Temperature-dependent nonlinear dynamic response of a KH_2PO_4 crystal near phase transition

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(Received 8 July 1988)

Nonlinear dynamic characteristics of a KH_2PO_4 crystal near the ferroelectric phase transition were observed to change across the transition temperature T_c. Above T_c period-doubling cascades and intermittency chaos were dominant signals, while phase locking and quasiperiodicity were dominant observations below T_c. This difference was attributed to a change in nature of the polarization dynamics in the KH_2PO_4 crystal across T_c: a damped-anharmonic-oscillator behavior above T_c and a coupled-mode behavior below T_c.

Nonlinear dynamic susceptibility of potassium dihydrogen phosphate [KH_2PO_4 (KDP)] crystal was found to be significant only near the phase transition temperature $T_c \simeq 123$ K.[1] KDP crystal, undergoing both a displacive structural phase transition and a proton-ordering order-disorder phase transition, becomes very unstable near the transition temperature T_c both lattice dynamically, with a soft-mode softening, and thermodynamically, with a Landau-potential softening. Landau theory of phase transitions in KDP involves a double-well-type nonlinear potential, and the nonlinearity of the polarization dynamics is an intrinsic characteristic of the ferroelectric phase transition in KDP. Polarization fluctuation grows as the instability increases with increasing nonlinearity of the potential when the transition temperature is approached. We want to report here experimental studies on the nonlinear dynamic response of KDP crystal near the phase transition. A simple inductance-capacitance-resistance (LCR) circuit of Fig. 1, with KDP as a temperature-dependent nonlinear element, was used to observe the signals presented in this work.

KDP crystal near the ferroelectric phase transition can be described by the following thermodynamic potential of Landau theory:[2]

$$G = \alpha_0 (T-T_0)\frac{P^2}{2} - \beta\frac{P^4}{4} + \gamma\frac{P^6}{6} + \cdots , \tag{1}$$

where T_0 represents the stability limit and the order parameter P, the fluctuation (ΔP) or induced polarization at $T > T_0$. The electric field along the ferroelectric axis, E_3, is then given by $E_3 = -\partial G/\partial P_3$

$$E_3 = \alpha_0 (T-T_0) P_3 - \beta P_3^3 + \gamma P_3^5 . \tag{2}$$

From Kirchhoff's laws the circuit equation of our LCR circuit, Fig. 1, is given by

$$L\ddot{Q} + R\dot{Q} + V_{\text{KDP}} = V_0 \cos(\omega t) , \tag{3}$$

where $Q = P_3 A$, $V_{\text{KDP}} = E_3 d$, and A and d are area and thickness of the KDP sample respectively. From Eqs. (1), (2), and (3) we obtain

$$LA\ddot{P}_3 + RA\dot{P}_3 + [\alpha_0(T-T_0)P_3 - \beta P_3^3 + \gamma P_3^5] d = V_0 \cos(\omega t) . \tag{4}$$

Reducing the variables to dimensionless ones we can obtain

$$\ddot{P} + \rho\dot{P} + \alpha P - 2P^3 + P^5 = V\cos(\Omega\tau) , \tag{5}$$

where

$$P = P_3/\sqrt{\beta/2\gamma}, \quad \Omega = \omega/\omega_0, \quad \tau = \omega_0 t ,$$
$$\rho = R/L\omega_0, \quad \alpha = 4\gamma\alpha_0(T-T_0)/\beta^2 ,$$
$$V = V_0/(LA\omega_0^2\sqrt{\beta/2\gamma}), \quad \omega_0^2 = d\beta^2/4LA\gamma ,$$

and time derivatives are now with respect to τ. Equation (5) belongs to the Duffing's nonlinear oscillator equation, where we have solutions of period-doubling cascades and intermittencies.[3] Period-doubling nonlinear oscillations in KDP crystal were observed indeed at $T \gtrsim T_c$ very near the ferroelectric transition.[4] Period tripling has been observed to be an important precursor to the type-II intermittency in our previous work.[5] Period-tripling signals of Fig. 2, observed at $T \gtrsim T_c$, thus represent the Duffing's-oscillator behavior of KDP at $T \gtrsim T_c$.

In Fig. 3 we have shown signals observed at $T \lesssim T_c$, corresponding to phase locking and quasiperiodicity. In KDP

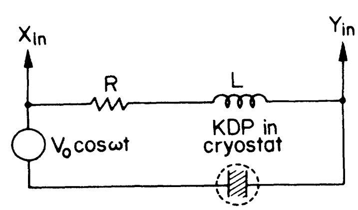


FIG. 1. LCR circuit to study the nonlinear-dynamic response of KH_2PO_4 crystal near ferroelectric transition ($R = 390$ Ω, $L = 5$ mH) at $T_c \simeq 123$ K.

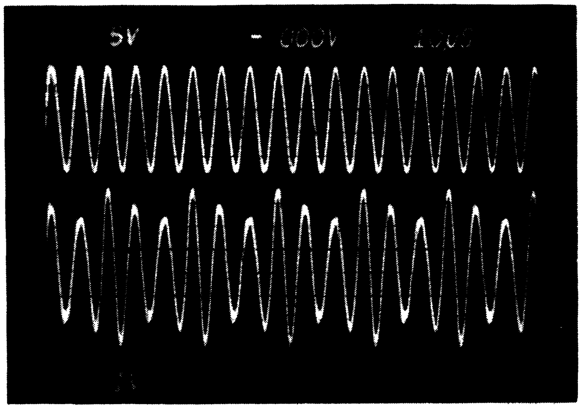


FIG. 2. Period tripling (3T) signal observed at $T=124$ K ($\omega=69.01$ kHz, $V_3=6.5$ V).

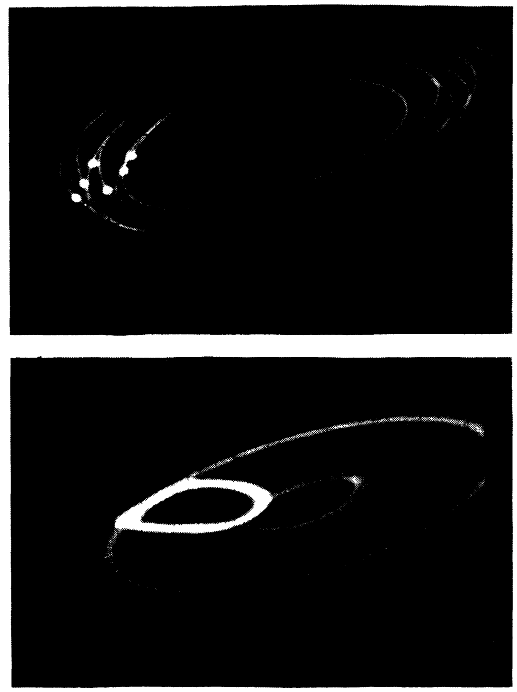
crystal the soft-mode polarization can be coupled with the acoustic shear deformation. We then have in the piezoelectric low-frequency regime the coupled equations of motion given in the form[6]

$$\ddot{P}+a\dot{P}+u(P,X)=V\cos(\Omega\tau)\,,$$
$$\ddot{X}+b\dot{X}+v(X,P)=0\,, \qquad (6)$$

where P denotes ferroelectric polarization, X shear strain, and $u(P,X)$ and $v(X,P)$ piezoelectric coupling terms. Equations (6) can be reduced to a pair of coupled Poincaré return maps of the type

$$P_{n+1}=f(P_n,X_n),\quad X_{n+1}=g(X_n,P_n)\,. \qquad (7)$$

This coupled Poincaré return map leads to the phase lock-

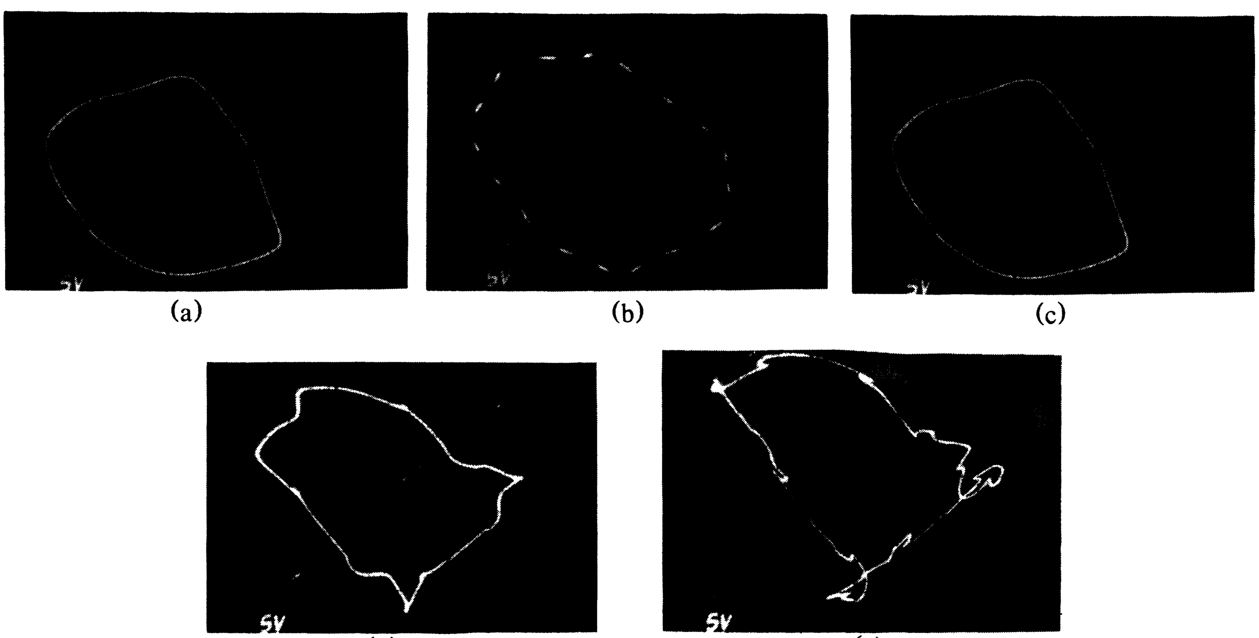


(a)

(b)

FIG. 3. (a) Signals of phase-locked and (b) quasiperiodic motions on a torus: from Lissajous figures constructed of (X_{in}, Y_{in}) observed at $T=121$ K.

ing and quasiperiodicity routes to chaos.[7,8] We can thus expect to observe phase locking and quasiperiodicity from the KDP when KDP can be described by Eq. (6). In the present work we have succeeded to observe both the phase locking and the quasiperiodicity signals from KDP slight-



(a) (b) (c)

(d) (e)

FIG. 4. Circle maps of quasiperiodicity route to chaos (return maps of peak-sampled Y_{in}) observed at $T=121$ K ($\omega_0=30.5$ kHz, $L=3$ mH): (a) $V=3.7$ V, (b) 4.0 V, (c) 4.25 V, (d) 4.55 V, (e) 4.75 V.

ly below the ferroelectric transition temperature T_c. Figure 4 shows the circle maps on the Poincaré section through the torus as observed in the KDP circuit. The continuous-circle map of the quasiperiodicity is seen to be sharply cornered, an indication of the Ruelle-Takens-Newhouse transition[8] to chaos, as the control parameter V is increased. Dotted-circle maps were also easily observed in the same ferroelectric vicinity near T_c, corresponding to phase locking states.

In conclusion we have shown that nonlinear dynamic nature of a KDP crystal also changes across the ferroelectric phase transition. Period-doubling cascades and intermittencies were observed to be dominant at temperatures above T_c while phase locking and quasiperiodicity were dominant at temperatures below T_c. This difference seems to suggest that in the paraelectric neighborhood of T_c, where $\langle P \rangle = 0$ but $\langle P^2 \rangle \neq 0$, the polarization dynamics of KDP is fluctuation dominant and may be represented by a damped anharmonic oscillator of Eq. (5) leading to period doubling and intermittencies but in the ferroelectric vicinity of T_c, where $\langle P \rangle \neq 0$, the piezoelectric coupling between $\langle P \rangle$ and shear strain X may control the polarization dynamics of KDP, and the dynamics may be described by the coupled equations (6) liable to phase locking and quasiperiodicity. This observation is consistent with the results[9] of the dynamic mean-field theory of the KDP pseudospin system that for $T > T_c$ only the transverse modes contribute to the polarization fluctuations but both longitudinal and transverse modes contribute for $T < T_c$.

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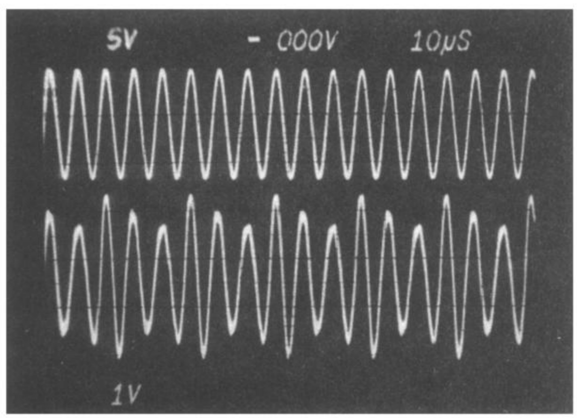

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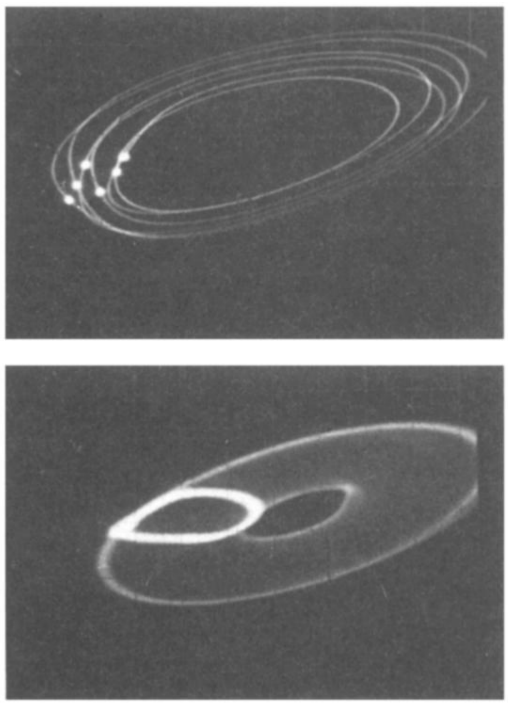

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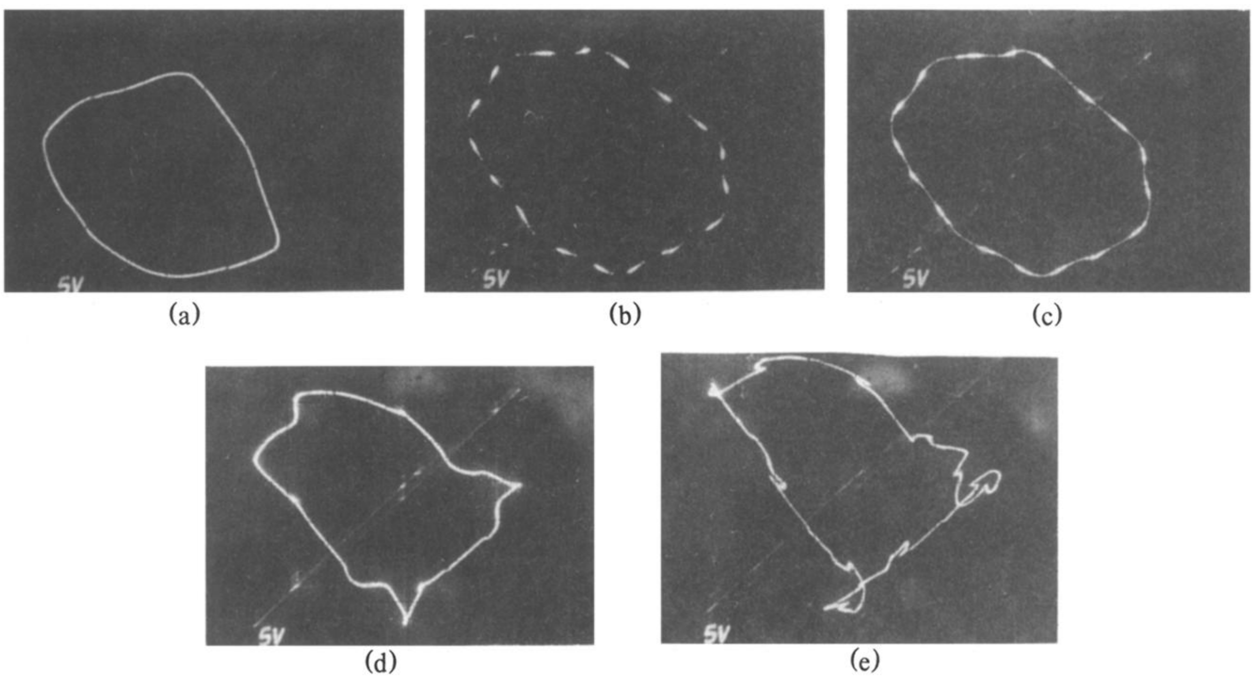

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