Condensed Matter Physics, 2003, Vol. 6, No. 2(34), pp. 189–196

What can we learn about ferroelectrics using methods of nonlinear dynamics?

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Received September 3, 2002

The nonlinear series resonance circuit with ferroelectric capacitor has been extensively investigated. If the ferroelectric within the capacitor is in its polar phase many of the features well known from model systems of nonlinear dynamics may be observed. These characteristics are the shift of resonance frequency with increasing driving voltage, bifurcations and chaotic behaviour. Considerations of the system on the basis of simple Landau-theory suggest to describe the resonance circuit by means of a Duffing-equation. Because of the switching process during the nonlinear vibrations the real situation is more complicated as can be shown by nonlinear time series analysis [1].

Key words: *ferroelectricity, domains, TGS, nonlinear dynamics, Duffing-equation, chaos*

PACS: *05.45.a, 05.45.Tp, 77.80.-e, 77.84.Fa*

1. Introduction

Switching processes in ferroelectrics are caused by electric fields of magnitudes above the coercive field strength E_{C}. One of the characteristics of switching is the occurrence of hysteresis-loops which can be observed below the so-called Curie-temperature T_{C}. They are accompanied by large dielectric nonlinearities [2]. The description of these switching processes is one of the problems to be solved in the physics of ferroelectricity and in the other fields of condensed matter physics. On the other hand, great efforts have been made during the recent years to analyse nonlinear dynamic systems [3]. So, the motivation of our work is twofold. Using ferroelectric materials like TGS in its polar phase as a capacitor in a series resonance circuit we have a nonlinear dynamic system that makes it possible to test the predictions of the results of the theory of nonlinear dynamics, e.g. bifurcations and chaotic behaviour. Another problem is the very switching process of the ferroelectrics underlying these bifurcations. Therefore, we are interested in answering the question whether it is possible to draw some conclusions concerning the ferroelectric switching by analysing the experimentally recorded time series of our nonlinear resonance circuit.

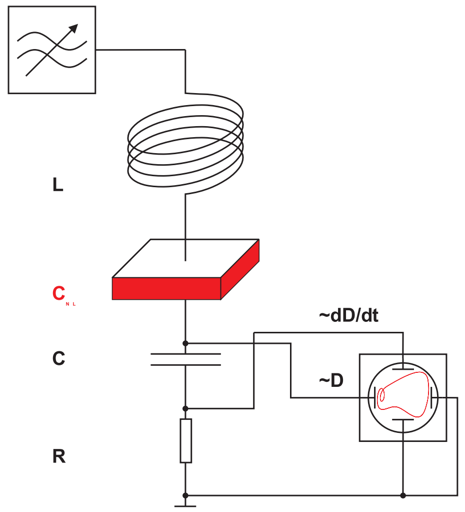

Figure 1. The nonlinear series resonance circuit with ferroelectric capacitor.

2. Experimental setup

The nonlinear series resonance circuit is shown in figure 1. It consists of a linear inductance L, and a nonlinear capacitance C_{NL}, which is a ferroelectric crystal. In our special case we used triglycine sulfate (TGS) as ferroelectric. The driving voltage $U_{\mathrm{ext}} = U_0 \cos(\omega t + \phi)$ may be varied both in amplitude and frequency to force the circuit in order to exhibit bifurcations and chaotic behaviour.

The linear capacitor C and the resistor R have been added to derive signals proportional to the charge of the TGS-capacitor and the current of the circuit, respectively. To minimize the influence of these two elements it is necessary to fulfill the condition $R \ll 1/C \ll 1/C_{\mathrm{NL}}$ which reminds of the Sawyer-Tower circuit. So, two signals $U_{\mathrm{C}} \sim D$ and $U_{\mathrm{R}} \sim \dot{D} = j$ may be derived [1,4], where D and j are the dielectric displacement and the current density, respectively. These signals are recorded by a digital storage oscilloscope and can be used for direct observation of the phase portrait as demonstrated in figure 2. Alternatively, it is possible to record only the signal U_{C} and to calculate the temporal derivatives numerically [1,5].

According to Kirchhoff's laws and neglecting the small voltages U_{C} and U_{R}, the behaviour of the circuit may be written by the following equation:

$$U_{\mathrm{ext}} = U_{\mathrm{L}} + U_{\mathrm{R_V}} + U_{\mathrm{NL}}\,, \tag{1}$$

where U_{L} and U_{NL} are the voltage drops in the inductance and the ferroelectric, respectively, and $U_{\mathrm{R_V}}$ is a voltage drop both across the ferroelectric and the inductance. From equation (1) one gets a second order differential equation [1,5,6], that is the equation of motion in the sense of the theory of dynamic systems:

$$\ddot{D} = -\frac{h}{LS} E_{\mathrm{NL}}(D, \dot{D}) - \frac{R_{\mathrm{V}}}{L} \dot{D} + \frac{U_{\mathrm{ext}}(t)}{LS}. \tag{2}$$

Here, $D, \dot{D}, \ddot{D}$ are the dielectric displacement and its temporal derivatives, E_{NL} is the electric field strength in the ferroelectric which is assumed to depend on both D

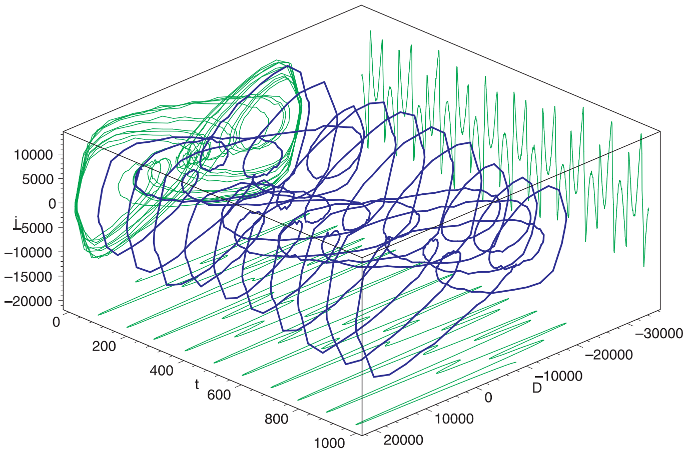

Figure 2. Experimentally observed phase portrait constructed from the two time series $D(t)$ and $j(t)$ (both in arbitrary units).

and $\dot{D}$ in general case. S and h are the electroded surface and the thickness of the ferroelectric sample, respectively.

For a further analysis, time series of D and $\dot{D}$ were recorded at different temperatures of the ferroelectric (see figure 3). The ferroelectric was a rectangular TGS sample with thickness $h = 0.28$ mm and the electroded surface $S = 7.31 \times 4.66\ \mathrm{mm}^2$. The other parameters were as follows: inductance $L = 100$ mH, amplitude of the sinusoidal driving voltage $U_0 = 15.5$ V and frequency $f = 2369$ Hz. The temperatures were $T_a = 311.65$ K, $T_b = 308.67$ K and $T_c = 306.67$ K.

3. Analysis of the experimental data

As mentioned above, this relatively simple circuit is interesting both from the viewpoint of nonlinear dynamics and ferroelectricity. In our case we can combine these fields and hope to get additional information for the characterization of the ferroelectric switching process using the methods proposed in order to extract the parameters of the equation of motion. For a review of such methods see [3] and references therein.

As is well known, many properties of ferroelectrics in its polar phases are dominated by the effect of the domain structure (see e.g. [2,7,8]). Nevertheless, already the simple Landau-theory of second order phase transitions motivates the consideration of the resonance circuit with ferroelectric TGS-capacitor as an experimental realization of a nonlinear dynamic system.

Using the relation $E_{\mathrm{NL}} = \alpha_0(T - T_C)D + \gamma D^3$ equation (2) yields directly:

$$\ddot{D} = -\frac{h}{LS}(\alpha_0(T - T_{\mathrm{C}})D + \gamma D^3) - \frac{R_{\mathrm{V}}}{L}\dot{D} + \frac{U_{\mathrm{ext}}(t)}{LS}. \tag{3}$$

This equation is the so-called Duffing-equation which is one of the model equations of nonlinear dynamics [9,10]. An interesting feature of equation (3) is that it represents a single-well oscillator above T_{C} and a double-well oscillator if the temperature of the

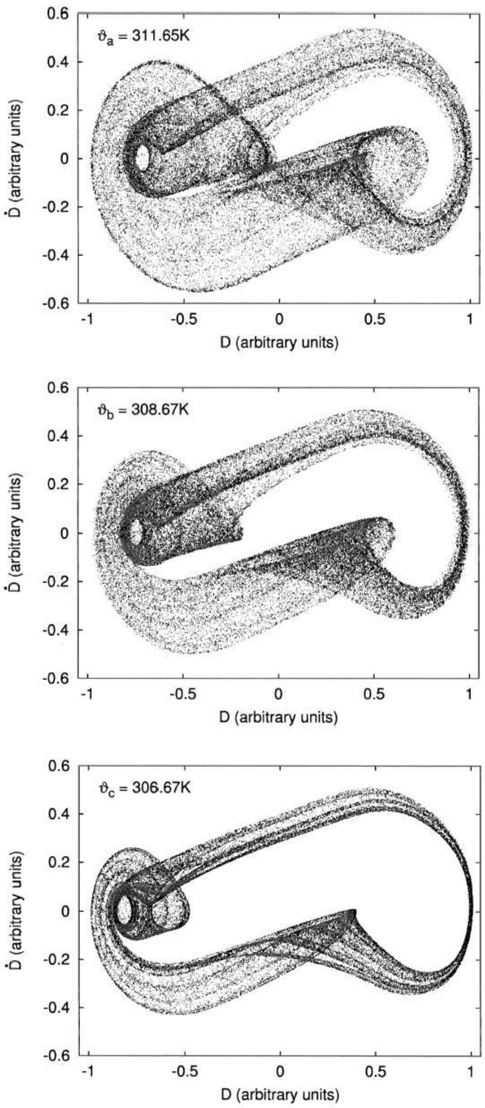

Figure 3. Three phase portraits which were recorded experimentally at different temperatures of the TGS-crystal [5].

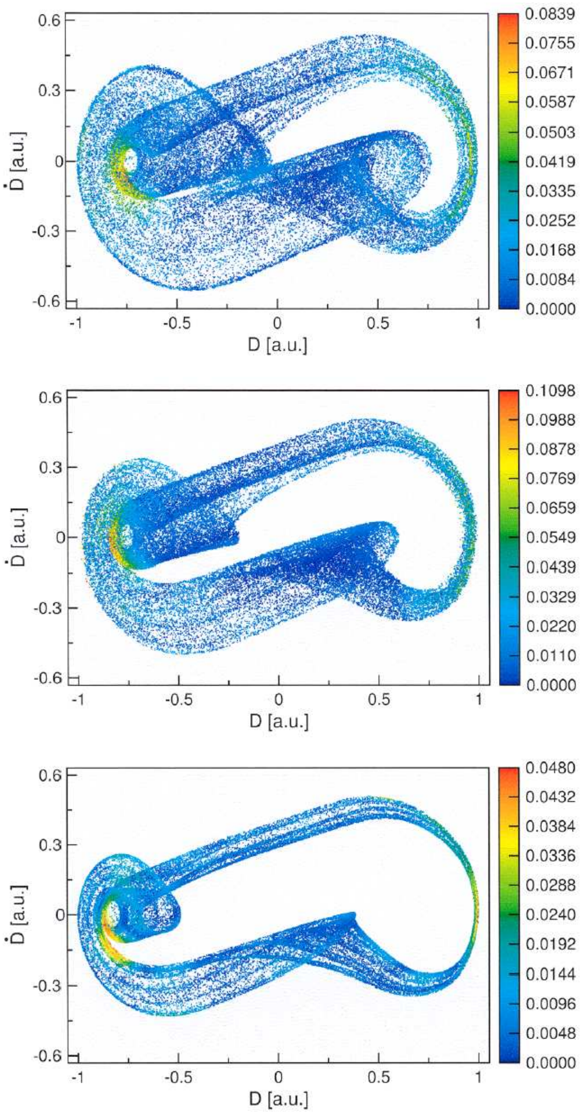

Figure 4. One-step prediction errors χ_n for the investigated phase portraits [5].

ferroelectric is below T_C. A wide variety of theoretical results for that model could be confirmed experimentally in our circuit. So, it was possible to measure the shift of resonance frequency with the increasing amplitude of the driving voltage [11] as well as period-doubling bifurcations and chaotic behaviour by tuning the parameters of the circuit such as temperature of the ferroelectric, amplitude and frequency of the driving voltage [4,6]. But more detailed investigations revealed that the situation is more complicated [12]. So, it is not possible to describe the behaviour of the circuit with TGS in its ferroelectric phase by parameters α_0 and γ determined from the shift of resonance frequency at temperatures of the same TGS capacitor above T_C. The reason is that linear and nonlinear dielectric properties in the paraelectric phase are not affected by domain switching processes. This is a confirmation of the well-known fact that Landau-theory is not capable of describing the switching [2].

The task is now to modify the model such that it becomes capable of characterizing the nonlinear resonance circuit in the polar region of the ferroelectric. Therefore, we tested the possibilities of time series analysis of chaotic signals from the viewpoint of an experimentalist which may be a little questionable at first glance. But as we have checked even in the regime of chaotic vibrations, the behaviour of the circuit is reproducible. It means that bifurcations, chaos and periodic windows [13] occur at the same control parameters (temperature of the ferroelectric, amplitude and frequency of the driving voltage) even when the ferroelectric is heated above the phase-transition temperature and cooled back to the initial temperature. A first attempt was made to get a better agreement between the experimental data and a model equation of extended Duffing-type:

$$\ddot{D} = \underbrace{\sum_{i=0}^{n} a_i D^i - \frac{R_\mathrm{V}}{L}\dot{D}}_{F(D,\dot{D})} + \frac{U_\mathrm{ext}(t)}{LS}, \tag{4}$$

where the degree of the polynomial in D was increased up to $n = 9$ [1].

What follows is a short description of the fitting procedure. For a detailed outline see [1,5]. Having recorded a long time series of $U_C \sim D$ (129907 data points D_n per time series in our experiments) it is possible to estimate the temporal derivatives $\dot{D}_n$ and $\ddot{D}_n$ from adjacent data points of the time series D_n according to the following formulas:

$$\begin{aligned}\dot{D}_n &= \frac{1}{60\tau}(D_{n+3} - 9D_{n+2} + 45D_{n+1} \\ &\quad -45D_{n-1} + 9D_{n-2} - D_{n-3}) + O(\tau^5), \\ \ddot{D}_n &= \frac{1}{180\tau^2}(2D_{n+3} - 27D_{n+2} + 270D_{n+1} \\ &\quad -490D_n + 270D_{n-1} - 27D_{n-2} + 2D_{n-3}) + O(\tau^5).\end{aligned} \tag{5}$$

Having calculated the derivatives for each point of time $t_n = n\tau$ we have quadruples $(D_n, \dot{D}_n, \ddot{D}_n, n)$ which are used to determine the parameters in the ansatz equation (4) by a least-squares fit which minimizes:

$$\chi^2 = \sum_{n=1}^{N} \left(\ddot{D}_n - F(D_n, \dot{D}_n) - \frac{U_{\mathrm{ext}}(n\tau)}{LS} \right)^2 = \sum_{n=1}^{N} \chi_n^2 \,. \tag{6}$$

The ansatz equation (3) yielded a relative error of the fit ϵ_N of 30%–27% for $N = 5-9$, where ϵ_N is calculated as

$$\epsilon_N = \frac{100\sqrt{\chi^2}}{\sqrt{\sum_{n=1}^{N} \ddot{D}_n^2}}. \tag{7}$$

A further improvement of the fit was achieved with a more general ansatz for the function $F(D, \dot{D})$ (see underbrace in equation (4)). Taking into account possible couplings between D and $\dot{D}$ which may be physically motivated by various attempts of modelling the nucleation processes [2] we made an ansatz of a complete bivariate polynomial with the terms up to the power k in D and l in $\dot{D}$, including all mixed terms:

$$F(D, \dot{D}) = \sum_{i=0}^{k} \sum_{j=0}^{l} \alpha_{ij} D^i \dot{D}^j. \tag{8}$$

It could be shown ([5]) that the relative error ϵ_N saturates at approximately 8.5% for $k > 12$ and $l > 5$. A further increase of the degree of the polynomial $F(D, \dot{D})$ to $k = 15$ and $l = 7$ gave no significant reduction of the error. But the model contains a lot of parameters (128 α_{ij} for the basis functions $D^i\dot{D}^j$ plus 2 parameters for amplitude and phase of the driving voltage U_{ext}). So, the question arises whether it is possible to reduce the number of coefficients without significantly increasing the relative error ϵ_Nv omitting the terms which give only a small contribution to the model. The first step towards such a simplified model results from the knowledge of the symmetry of the system. From the symmetry of the driving voltage $U_{\mathrm{ext}}(t) = -U_{\mathrm{ext}}(t + T/2)$, where T is the period of the driving, it can be concluded that the attractor in the phase space $\mathbf{X} = (D, \dot{D})$ has the same symmetry or there exist two attractors related to each other by the relation $\mathbf{X}_{A_1}(t) = -\mathbf{X}_{A_2}(t + T/2)$. Taking into account this symmetry we can neglect all the basis functions $D^i\dot{D}^j$ where $i + j$ equals an even number except α_{00} to describe an asymmetry (e.g. inner field) in the ferroelectric. The result of this reduction was a small increase of the relative errors $\epsilon_{N_{\mathrm{odd}}} \approx \epsilon_{N_{\mathrm{all}}} + 0.5\%$ for the three different cases. Further reduction of the number of the remaining 65 α_{ij} was achieved by a modified backward elimination algorithm (for details see [5]). Nevertheless, it was necessary to keep about 20–30 basis functions to avoid a dramatic increase of the error. As a result of the procedure, we present the numerically calculated phase portraits for the "optimal" models for the three experimental time series (figure 4). Obviously, the main features of the experimental data are correctly reproduced. Additionally, figure 4 contains the absolute values of the one-step prediction errors χ_n (see equation (6)) in color coding over the phase portrait. It is not surprising that the largest errors occur at the normalised values $|D| > 0.5$, that is in the vicinity of the values of the spontaneous polarization, where nucleation processes cannot be neglected.

The most surprising result was the fact that we can extract a nonlinear relation $E_{\mathrm{NL}}(D, \dot{D})$ for the ferroelectic TGS from our model with no significant indication of a double-well potential [5]. Nevertheless, we showed that it is possible to fit even chaotic time series of the nonlinear resonance circuit with high accuracy with only few assumptions concerning the ferroelectric. Further steps towards an improvement of the model will follow including more physically motivated models of ferroelectric switching processes.

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Що можна дізнатися про сегнетоелектрики методами нелінійної динаміки?

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Отримано 3 вересня 2002 р.

Об'єктом дослідження у даній роботі є нелінійний послідовний резонансний контур з сегнетоелекричним конденсатором. Якщо сегнетоелектрик у конденсаторі знаходиться у полярному стані, спостерігаються властивості, відомі для модельних систем у нелінійній динаміці. Це, зокрема, зсув резонансної частоти зі збільшенням прикладеної напруги, біфуркації та хаотична поведінка. Розгляд системи в рамках простої теорії Ландау веде до опису резонансного контура за допомогою рівняння Дуффінга. Насправді ж, як показано з допомогою аналізу нелінійних часових послідовностей [1], процеси перемикання підчас нелінійних коливань значно ускладнюють ситуацію.

Ключові слова: *сегнетоелектрики, домени, ТГС, нелінійна динаміка, рівняння Дуффінга, хаос*

PACS: *05.45.a, 05.45.Tp, 77.80.-e, 77.84.Fa*