Efficient excitation of nonlinear phonons via chirped pulses: Induced structural phase transitions

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Nonlinear phononics play important role in strong laser-solid interactions. We discuss a dynamical protocol for efficient phonon excitation, considering recent inspiring proposals: inducing ferroelectricity in paraelectric perovskites, and inducing structural deformations in cuprates [Subedi et al., Phys. Rev. B 89, 220301(R) (2014); 95, 134113 (2017)]. High-frequency phonon modes are driven by midinfrared pulses, and coupled to lower-frequency modes those indirect excitations cause structural deformations. We study in more detail the case of KTaO$_3$ without strain, where it was not possible to excite the needed low-frequency phonon mode by resonant driving of the higher frequency one. Behavior of the system is explained using a reduced model of coupled driven nonlinear oscillators. We find a dynamical mechanism which prevents effective excitation at resonance driving. To induce ferroelectricity, we employ driving with sweeping frequency, realizing so-called capture into resonance. The method can be applied to many other related systems.

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I. INTRODUCTION

Research in ultrafast light control of materials has attracted a lot of interest recently [1–13]. Intense midinfrared pulses have been used to directly control dynamics of the crystal lattice [5–10,12,13], in particular, inducing melting of orbital and magnetic orders. In many recent suggestions and experiments phonon modes are driven indirectly: a laser drives a high-frequency infrared-active phonon mode $Q$ which then excites required modes, not easily accessible by direct drive, by means of nonlinear couplings. Here we suggest to add a useful tool to the arsenal of nonlinear phononics: capture into a resonance (see [14–16] for a detailed mathematical introduction). Such a phenomenon is encountered in classical and celestial mechanics [14,15,17,18] and was employed, e.g., in plasma and accelerator physics [19–26]. In a nonlinear system with near-resonant driving, as amplitude of perturbation grows, frequency of the system varies, and it stops to absorb energy efficiently. Driving with changing frequency enables us to lock the system into the regime where the resonance between the drive and the system is dynamically sustained [16].

We find that in perovskite paraelectric KTaO$_3$ (being described below), as well as in other systems [like La$_2$CuO$_4$ cuprate (LCO)], it is possible to considerably excite a high-frequency phonon mode using a protocol based on capture into the resonance, which requires a pulse with chirped frequency. This excitation makes a coupled low-frequency phonon mode dynamically unstable. Pulses with frequency chirps on the picosecond time scale have been generated, e.g., in FELIX [27,28].

II. THE MODEL

In all considered examples below, a zone-center optical-phonon mode $Q$ with (at least) quartic nonlinearity is driven by a laser pulse creating effective potential for a coupled lower-frequency phonon mode, those excitations trigger a structural phase transition. So let us start with a single driven mode (adding the coupled modes later). Neglecting dissipation, our starting Hamiltonian $H = \frac{p^2}{2} + \frac{\delta Q^4}{4} = Q_0 \sin \Phi(t)$, where $\Phi(t)$ is the phase of the driving, $\Phi(t) = \Omega(t)$, $\Omega_0$ is the linear frequency of the driven phonon mode, $Q$ is the normal coordinate, and $P$ is the conjugate momentum; $c_4, c_6, \ldots$ are anharmonic coefficients, $F_0(t)$ is the amplitude of the external field. Introducing symplectic coordinates $P = \sqrt{2J/\Omega_0} \sin \Phi$, $Q = \sqrt{2J/\Omega_0} \sin \Phi$ we then make a transformation to the resonance phase $\xi = \phi - \Phi$ using the time-dependent generating function $W = J(\phi - \Phi)$. The new Hamiltonian $H' = H - J \Omega$ can be averaged over the fast phase, with the result (neglecting nonlinearities higher than the quartic for a while) $H' = \delta \Omega J + \frac{3}{2} c_4 J^2 - J F_0 \sqrt{2J/\Omega_0} \cos \xi$, where $\delta \Omega \equiv \Omega_0 - \Omega$. Introducing now $x = \sqrt{2J} \sin \xi$, $y = \sqrt{2J} \cos \xi$, we get an effective Hamiltonian $H = \frac{3}{2} c_4 x^2 + \frac{3}{2} \mu y^2 + \frac{3}{2} \mu^2 (x^2 + y^2)^2 - \frac{F_0}{2} x^2$. Upon rescaling $H \rightarrow H' / \sqrt{3} c_4$, $t \rightarrow t \sqrt{3} c_4$, and introduction of $\lambda = -\frac{\delta \Omega}{2} / \sqrt{3} c_4$, $\mu = -\frac{F_0}{2 \sqrt{3} c_4}$ we bring the Hamiltonian to the form

$$H = (x^2 + y^2)^2 - \lambda(t)(x^2 + y^2) + \mu(t)y.$$ (1)

This Hamiltonian is often encountered in problems of celestial mechanics and plasma physics [15]. Under slow change of frequency and/or amplitude of driving, parameters of (1) are changing (increasing frequency corresponds to $\lambda > 0$). Corresponding phase portraits are shown in Fig. 1. A bifurcation happens at $\lambda_* = \frac{1}{2} \mu^{2/3}$. Below this value, there is a single equilibrium (A), while at higher values of $\lambda$ there are two stable (A, B) and one unstable equilibrium (C). Provided certain conditions are met, a phase particle can follow the initial equilibrium point (A) which moves away from origin (correspondingly, in the original system amplitude of oscillation grows, while its frequency remains approximately equal to the instantaneous frequency of the drive, so this regime...
excitation. With dissipation, the equation of motion becomes

to construct a simple and effective protocol for phonon system nonadiabaticity and dissipation become very important, point will be thrown away from the resonance. In our realistic inside separatrix loops it can be predicted when the phase are changing, phase space area within the trajectory remains a great detail [15,16]. E.g., as the parameters of the system are changing, phase space area within the trajectory remains unaffected (Fig. 6(b), bottom curve).

is called capture into the resonance). Under influence of a Gaussian pulse with fixed frequency, the point A is shifted by a certain amount and then returns back to the origin [see Fig. 6(b), upper curve]. In contrast, a pulse with sweeping frequency can shift the equilibrium far away [Fig. 6(b), bottom curve]. In the adiabatic approximation, dynamics can be described in a great detail [15,16]. E.g., as the parameters of the system are changing, phase space area within the trajectory remains approximate adiabatic invariant, and from behavior of the areas inside separatrix loops it can be predicted when the phase point will be thrown away from the resonance. In our realistic system nonadiabaticity and dissipation become very important, nevertheless qualitative understanding of dynamics allows us to construct a simple and effective protocol for phonon excitation. With dissipation, the equation of motion becomes

which is the same as in a damped and driven Duffing oscillator (the coefficient $\gamma$ quantifies damping). At fixed frequency, searching for a periodic solution $Q = A \sin(\Omega t + \phi_0)$, one gets approximately a cubic equation for the amplitude:

$$A^2(\gamma^2 \Omega^2 + (\Omega_0^2 - \Omega^2 + 3c_4 A^2))^2 = F_0^2. \quad (3)$$

III. RESULTS

Solutions of Eq. (3) are shown in Fig. 1(a) for different values of the driving amplitude $F_0$. They are dissipative counterparts of fixed points in phase portraits of (1). One can see that driving frequency higher than the resonant one allows us to achieve higher steady-state amplitudes. To demonstrate induced ferroelectricity, consider a specific example of KTaO$_3$: a perovskite oxide with cubic structure, possessing a paraelectric phase. Such materials have four triply degenerate optical-phonon modes at the zone center. Three of these modes are infrared active (have the irreducible representation $T_{1u}$ [3]). The remaining one is optically inactive [3]. Ferroelectricity is related to dynamical instability of an infrared-active transverse optical-phonon mode: most ferroelectric materials show a characteristic softening of an infrared transverse optical mode as the transition temperature is approached. In Ref. [3] it was investigated whether a similar softening and instability of the lowest frequency $T_{1u}$ mode can be achieved by an intense laser-induced excitation of the highest frequency $T_{1u}$ mode. In the case of cubic structure it was not achieved due to certain dynamical reasons (discussed below), however with addition of internal stress which modifies the crystal lattice such a mechanism worked. Let us consider in more detail the cubic structure case (without stress). The phonon frequencies calculated in Ref. [3] are $\Omega_1 = 85 \text{ cm}^{-1}$ and $\Omega_2 = 533 \text{ cm}^{-1}$ for the lowest and highest frequency $T_{1u}$ modes, respectively ($\Omega_0$ from previous discussion plays the role of $\Omega_0$ now). Following [3] we simplify the analysis by considering a case where the $x$ component of the highest frequency mode $Q_{hx}$ is pumped by an intense light source and influences the dynamics of the lowest frequency $T_{1u}$ mode along the longitudinal $Q_{iz}$ and transverse $Q_{iz}$ coordinates. Dynamics along the second transverse coordinate $Q_{ly}$ is neglected.

The energy surface $V(Q_{hx}, Q_{iz}, Q_{1z})$ has a complicated form with many kinds of nonlinear couplings and anharmonicities: $V(Q_{hx}, Q_{iz}, Q_{1z}) = \frac{3}{2} Q_{hx}^2 + \frac{5}{2} Q_{iz}^2 + \frac{5}{2} Q_{1z}^2 + V^{nl}(Q_{hx}, Q_{iz}, Q_{1z})$, where $V^{nl}$ is the nonlinear part of the energy, obtained in state-of-the-art calculations of [3], and given in Appendix A for convenience. We use these coefficients in our numerical calculations. The equations of motions are

$$\ddot{Q}_{hx} + \gamma_h \dot{Q}_{hx} + \Omega_{hx}^2 Q_{hx} = -\frac{\partial V^{nl}(Q_{hx}, Q_{iz}, Q_{1z})}{\partial Q_{hx}} + F(t),$$

$$\ddot{Q}_{1z} + \gamma_1 \dot{Q}_{1z} + \Omega_{1z}^2 Q_{1z} = -\frac{\partial V^{nl}(Q_{hx}, Q_{iz}, Q_{1z})}{\partial Q_{1z}}, \quad (4)$$

where $j = x, z$; $\gamma_0$ and $\gamma_1$ are damping coefficients (typically a few percent of the corresponding harmonic frequencies), external force $F(t) = Z_{hx}^* E_0 \sin(\Omega t) e^{-r^2/2\sigma_2^2}$, $Z_{hx}^*$ is the effective charge, $\Omega$ is the driving frequency, and $E_0$ is the amplitude of the electric field. $\sigma_2 \equiv \sigma/2\sqrt{2\ln2}$, where $\sigma$ is the width of the pulse.

Qualitative understanding of possible dynamics can be captured by drawing a projection of the potential energy $V(Q_{1z}, Q_{hz})$ by the plane $Q_{1z} = 0$ (Fig. 3 of [3]). Resulting curves $V(Q_{hz})$ at fixed values of $Q_{hz}$ have a single-well form (at small absolute values of $Q_{hz}$), or a double well form (at larger $Q_{hz}$). In the latter case, induced ferroelectricity is possible, as finite value of $Q_{1z}$ at equilibrium corresponds to the ferroelectric phase. However, to reach such a state dynamically is a nontrivial issue. Excitation of the $Q_{hx}$ mode should be done with a pulse of limited power and duration. When the driving frequency $\Omega$ is chosen close to the phonon mode frequency $\Omega_h$ [3], the transverse mode $Q_{1z}$ remains almost unaffected (Fig. 2) due to insufficient amplitude of the $Q_{hz}$.
mode. In Ref. [3], the amplitude of driving was varied in a large range, up to pump amplitudes of 100 MV cm\(^{-1}\), with no signs of dynamical instability in \(Q_{1z}\). An increase of the pump amplitude makes the dynamics of \(Q_{1z}, Q_{hx}\) chaotic, but does not result in a noticeable response of the \(Q_{1z}\) mode: at strong driving the “auxiliary” longitudinal mode \(Q_{1z}\) becomes excited (see Appendix B), and chaotic dynamics prevents efficient excitation of \(Q_{hx}\).

There is a range of driving frequencies (away from the exact resonance with \(\Omega_{0}\)), where a pulse of the same amplitude can effectively excite all three modes, inducing transient dynamical instability in \(Q_{1z}\). Indeed, shifting driving frequency about 15–20\% from the exact resonance with \(\Omega_{0}\) leads to considerable excitation of the modes \(Q_{1z}, Q_{hx}\) (Fig. 3). The mode \(Q_{hx}\) experiences beatings which create transient double-well potential for the \(Q_{1z}\) mode [Fig. 3(d)]. From the full potential energy of the system, we can single out the term quadratic in \(Q_{1z}: (\frac{\Omega_0^2}{2} + G(t))Q_{1z}^2\), where \(G(t) = m_3 Q_{hx}^4 + d Q_{hx}^2 Q_{1z} + \Omega_0^2 Q_{hx}^2 + p Q_{1z}^2 + e_2 Q_{1z}^2 Q_{1x}^2 + e_3 Q_{1x}^2 Q_{hx}\) (see Appendix A). Due to strong coupling between \(Q_{hx}\) and \(Q_{1z}\) modes, they oscillate synchronously although their linear frequencies are very different. When the average value of the coefficient \((G(t))\) exceeds \(-\Omega_0^2/2\), the effective potential for the mode \(Q_{1z}\) becomes unstable. Due to violent beatings in the \(Q_{hx}\) mode, it does not happen smoothly, and only for a short fraction of the pulse excitation time the mode \(Q_{1z}\) experiences the inverted parabolic potential [at the first maxima of the beatings; see Fig. 3(d)].

There is a way to create the needed effective potential in a more robust way. Consider driving with sweeping frequency. A chirped pulse has the form \(F = F_0(t)\sin\Phi(t)\), where \(\Phi(t) = \Omega_0 t + \frac{\omega t^2}{2}, F_0(t) = \exp[-t^2/2(\sigma^2/8\text{ln}2)]\). Time dependence of the instantaneous frequency and the amplitude translates into the dependence of parameters \(\mu, \lambda\) of the Hamiltonian (1) on time. Corresponding phase portraits are slowly deformed, and, if our phase point is not thrown away from the region where the initial equilibrium is located (see [16] for details), it oscillates around the equilibrium point moving away from the origin. Such a regime, illustrated in Fig. 4, is not only effective for excitation of the \(Q_{hx}\) mode, but also provides smooth generation of the effective potential for the \(Q_{1z}\) mode [Fig. 4(d)].

The important feature of the dynamics is that the auxiliary longitudinal mode \(Q_{1z}\) also gets excited considerably. Unlike the case of very strong resonant driving (Appendix B), where chaotic dynamics happens after excitation of the \(Q_{1z}\) mode, here the longitudinal modes oscillate synchronously (in 1:1 resonance), and the resulting dynamics is regular (because the pulse with sweeping frequency excites the system in such a way that a phase point remains not far from the instantaneous equilibrium point).

Note that damping plays an important role in dynamics. Damping parameters typically are 5–10\% of corresponding linear frequencies. Assuming higher damping for low-frequency phonons, the mode \(Q_{1x}\) is not excited considerably, and dynamics can be understood from the driven Duffing oscillator model for the \(Q_{hx}\) mode alone. While in the Hamiltonian model there are three equilibria above the critical frequency detuning and a phase point captured into resonance can reach large amplitudes of \(Q\) moving near one of them, damping leads to termination of this process at the tip of the resonance “tongue,” where stable and unstable equilibria collide and annihilate. At weak driving amplitudes, the tips lie at a “backbone” defined as \(A_{np} = \frac{E_0}{\Omega_0^2}, \omega_{np} = 1 + \frac{3c_3 F_0^2}{\gamma^2 \Omega_0^2}\). For a rough estimate of the optimal sweep, assume that a pulse starts with the linear resonance frequency \(\Omega_0\), and reaches the tip of the resonance tongue at its maximum. Then, the estimate for the sweeping rate is \(\alpha = 3c_3 F_0^2 / \gamma^2 \Omega_0^2\). We make numerical experiments with various sweeping rates and amplitudes of driving [see Figs. 4(e) and 4(f)], and find a remarkable improvement in the efficiency of excitation compared to pulses with constant frequency. Most exciting, the protocol with sweeping frequency works also for much shorter, subpicosecond pulses. We show in
potential energy. (e),(f) Maximal amplitude of (e) with sweeping frequency. Instantaneous frequency in the center before the maximum of the pulse.

drive with the constant frequency of modes \( G \) instantaneous coefficient rates modes under driving with sweeping frequency at different sweeping rates. Only the lowest curve lead to excitation of the Raman mode.

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Fig. 4. Dynamics of the KTaO\(_3\) model system under driving with sweeping frequency. Instantaneous frequency in the center of the pulse is \( \Omega = 1.2 \Omega_0 \). (a)–(d) \( Q_{1x}, Q_{1z}, Q_{h z} \) modes, and the instantaneous coefficient \( G(t) \) of the \( \frac{1}{2} \Omega_0^2 + G(t)Q_z^2 \) term in the potential energy. (e),(f) Maximal amplitude of (e) \( Q_{h z} \), and (f) \( Q_{1z} \) modes under driving with sweeping frequency at different sweeping rates \( a \). Curves from top to bottom correspond to amplitudes of driving \( F_0 = 30, 20, 10 \) MV cm\(^{-1}\). Damping rates are 5% of the linear frequencies. Instantaneous frequency of the drive is linearly increased, reaching the linear frequency of the phonon mode \( \Omega_0 \) at \( t_a = 0.75 \sigma \) before the maximum of the pulse. \( \sigma = 2 \) ps. \( a = 0 \) corresponds to the drive with the constant frequency \( \Omega = \Omega_0 \).

Fig. 5 an example with \( \sigma = 350 \) fs, which corresponds to laser parameters used in the Cavalleri group.

The same approach applies not only to ferroelectrics, but to many other systems, e.g., laser driven LCO [1]. There, a relevant reduced model consists of an infrared-active \( Q_{IR} \) mode (described by a driven Duffing oscillator) coupled to a Raman mode \( Q_R \) by a quadratic-quadratic term. The \( Q_R \) \([18]\) mode describes in-plane rotations of CuO\(_6\) octahedra, whereas the \( Q_{IR} \) mode describes in-plane stretching of Cu-O bond. The energy surface has the form \( V = \Omega_0^2 Q_{IR}^2 + \Omega_1^2 Q_{IR}^2 + c_4 Q_{IR}^4 + b_4 Q_R^2 - \frac{g}{2} Q_{IR}^2 Q_R^2 \). Values of the coefficients were derived in elaborate calculations of [1]. There is a single-potential well around the equilibrium value for the \( Q_R \) mode at small amplitudes of \( Q_{IR} \), which becomes a double-well potential at larger amplitudes of \( Q_{IR} \) [the instantaneous quadratic potential felt by the slow mode is \( \frac{Q_{IR}^2}{\Omega_0^2} (\Omega_1^2 - g Q_{IR}^2) \), which becomes an inverted parabolic potential for sufficiently high amplitudes of the driven IR mode]. The critical value of driving force \( F_c \) depends on detuning \( \delta \Omega = \Omega_0 - \Omega \) and can be made smaller than its value on the resonance (being used in Ref. [1]). Indeed, to the first approximation, the averaged potential for the slow mode is \( \frac{Q_{IR}^2}{\Omega_0^2} (\Omega_1^2 - g Q_{IR,max}^2/2) \) and becomes unstable at critical value of the fast mode amplitude \( Q_{IR,max} = \Omega_1 \sqrt{2g/\Omega_0} \). This can be achieved at sufficiently smaller driving force amplitudes provided a sweeping frequency pulse is used. We show corresponding results of numerical calculations in Fig. 6. Figure 6(c) shows also instantaneous locations of stable equilibrium of the effective potential as a function of time for different values of sweeping rates.

Excitation of the in-plane rotations associated with the \( Q_R \) mode can be used to modulate superexchange coupling in this cuprate [1]. Recently there has been a lot of interest in effective models arising from periodic driving [29–39], and the suggested method can be useful for this area of research as well. It can be useful also for recent proposals and experiments on driven orthorhombic perovskites (like ErFeO\(_3\); see [11,13]), where three-linear phonon coupling is realized: two high-frequency infrared-active modes are coupled to the third, Raman mode.

IV. CONCLUSIONS

To conclude, we demonstrate that drastic improvement in efficiency of excitation of nonlinear phonons can be achieved by chirped pulses. In terms of nonlinear dynamics of reduced classical models, capture into the resonance happens and the driven mode is transferred to a higher amplitude state efficiently, which triggers instabilities in the coupled slow modes, and corresponding phase transitions.
TABLE I. The values of the coefficients of the polynomial for energy surfaces of Raman and IR modes obtained in Refs. [1,3]. The units of a $Q^m Q^n Q^p$ term are meV $A^{-(m+p+n)}$.

| Coefficient | Term | KTaO$_3$ | Coefficient | Term | LCO |
|-------------|------|----------|-------------|------|-----|
| $\Omega_2^{2}$ | $Q_{3z}^2$ | 1043.77 | $\Omega_5^{2}$ | $Q_{3r}^2$ | 1462.3 |
| $\Omega_2^{4}$ | $Q_{3z}^4$ | 27.06 | $\Omega_1^{2}$ | $Q_{3r}^2$ | 103.55 |
| $\Omega_2^{6}$ | $Q_{3z}^6$ | 27.06 | $g$ | $Q_{3r}^2 Q_{3z}^2/2$ | 46.98 |
| $a_4$ | $Q_{3z}^4$ | 47.55 | $a_4$ | $Q_{3r}^4$ | 8.36 |
| $c_4$ | $Q_{3z}^2 Q_{3r}^2$ | 63.17 | $c_4$ | $Q_{3r}^2$ | 103.5 |
| $a_6$ | $-6.45$ | | | | |
| $c_6$ | $-0.733$ | | | | |
| $a_8$ | $1.47$ | | | | |
| $c_8$ | $0.438$ | | | | |
| $a_{10}$ | $-0.235$ | | | | |
| $c_{10}$ | $-0.0168$ | | | | |
| $a_{12}$ | $0.0243$ | | | | |
| $c_{12}$ | $-0.000129$ | | | | |
| $l$ | $-5.95$ | | | | |
| $m_1$ | $-1.03$ | | | | |
| $m_2$ | $-3.05$ | | | | |
| $n_1$ | $0.185$ | | | | |
| $n_2$ | $0.00435$ | | | | |
| $n_3$ | $-0.237$ | | | | |
| $n_4$ | $118.34$ | | | | |
| $n_5$ | $215$ | | | | |
| $n_6$ | $-175.58$ | | | | |
| $u_1$ | $-2.72$ | | | | |
| $u_2$ | $10.64$ | | | | |
| $u_3$ | $-22.81$ | | | | |
| $u_4$ | $25.38$ | | | | |
| $u_5$ | $19.09$ | | | | |
| $d$ | $6.29$ | | | | |
| $p$ | $-1.7$ | | | | |
| $q_1$ | $-1.7$ | | | | |
| $q_2$ | $-1.7$ | | | | |
| $r_1$ | $0.0935$ | | | | |
| $r_2$ | $0.00523$ | | | | |
| $r_3$ | $0.00523$ | | | | |
| $e_1$ | $6.61$ | | | | |
| $e_2$ | $-13.16$ | | | | |
| $e_3$ | $11.3$ | | | | |
| $f_1$ | $0.299$ | | | | |
| $f_2$ | $-0.68$ | | | | |
| $f_3$ | $1.39$ | | | | |
| $g$ | $1.02$ | | | | |
| $h_1$ | $-0.462$ | | | | |
| $h_2$ | $0.813$ | | | | |
| $h_3$ | $-0.745$ | | | | |

The method is especially remarkable in cases where a system cannot be excited by bare increase of the power of drive, like in KTaO$_3$. The approach can be useful in many recent proposals on laser-induced phase transitions.

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FIG. 7. Resonant driving, the same as in Fig. 2, but with a larger driving amplitude. From top to bottom: $Q_{3z}, Q_{3x}, Q_{hx}$ modes. $E_0 = 60$ MV cm$^{-1}$. Amplitude of driving is so large that dynamics become chaotic; nevertheless the mode $Q_{3z}$ is unexcited.

APPENDIX A: NONLINEAR PART OF THE POTENTIAL-ENERGY SURFACE FOR KTAO$_3$ AND LCO

The nonlinear part of the potential-energy surface of KTaO$_3$ is

$$V^{nl}(Q_{hx}, Q_{3z}, Q_{3x}) = \sum_{k=2}^{6}(a_{2k}(Q_{3z}^{2k} + Q_{3x}^{2k}) + c_{2k} Q_{hx}^{2k})$$

$$+ l Q_{3z}^2 Q_{hx}^2 + m_1 Q_{3z}^4 Q_{hx}^2 + m_2 Q_{3x}^2 Q_{hx}^4$$

$$+ n_1 Q_{3z}^4 Q_{hx}^4 + n_2 Q_{3z}^6 Q_{hx}^2 + n_3 Q_{3x}^2 Q_{hx}^6$$

$$+ t_1 Q_{3z}^4 Q_{hx}^2 + r_2 Q_{3z}^4 Q_{hx}^2 + r_1 Q_{3x}^4 Q_{hx}^4$$

$$+ p Q_{3z}^2 Q_{hx}^2 + q_1 Q_{3z}^2 Q_{hx}^2 + q_2 Q_{3x}^2 Q_{hx}^2$$

$$+ r_1 Q_{3z}^4 Q_{hx}^2 + r_2 Q_{3z}^4 Q_{hx}^2 + r_3 Q_{3x}^4 Q_{hx}^2$$

$$+ d Q_{3z}^2 Q_{hx}^2 + g Q_{3z}^2 Q_{hx}^2.$$
\[ + \sum_{k=1}^{3} f_k Q_{1z}^k 2Q_{1x}^{l-k} Q_{hx}^k + \sum_{k=1}^{5} h_k Q_{1z}^{6-k} Q_{hx}^k \]
\[ + \sum_{k=1}^{3} f_k Q_{1z}^{l-k} Q_{hx}^k + \sum_{k=1}^{3} h_k Q_{1z}^{6-k} Q_{hx}^k. \quad (A1) \]

Values of the main coefficients are in Table I (we also give corresponding coefficients for LCO).

**APPENDIX B: STRONG RESONANT DRIVING: CHAOTIC DYNAMICS**

Here we present illustrative examples of large-amplitude driving, clarifying the absence of the transverse mode $Q_{1z}$ response even at very strong driving in case it is done at fixed frequency (see Fig. 7). Strong drive here causes high-frequency modes to oscillate chaotically; as a result the amplitude of the $Q_{1z}$ mode does not reach the threshold for triggering the instability of the transverse mode.

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