Bifurcation response in a KH_2PO_4 crystal near the ferroelectric transition

Jung-Yun Huang and Jong-Jean Kim*
Department of Physics, Korea Advanced Institute of Science and Technology, P. O. Box 150, Cheongryang-ni, Seoul, Korea
(Received 10 July 1986)

Phase locking and period-doubling bifurcation are observed to characterize the nonlinear response of a driven KH_2PO_4 crystal at temperatures near the ferroelectric phase transition, where the crystal may well be represented by a coupled system of two anharmonic oscillators, a polarization-mode oscillator and an elastic shear-mode oscillator coupled by the piezoelectric effect.

I. INTRODUCTION

Nonlinear dissipative dynamics, which underlies most real systems, has been a concern of many theoretical and experimental research workers in recent years.[1-3] Even a simple nonlinear dynamical equation can show surprisingly irregular properties of its solution as a control parameter is slightly adjusted. The most striking feature of nonlinear dynamical equations, however, is the universality of the solutions from periodic to chaotic dynamics.[2]

Fluctuation is very important in both the critical dynamics of phase transitions and the chaotic dynamics of nonlinear systems.[2] The nonlinear dynamic response of the KH_2PO_4 (KDP) crystal near the phase transition may thus be informative about the dynamics of the phase transition in the crystal. A ferroelectric Rochelle-salt crystal was found to exhibit period-doubling bifurcations and chaos when it was employed as a biased piezoelectric element in a nonlinear electrical oscillator.[4] The KDP crystal is well known for its use as a device element in nonlinear optics. However, the nonlinear response of KDP crystal has not been much studied in the low-frequency region, where the nonlinearity seems to become significant only near the phase transition at $T=123$ K.

In this work we report our observation of nonlinear dynamic responses in a KDP crystal near T_c, which may help to study the correlation between nonlinearity and phase-transition dynamics.

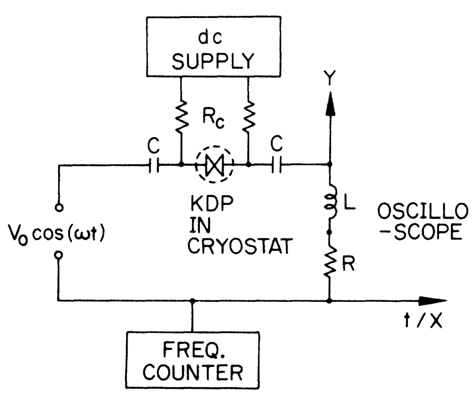


FIG. 1. Circuit to observe bifurcations in KDP: $R_c=1$ MΩ, $C=1$ μF, $R=390\Omega$, $L=5$ mH. Sample dimension is $1.5\times10.8\times0.4$ mm^3.

II. EXPERIMENT

In the KDP crystal the shear strain S_6 is coupled piezoelectrically with the c-axis lattice polarization P_3, and the crystal can be tuned to a piezoelectric oscillation along the 45° diagonal in the ab plane. We prepared 45° c-cut samples, where vibrational modes other than the longitudinal oscillations along the sample length were decoupled from the piezoelectric excitation.

We employed the RLC circuit of Fig. 1 to observe the nonlinear response of the KDP crystal to a sinusoidal excitation. An external bias field was used to adjust the resonant frequency of the KDP sample. The sample temperature was controlled within 0.05 K at temperatures less than 1 K above the phase-transition temperature. We had to avoid resonant driving conditions for which samples soon shattered due to a large S_6 strain. Instead, we performed experiments near the antiresonance frequencies. Real-time display (x-t mode) as well as Lissajous-figure construction (x-y mode) of the response signals against the driving sine wave were made on an oscilloscope.

III. RESULTS AND DISCUSSION

Figure 2 shows a real-time display of a period-doubling response observed in KDP. When the frequency ω was

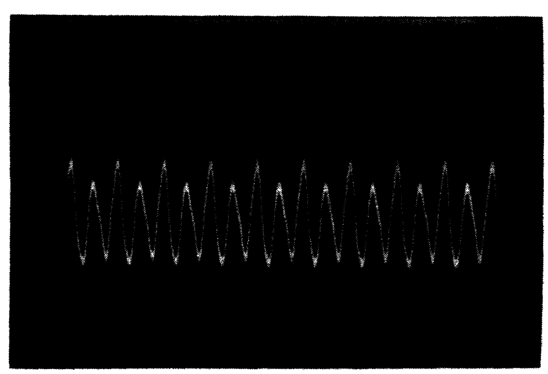
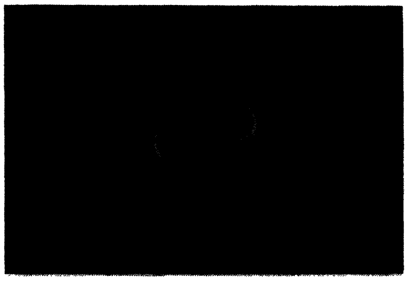
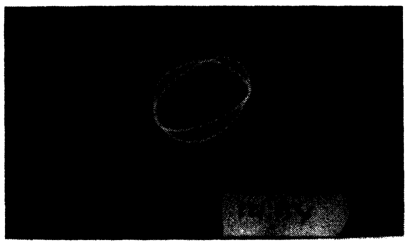


FIG. 2. Period-two signal: $\omega=88.18$ kHz, $V_0=6.5$ V.

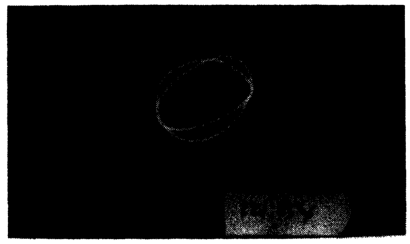


(a)

(c)



(b)



(d)

FIG. 3. Period-doubling bifurcation and chaos: external bias =928 V/cm (x: 5 V/div, y: 5 V/div), (a) 120.2 kHz, (b) 120.637 kHz, (c) 120.644 kHz, (d) 121.154 kHz.

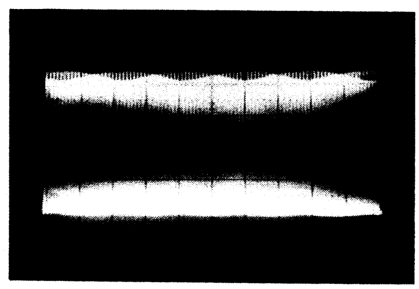
used as a bifurcation parameter, we could observe a complete period-doubling sequence to chaos as displayed in Fig. 3. Phase locking between mode resonance and the driving signal was also observed when the mode-resonance frequency became a rational fraction of the driving frequency. In the KDP crystal, phase locking of the type $l/(l+\Delta l)$ or $l/(2l+\Delta l)$, where l and Δl are integers, was easily observed. Figure 4(a) shows a $l/(2l+\Delta l)$-type phase locking, where the mode-resonance and driving frequencies are 46.8 and 90.93 kHz, respectively, to give $\frac{46.8}{90.93} \simeq \frac{17}{33}$ corresponding to $l=17$, $\Delta l=-1$. Because the resonant frequencies of the sample are dependent on the bias field, the KDP sample may experience broad resonances during the full cycle evolution of the driving sinewave signal. Mode coupling between resonances may also exist to produce the quasiperiodic responses[5,6] in the driven KDP system. If we consider only the dielectric response of the KDP crystal near T_c in the low-frequency circuit of Fig. 1, we obtain a nonlinear oscillator equation for the circuit. The Gibbs free energy appropriate for the phase transition in KDP,[7]

$$G=\alpha(T-T_0)P^2/2-\beta P^4/4+\gamma P^6/6 \ , \tag{1}$$

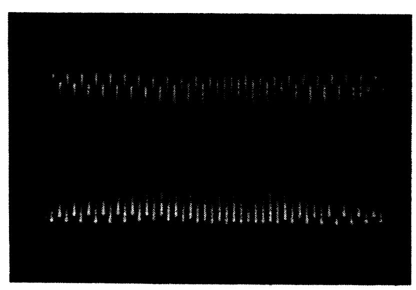
gives the electric field E $(=\partial G/\partial P)$ as a function of the polarization field P in the crystal. When the RLC circuit of Fig. 1 is driven by a sinusoidal input $V_0\cos(\omega t)$, we thus obtain

$$(1/\omega_0^2)\frac{d^2}{dt^2}P+(R/L\omega_0^2)\frac{d}{dt}P+(4\alpha\gamma/\beta^2)(T-T_0)P - (4\gamma/\beta)P^3+(2\gamma/\beta)^2P^5=(4\gamma V_0/\beta^2 d_0)\cos(\omega t) \ , \tag{2}$$

where R represents resistance, L inductance, T temperature, d_0 sample thickness, A sample area, and ω_0 equal to $\beta\sqrt{d_0/4\gamma LA}$ has the dimensions of frequency. Solutions of this nonlinear oscillator equation include phase locking and intermittency.[8] In real KDP samples near T_c the polarization mode is coupled with the elastic shear mode by the piezoelectric effect, and we can also explain the bifurcations in the KDP sample as those of a coupled system of two anharmonic oscillators. The KDP system may also be



(a)



(b)

FIG. 4. (a) Phase-locked signal: $\omega=90.93$ kHz, $V_0=8$ V (x: 0.2 ms/div, y: 5 V/div); each envelope contains about 34 cycles of the driving frequency. (b) Enlarged view of (a) (x: 50 μs/div); large-amplitude signals become small-amplitude signals and small signals become large signals as passing from one envelope to another.

represented by a many-body-interaction pseudospin system, which shows chaotic responses to an external oscillating field.[9]

All these nonlinear response signals of KDP were observable only near the phase-transition temperature in accordance with the piezoacoustic soft-mode signals.[10] Optical nonlinearity of the KDP crystal is well known as a room-temperature observable. However, the low-frequency nonlinear behavior of KDP in the piezoelectric regime seems to be strong only near the phase transition at $T_c = 123$ K, where the shear elastic stiffness decreases rapidly with temperature and the restoring force becomes strongly nonlinear due to the large-amplitude atomic displacements of the soft mode.[7]

---

*Author to whom correspondence should be addressed.

[1] *Proceedings of the International Conference on Order in Chaos, Los Alamos, 1982,* edited by D. Campbell and H. Rose [Physica D **7**, 3 (1983)].

[2] J. P. Crutchfield, J. D. Farmer, and B. A. Huberman, Phys. Rep. **92**, 45 (1982).

[3] H. G. Schuster, *Deterministic Chaos* (Physik-Verlag, Weinheim, 1984).

[4] D. J. Jefferies, Phys. Lett. **90A**, 316 (1982).

[5] J. Stavans, F. Heslot, and A. Libchaber, Phys. Rev. Lett. **55**, 596 (1985).

[6] Da Ren He, W. J. Yeh, and Y. K. Kao, Phys. Rev. B **31**, 1359 (1985).

[7] M. E. Lines and A. M. Glass, *Principles and Applications of Ferroelectric and Related Materials* (Clarendon, Oxford, 1977).

[8] (a) T. Yamada and H. Fujisaka, Prog. Theor. Phys. Suppl. **64**, 269 (1978). (b) We also found numerical solutions of intermittency and phase locking from Eq. (2) (unpublished).

[9] M. Inoue and H. Koga, Prog. Theor. Phys. **69**, 1403 (1983).

[10] J. Y. Koo and J. J. Kim, Jpn. J. Appl. Phys. **24**, Suppl. 2, 704 (1985).

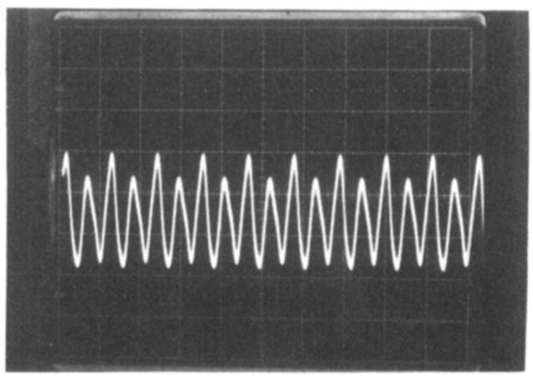


FIG. 2. Period-two signal: $\omega = 88.18$ kHz, $V_0 = 6.5$ V.

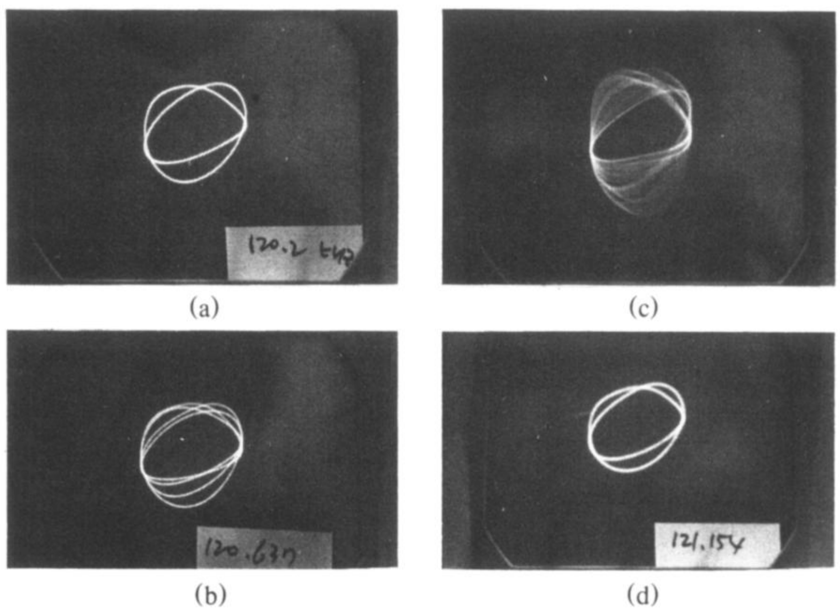
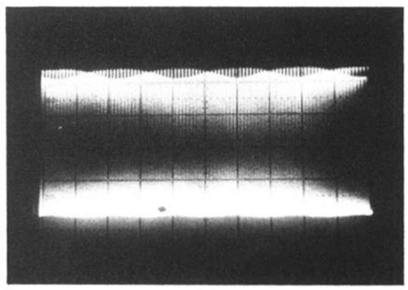
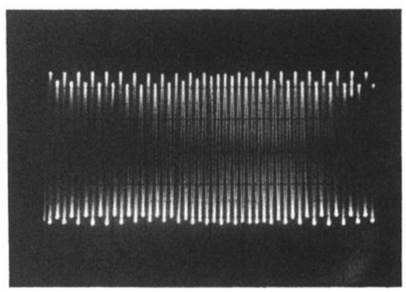


FIG. 3. Period-doubling bifurcation and chaos: external bias =928 V/cm (x: 5 V/div, y: 5 V/div), (a) 120.2 kHz, (b) 120.637 kHz, (c) 120.644 kHz, (d) 121.154 kHz.



(a)



(b)

FIG. 4. (a) Phase-locked signal: $\omega = 90.93$ kHz, $V_0 = 8$ V (x: 0.2 ms/div, y: 5 V/div); each envelope contains about 34 cycles of the driving frequency. (b) Enlarged view of (a) (x: 50 μs/div); large-amplitude signals become small-amplitude signals and small signals become large signals as passing from one envelope to another.