In the left hand side the control is switched off and then is switched on. One can see, that the perturbation is smaller than 1% for a successful chaos control.

In the first time after the control is switched on, the perturbation is large — the system is forced to a stable orbit. A smaller perturbation is required to hold the system in a periodic window, if this orbit is reached.

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