Period Doubling of a Torus near the Ferroelectric Phase Transition of a KH_2PO_4 Crystal

Jong Cheol Shin*
Research Institute for Basic Sciences, Seoul National University, Seoul 151-742, Korea

Sook-Il Kwun
Department of Physics, Seoul National University, Seoul 151-742, Korea
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We have studied the transition to chaos in the ferroelectric KH_2PO_4 crystal near the phase transition temperature. An unusual scenario has been found: the transition from a fixed point to a quasiperiodic motion on a torus (T) through Hopf bifurcation to a quasiperiodic motion on a two-torus (T^2) through a cascade of torus doublings, and then directly to chaos. These results point to the essential role of the polarization fluctuations near the phase transition and the electromechanical couplings between the soft-mode polarization P and the elastic shear strain X of the crystal, whose dynamics are described by a Duffing's anharmonic oscillator coupled by a piezoelectric oscillator. [S0031-9007(99)08532-4]

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Recent studies on nonlinear dynamics and chaos have shown that many dissipative nonlinear dynamical systems, though the governing equations of motion for the systems are very simple and deterministic, can produce an apparently pseudorandom behavior, which is called chaos. One of the most interesting results of nonlinear dynamical systems theory is that these random or chaotic behaviors can be reached by following one or another of the known scenarios, regardless of the details of the dynamical systems concerned, which leads to the concept of universality in dynamical systems theory [1]. Period doubling, quasiperiodicity, and intermittency are three representative examples.

Another interesting but less widely known scenario is the cascade of torus doublings. In this scenario the torus, represented by a circular ring in phase space, is separated into two circular rings, followed by the emergence of a strange attractor. This scenario was first studied extensively in theory by Kaneko [2]. He combined two kinds of maps belonging to the two different universality classes, viz., period doubling and quasiperiodicity, associated with the frequency locking, and found a new scenario, i.e., torus doubling, as a result of the interaction between the two scenarios. He also found that, unlike with the infinite number of period doubling from a fixed point, the torus undergoes a finite number of doublings.

The cascade of torus-doubling transition to chaos is similar to the period-doubling transition to chaos. But it differs in the following two significant ways: (i) A cascade of torus doublings occurs in high-dimensional dynamical systems. It requires at least four dimensions in a continuous flow or three dimensions in a discrete map. Unlike the period-doubling transition to chaos, it does not occur in a low-dimensional map or continuous flow. (ii) Instead of an infinite number of period doublings toward the accumulation point, only a finite number of torus doublings have been observed before the onset of chaos. In fact, no more than two successive torus-doubling events have ever been observed in experiments. Compared with the Ruelle and Takens conjecture [3] that a torus in low-dimensional dynamical systems undergoes a transition into chaos through quasiperiodicity, a torus in high-dimensional flow loses its stability and develops into chaos through period doubling.

The torus-doubling transition to chaos has been reported in high-dimensional dynamical systems: in a numerical study of seven-mode Navier-Stokes equations [4], in a three-dimensional map with dissipation by Arneodo *et al.* [5], and in a quintic complex Ginzburg-Landau equation describing the dynamics of plane Poiseuille flow [6]. This scenario has also been reported in recent experiments on fluid convection [7] and chemical reactions [8], all of which are infinite-dimensional dynamical systems. But those few works reporting on the torus-doubling scenario in condensed matter physics are given in Ref. [9].

In this paper, we report on an experimental observation of a torus-doubling transition to chaos, i.e., the transition from a fixed point to a quasiperiodic motion on a torus (via Hopf bifurcation), to a motion on a two torus (via torus doubling), and then to chaos, as suggested by Kaneko [2], in a KH_2PO_4 (KDP) crystal near the ferroelectric phase transition temperature. It should be stressed that this is the first experimental report on the torus-doubling transition to chaos occurring in condensed matter, and KDP is a good candidate to study this scenario. The dynamical behavior of the KDP crystal near the phase transition is described by the coupled five-dimensional dynamical systems; a Duffings' anharmonic oscillator, driven by an external stimulus, is coupled to the elastic shear-mode oscillator by piezoelectricity. As a result, a KDP crystal near the phase transition is expected to give a large variety of nonlinear dynamical behaviors including period doubling, quasiperiodicity, and torus doubling as a result of the interaction between them.

Until now, nonlinear dynamical behavior, especially in the ferroelectric materials, has been reported in the following three important situations:

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(a) *Near the transition temperature T_c.*—Fluctuation in the order parameter grows significantly as the temperature approaches the phase transition and the instability associated with the fluctuation is also greatly increased. As a result, nonlinearity of the crystal can be induced. A Rochelle salt was reported to show a period doubling and chaos when it was moderately biased near T_c [10], and this was the first report on chaotic phenomena in ferroelectric materials.

(b) *By mode couplings.*—In the ferroelectric KH_2PO_4, there exists an electromechanical coupling between a polarization mode along the ferroelectric c axis and an elastic shear mode along the diagonal direction in the ab plane near T_c. Period doubling, frequency locking associated with the quasiperiodicity, and intermittency have been reported [11]. Regarding the coupling between the two nonlinear mode oscillators, torus-doubling transition to chaos, which will be the main concern of this paper, is naturally expected in the equation of motion but it has not been experimentally reported so far.

(c) *By instabilities in the crystal.*—The mode-locking structure of the oscillatory and chaotic states in the electrical conduction has been reported for barium sodium niobate (BSN) crystal, which is in good agreement with the theoretical circle map [12]. These examples in ferroelectric materials show that the study of the nonlinear dynamical behavior of condensed matter gives much information on the correlation between the generation of nonlinearity and the order-parameter dynamics near the phase transition temperature.

For our experiment, KDP samples were prepared in two stages. The KDP was cut along the c axis to get a thin slab and then cut again along the 45° diagonal in the ab plane to couple the piezoelectric excitation to the c-axis polarization. Typical dimensions of the samples were $13 \times 3 \times 0.5\ \mathrm{mm}^3$. To make electrodes on the sample, it was evaporated with gold and then painted with silver paste. With the sample geometry prepared in this way, shear strain X_6 is coupled piezoelectrically to the polarization P along the c axis so it simply dilates along the length. The KDP samples prepared were used as a nonlinear capacitor in the simple RLC resonator (Fig. 1) with $L = 10$ mH and $R = 390\ \Omega$.

We used cryogenic equipment (e.g., a closed cycle helium refrigerator and a cryostat), provided in a package by the JANIS Research Co., Inc. We also used the Lakeshore 330 autotuning temperature controller whose control stability is given by ± 25 mK at 300 K with silicone diode sensor. With the cryostat set at $\sim$3–5 mTorr, the temperature of the KDP sample was controlled to within ± 0.01 K around T_c. As a signal generator, we used the HP3325B function generator whose resolution is given by 1 μHz at $f \le 100$ kHz from 20 to 30 °C. To characterize the sample, we performed the dielectric measurements first. An HP4275 multifrequency spectrum analyzer was used at $f = 10$ kHz and $T = \sim$150–50 K in steps of 1 K. We then carried out the same experiments again in steps

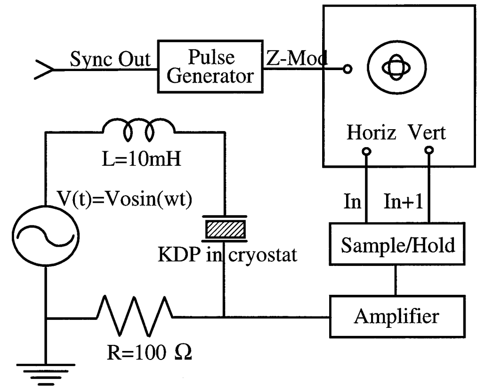

FIG. 1. Schematic of the apparatus used in the experiment to generate the Poincaré section directly on the oscilloscope.

of 0.02 K near T_c to determine exactly the phase transition temperature of the KDP crystal. T_c was found to be 120.98 K and the voltage signal across the resistor was measured as a dynamical variable.

Now let us suppose that the KDP crystal is driven by a sinusoidal voltage $V(t) = V_0 \sin(2\pi f t)$ in a simple RLC circuit. Then the equation of motion for P near the phase transition temperature is described by a Duffing's nonlinear oscillator equation [11]. A lot of numerical studies have been devoted to the equation and it is now one of the representative examples of a nonlinear dynamical system showing a finite number of period doublings [13].

KDP crystal also has a piezoelectricity near the phase transition temperature between the soft-mode polarization P and the elastic shear deformation X. Regarding bilinear coupling $\eta P X$ with η the coupling strength in the interaction potential, which is the simplest coupling form, the following equation of motion has been suggested [11]:

$$\begin{aligned} &\ddot{P} + a\dot{P} - P + P^3 + \eta X = \nu \sin(\Omega \tau), \\ &\ddot{X} + b\dot{X} - X + X^3 + \eta P = 0, \end{aligned} \tag{1}$$

where τ is the dimensionless time, a and b are the damping constant related to the polarization fluctuation and shear strain, Ω is the dimensionless frequency, and the differentiation is with respect to τ. According to Eq. (1), dynamical behavior of KDP crystal near the phase transition is described by two anharmonic oscillators coupled by an electromechanical coupling, each of which is a representative example of period doubling and quasiperiodicity. When it is written as a set of first-order equations in the following form,

$$\begin{aligned} &\frac{dP}{d\tau} = Q, \quad \frac{dQ}{d\tau} = -aQ + P - P^3 - \eta X + \nu \sin\phi, \\ &\frac{dX}{d\tau} = Y, \quad \frac{dY}{d\tau} = bY + X - X^3 - \eta P, \\ &\frac{d\phi}{d\tau} = \Omega, \end{aligned} \tag{2}$$

to analyze the dynamical behaviors, it becomes the five-dimensional dynamical system in continuous phase flow and thus satisfies the necessary condition for the torus-doubling transition leading to chaos.

Four-dimensional Poincaré maps of the type,

$$\begin{aligned} P_{n+1} &= f_1(P_n, Q_n, X_n, Y_n)\,, \\ Q_{n+1} &= f_2(P_n, Q_n, X_n, Y_n)\,, \\ X_{n+1} &= f_3(P_n, Q_n, X_n, Y_n)\,, \\ Y_{n+1} &= f_4(P_n, Q_n, X_n, Y_n)\,, \end{aligned} \qquad (3)$$

which will be a combination of maps showing period doubling and quasiperiodicity, can be constructed by a Poincaré section technique. According to Eqs. (2) and (3), the KDP crystal near the phase transition can be regarded as a coupled dynamical system which belongs to the two different universality classes: period doubling and quasiperiodicity. Thus a KDP crystal is a good example of a nonlinear dynamical system in condensed matter to study the interaction between period doubling and quasiperiodicity. The mappings given by Eq. (3) can sometimes be simplified into low-dimensional maps showing period doubling or quasiperiodicity when one of the variables can be written as a function of the other three variables after the initial transients have died away. It should also be pointed out that, even though an exact analytic form of the equation is not available, maps obtained in experiments can be useful in illustrating the bifurcation nature of the dynamical system concerned. In the experiment, we have observed period doubling of a fixed point, quasiperiodicity, and period doubling of a torus, as will be discussed below.

To see the torus-doubling phenomena more clearly, we set $T = T_c + 0.2$ K and then varied T to see the temperature dependence of the phenomena. Too intense turbulent fluctuations of the order parameter P near T_c may degrade the fine structure of the phenomena, i.e., the torus develops directly into chaos without period doubling. Stabilizing the temperature, we scanned the frequency of the signal generator linearly from 100 to 30 kHz in automatic mode in 1000 sec. From this frequency scan, we could find the frequency interval displaying chaos. In that frequency region, we scanned the frequency again manually in steps of 1 Hz. At this scan rate we assumed a quasistatic response of the crystal.

We observed that a fixed point evolves into a quasiperiodic motion on a torus (T) through a Hopf bifurcation at $f = 62.534$ kHz. The second self-oscillation in the crystal is generated by a piezoelectric oscillation provided by an electromechanical coupling between the order parameter P and the shear strain X as given by Eqs. (1) and (2). A Poincaré map, a stroboscopic motion of the trajectory on a section plane in the phase space, is a common way of displaying the dynamics of quasiperiodic motion and, experimentally, this is generated by a series connection of a sample and hold circuit, which holds the maximum of the signal with one period delayed in succession. Additionally a pulse generator is also used to trigger the signal at the peak position. It consists of a differentiator and a zero-crossing detector. These pulses are used to strobe the Z axis of the oscilloscope to display the Poincaré section of the phase plots on the cathode ray tube clearly (Fig. 1). On the Poincaré section, a two-frequency quasiperiodic motion in the continuous flow is represented by a circular ring on the Poincaré section plane, and the result is shown in Fig. 2(a).

The transition from a motion on a torus to a motion on a two-torus occurs at $f = 61.096$ kHz through a period doubling. Figure 2(b) shows the period-doubled torus. A smooth circular curve in Fig. 2(a), showing that the motion is in a quasiperiodic state, is separated into two smooth circles, each of which is visited alternately by

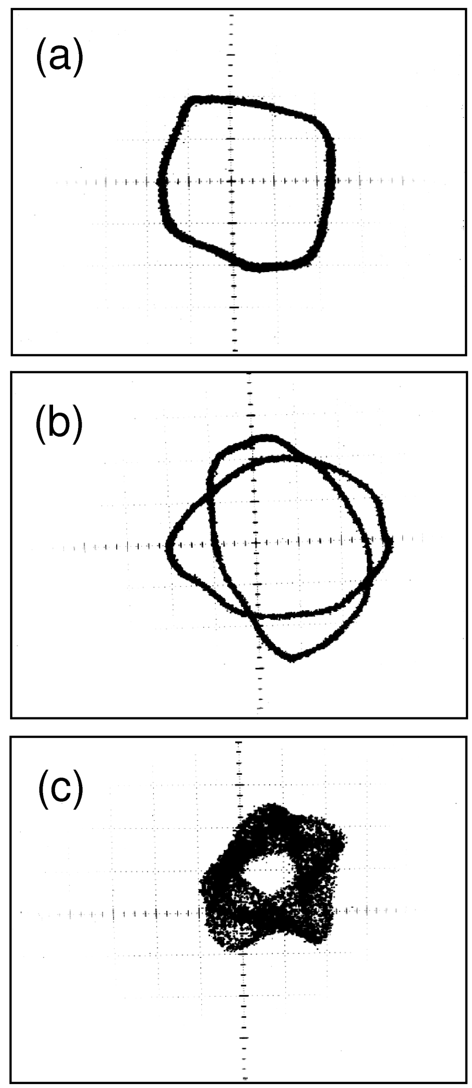

FIG. 2. Return map observed at $T = 121.18$ K and $V_0 = 10$ V. Scale of the horizontal and the vertical axis is the same, 0.2 V/div, with the gain of 10 dB. (a) A quasiperiodic motion on a torus ($f = 62.092$ kHz); (b) a quasiperiodic motion on a two-torus ($f = 61.074$ kHz); (c) a chaotic motion ($f = 60.404$ kHz).

the trajectory. After a period doubling of the torus, chaos appears suddenly at $f = 61.054$ kHz. The two-torus starts to merge to form a chaotic attractor and the result is shown in Fig. 2(c). The whole sequence in the figure means that a fixed point undergoes a transition to a quasiperiodic motion on a torus (T) through the Hopf bifurcation, to a quasiperiodic motion on a two-torus (T^2) through the period-doubling bifurcation, and then to a chaos. This is in sharp contrast with the period doubling of a fixed point in a low-dimensional dynamical system, which requires an infinite number of period-doubling bifurcations toward the onset of chaos. It should also be stressed that a torus in a high-dimensional dynamical system ($d \geq 3$) can lose its stability and develop into chaos by a finite number of period doublings, as predicted in theory by Kaneko [2]. On the other hand, a torus in a low-dimensional dynamical system ($d < 3$) develops into chaos by the quasiperiodicity associated with frequency locking as suggested by Ruelle and Takens [3].

In our investigation, torus-doubling phenomena have been observed within ± 1 K around T_c while period-doubling and intermittent routes to chaos are also observed ubiquitously. In particular, much richer nonlinear dynamical responses including quasiperiodicity, frequency locking, and a cascade of torus-doubling transition to chaos are found near the phase transition temperature T_c. More details will be reported elsewhere [14]. Such nonlinear dynamical behavior of the KDP crystal suggests that, in the paraelectric neighborhood of T_c, where the average polarization is zero, higher orders of the order-parameter fluctuation play an important role as is represented by Duffing's highly nonlinear equation. Along with the polarization fluctuation, electromechanical coupling, caused by the piezoelectric effect, gives another source of nonlinear phenomena in the low-frequency region of KDP crystal near the phase transition temperature T_c in a driven *RLC* circuit.

In conclusion, we have observed a large variety of nonlinear dynamical responses of the KDP crystal near the ferroelectric phase transition temperature T_c, including period doubling of a fixed point, quasiperiodicity associated with frequency locking, and period doubling of a torus. It is associated with the polarization fluctuation, given by Duffing's nonlinear damped-anharmonic oscillator leading to period doubling, and the electromechanical coupling between the polarization P and the shear strain X of the KDP crystal, leading to quasiperiodicity. Observation of a finite number of period doublings of a torus in the crystal suggests that polarization fluctuation, as well as an electromechanical coupling due to the large atomic displacement of the crystal near the phase transition, plays a crucial role in the nonlinear dynamical behavior of the crystal. Now it is our conjecture that torus-doubling transition to chaos can be observed frequently in most ferroelectric materials because they have a strong electromechanical coupling between the piezoelectric oscillation and the large polarization fluctuation near the phase transition.

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*Email address: jcshin@phya.snu.ac.kr

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