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Efficient excitation of nonlinear phonons via chirped mid-infrared pulses: induced structural phase transitions

A.P. Itin\textsuperscript{1,2} and M.I. Katsnelson\textsuperscript{3}

\textsuperscript{1}Radboud University, Nijmegen, The Netherlands, \textsuperscript{2}Space Research Institute, Moscow, Russia

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 material such as KTaO\textsubscript{3}, and inducing structural deformations in cuprates (e.g. La\textsubscript{2}CuO\textsubscript{4}) [A. Subedi et.al, Phys.Rev. B 89, 220301 (2014), Phys.Rev. B 95, 134113 (2017)]. High-frequency phonon modes are driven by mid-infrared pulses, and coupled to lower-frequency modes those indirect excitation causes structural deformations. We study in a more detail the case of KTaO\textsubscript{3} without strain, where (at first glance) it was not possible to excite the needed low frequency phonon mode by resonant driving of the higher frequency one. Behaviour 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. In order to induce ferroelectricity we employ driving with sweeping frequency, realizing so called capture into resonance. The method works for realistic femtosecond pulses and can be applied to many other related systems.

Research in ultrafast light-control of materials has attracted a lot of interest recently [1–9]. Intense mid-infrared pulses have been used to directly control the dynamical degrees of freedom of the crystal lattice [5–9], 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 which then excites required modes not easily accessible by direct drive by means of nonlinear couplings. Here we suggest to add a new useful tool to the arsenal of nonlinear phononics: capture into a resonance. Such phenomenon is encountered in classical and celestial mechanics [10–12] and was employed, e.g. in plasma and accelerator physics [13–17]. 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 to sustain resonance relation between the drive and the system [18].

We find that in perovskite paraelectric KTaO\textsubscript{3} (being described below), as well as in other systems (like La\textsubscript{2}CuO\textsubscript{4} cuprate (LCO)), it is possible to considerably excite a high-frequency phonon mode using a protocol based on capture into resonance, which requires a pulse with chirped frequency. This excitation makes a coupled low-frequency phonon mode dynamically unstable. Pulses with frequency chirps on picosecond timescale have been generated e.g. in FELIX [19].

In all considered examples below, a phonon mode with (at least) quartic nonlinearity is driven by a laser pulse, creating effective potential as a function of 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) $\mathcal{H} = \delta\Omega J + \frac{2}{3} c_4 J^2 - \frac{1}{2} F_0 \sqrt{2J/\Omega_0} \cos \gamma$. Introducing now $x = \sqrt{2J} \sin \gamma$, $y = \sqrt{2J} \cos \gamma$, we get an effective Hamiltonian.

$$P = \sqrt{2J/\Omega_0} \cos \phi, \quad Q = \sqrt{2J/\Omega_0} \sin \phi$$

we then make a transformation to the resonance phase $\gamma = \phi - \Phi$ 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) $\mathcal{H} = \delta\Omega J + \frac{2}{3} c_4 J^2 - \frac{1}{2} F_0 \sqrt{2J/\Omega_0} \cos \gamma$. Introducing now $x = \sqrt{2J} \sin \gamma$, $y = \sqrt{2J} \cos \gamma$, we get an effective Hamiltonian.
Hamiltonian $H = \frac{3}{2} \mu c_{4y} (x^2 + y^2)^2 + \frac{\Delta}{\Omega} (x^2 + y^2) - \frac{F_0}{2 \sqrt{10} \gamma_2}$. Upon rescaling $H \rightarrow H/\frac{3}{2} \mu c_{4y}$, $t \rightarrow t \frac{\mu c_{4y}}{\frac{\Delta}{\Omega}}$ and introduction of $\lambda = -\frac{\Delta}{3} \frac{3}{\mu c_{4y}}$, $\mu = -\frac{F_0}{2 \sqrt{10} \gamma_2}$ 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 [12]. 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 on Fig. 1. A bifurcation happens at $\lambda_c = \frac{3}{2} \mu^{2/3}$. 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 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. 1b, upper curve). In contrast, a pulse with sweeping frequency can shift the equilibrium far away (Fig. 1c, bottom curve). In the adiabatic approximation, dynamics can be described in a great detail [12, 13]. E.g., as the parameters of the system are changing, phase space area within the trajectory remains approximate adiabatic invariant, and from behaviour 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 non-adiabaticity and dissipation become very important, nevertheless qualitative understanding of dynamics allows to construct simple and effective protocol for phonon excitation. With dissipation, the equation of motion becomes

$$\dot{Q} + \gamma \ddot{Q} + \Omega_0^2 Q + 4c_4 Q^3 = F_0 \sin \Phi(t),$$  

(2)

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

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

(3)

Solutions of Eq. (3) are shown on Fig. 1k for different values of the driving amplitude $F_0$. They are dissipative counterparts of fixed points in phase portraits of 1i. One can see that driving frequency higher than the resonant one allows 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 $T_{1u}$ [2]). The remaining one is optically inactive ( $T_{2g}$). Ferroelectricity is related to dynamical instability of an infrared-active transverse optic phonon mode: most ferroelectric materials show a characteristic softening of an infrared transverse optic mode as the transition temperature is approached. In [2] it was investigated if a similar softening and instability of the lowest frequency $T_{1u}$ mode can be achieved.
Damping rates are 5% of the linear frequencies. Instantaneous maximum of the pulse. $\sigma = 2$ ps. $a = 13.2$ (d) Mode $Q_2$ at $z = 0$ corresponds to the ferroelectric state case (without stress). The calculated in [2] phonon frequencies are $\Omega_1 = 85$ cm$^{-1}$ and $\Omega_2 = 533$ cm$^{-1}$ for the lowest and highest frequency $T_{1u}$ modes, respectively ($\Omega_0$ from previous discussion plays the role of $\Omega$ now). Following [2] we simplify 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 $T_{1u}$ modes along the longitudinal $Q_{lx}$ and transverse $Q_{lz}$ coordinates. Dynamics along the second transverse coordinate $Q_{ly}$ is neglected. The energy surface $V(Q_{hx}, Q_{lz}, Q_{lx})$ has a complicated form with many kinds of nonlinear couplings and anharmonicities: $V(Q_{hx}, Q_{lz}, Q_{lx}) = \frac{\Omega_1^2}{2}(Q_{lx}^2 + Q_{lz}^2) + \frac{\Omega_2^2}{2}Q_{hx}^2 + V^{nl}(Q_{hx}, Q_{lx}, Q_{lz})$, where $V^{nl}$ is the nonlinear part of the energy, obtained in state-of-the-art calculations of [2], and given in [20] for convenience. Equations of motions are:

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

$$\ddot{Q}_{lj} + \gamma_l Q_{lj} + \Omega_{lj}^2 Q_{lj} = -\frac{\partial V^{nl}(Q_{hx}, Q_{lx}, Q_{lz})}{\partial Q_{lj}},$$

where $j = x, z$; $\gamma_h$ and $\gamma_l$ are damping coefficients (typically few percents of the corresponding harmonic frequencies), external force $F(t) = Z_{hx}^e E_0 \sin(\Omega t)e^{-t^2/2\sigma^2}$, $Z_{hx}^e$ is the effective charge and $\Omega$ is the driving frequency.

Qualitative understanding of possible dynamics can be captured by drawing a projection of the potential energy $V(Q_{lx}, Q_{lx}, Q_{hx})$ by the plane $Q_{lx} = 0$ (Fig. 3 of [2]). Resulting curves $V(Q_{hx})$ at fixed values of $Q_{hx}$ have single-well form (at small absolute values of $Q_{hx}$), or a double well form (at larger $Q_{hx}$). In the latter case, induced ferroelectricity is possible, as finite value of $Q_{lz}$ at equilibrium corresponds to the ferroelectric

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 (being discussed below), however with addition of internal stress which modifies the crystal lattice such mechanism worked. Let us consider in a more detail the cubic structure case (without stress). The calculated in [2] phonon frequencies are $\Omega_1 = 85$ cm$^{-1}$ and $\Omega_2 = 533$ cm$^{-1}$ for the lowest and highest frequency $T_{1u}$ modes, respectively ($\Omega_0$ from previous discussion plays the role of $\Omega$ now). Following [2] we simplify 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 $T_{1u}$ modes along the longitudinal $Q_{lx}$ and transverse $Q_{lz}$ coordinates. Dynamics along the second transverse coordinate $Q_{ly}$ is neglected. The energy surface $V(Q_{hx}, Q_{lz}, Q_{lx})$ has a complicated form with many kinds of nonlinear couplings and anharmonicities: $V(Q_{hx}, Q_{lz}, Q_{lx}) = \frac{\Omega_1^2}{2}(Q_{lx}^2 + Q_{lz}^2) + \frac{\Omega_2^2}{2}Q_{hx}^2 + V^{nl}(Q_{hx}, Q_{lx}, Q_{lz})$, where $V^{nl}$ is the nonlinear part of the energy, obtained in state-of-the-art calculations of [2], and given in [20] for convenience. Equations of motions are:

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$$\ddot{Q}_{lj} + \gamma_l Q_{lj} + \Omega_{lj}^2 Q_{lj} = -\frac{\partial V^{nl}(Q_{hx}, Q_{lx}, Q_{lz})}{\partial Q_{lj}},$$

where $j = x, z$; $\gamma_h$ and $\gamma_l$ are damping coefficients (typically few percents of the corresponding harmonic frequencies), external force $F(t) = Z_{hx}^e E_0 \sin(\Omega t)e^{-t^2/2\sigma^2}$, $Z_{hx}^e$ is the effective charge and $\Omega$ is the driving frequency. Qualitative understanding of possible dynamics can be captured by drawing a projection of the potential energy $V(Q_{lx}, Q_{lx}, Q_{hx})$ by the plane $Q_{lx} = 0$ (Fig. 3 of [2]). Resulting curves $V(Q_{hx})$ at fixed values of $Q_{hx}$ have single-well form (at small absolute values of $Q_{hx}$), or a double well form (at larger $Q_{hx}$). In the latter case, induced ferroelectricity is possible, as finite value of $Q_{lz}$ at equilibrium corresponds to the ferroelectric
phase. However, to reach such a state dynamically is a nontrivial issue. Excitation of \(Q_{hx}\) 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 \(\Omega_h\), the transverse mode \(Q_{1x}\) remains almost unaffected (Fig. 2) due to insufficient amplitude of the \(Q_{hx}\) mode. In \(\Omega\), the amplitude of driving was varied in a large range, up to pump amplitudes of 100 MIV/cm\(^{-1}\), with no signs of dynamical instability in \(Q_{1z}\). Increase of the pump amplitude makes the dynamics of \(Q_{hx}, Q_{1x}\) chaotic, but does not result in noticeable response of the \(Q_{1z}\) mode: at strong driving the 'axuilary' longitudinal mode \(Q_{1z}\) becomes excited (20), and chaotic dynamics prevents efficient excitation of \(Q_{hx}\). There is a range of driving frequencies (away from the exact resonance with \(\Omega_h\)), 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 \(\Omega_h\) 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. 3d). From the full potential energy of the system, we can single out the potential felt by the slow mode is \(G(t) = \sqrt{2}Q_{hx}^2 + Q_{1x}^2 + pQ_{1z}^2 + c_2Q_{hx}^2 + c_3Q_{hx}^2 + c_4Q_{hx}^2\) (20)). Due to strong coupling between \(Q_{hx}\) and \(Q_{1x}\) modes, they oscillate synchronously although their linear amplitudes are almost unaffected (Fig. (2)) due to insufficient ampli-

dics. Damping parameters typically are 5-10% of corresponding linear frequencies. In case we assume higher damping for low-frequency phonons, the auxiliary longitudinal mode \(Q_{1z}\) is not excited considerably, and dynamics can be understood from the driven Duffing oscillator model for the \(Q_{hx}\) mode alone (with small corrections from \(Q_{1x}\)). 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 \(A_{tip} = \frac{F_0}{m_0}, \omega_{tip} = 1 + \frac{3c_4F_0^2}{\gamma_0^2}\). 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 \(\alpha = \frac{3c_4F_0^2}{\gamma_0^2}\). We make numerical experiments with various sweeping rates and amplitudes of driving (see Fig 3b,f), and find a remarkable improvement in efficiency of excitation compared to pulses with constant frequency. Most exciting, the protocol with sweeping frequency works also for much shorter, sub-picosecond pulses. We show in Fig 4 an example with \(\sigma = 350\) fs, which corresponds to laser parameters used in A.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 infrared-active \(Q_{IR}\) mode (described by a driven Duffing oscillator) coupled to a Raman mode \(Q_R\) by a quadratic term. \(Q_R (B_{1g}(18))\) mode describes in-plane rotations of CuO\(_6\) octahedra, whereas \(Q_{IR}\) mode describes in-plane stretching of Cu-O bond. The energy surface has the form \(V = \Omega_{IR}^2Q_{IR}^2 + \Omega_R^2Q_R^2 + c_4Q_{IR}^4 + b_4Q_{IR}^4 - \frac{4}{\sigma}Q_{IR}Q_R^2\). Values of the coefficients were derived in elaborate calculations of \([\text{int}]\). There is a single-potential well around the equilibrium value for \(Q_R\) mode at small amplitudes of \(Q_{IR}\), which becomes double-well potential at larger amplitudes of \(Q_{IR}\) (instantaneous quadratic potential felt by the slow mode is \(\frac{1}{\sigma}Q_{IR}^2 - gQ_{IR}^2\)), which becomes inverted parabolic potential for sufficiently high amplitudes of the driven \(IR\) mode. The critical value of driving force \(F_c\) depends on detuning \(\delta \Omega \equiv \Omega - \Omega_0\) and can be made smaller than its value on the resonance (being used in [\text{int}]). Indeed, to the first approximation, the averaged potential for the slow mode is \(\frac{1}{2\sigma}Q_{IR}^2 (\Omega^2 - gQ_{IR}^2)\) and becomes unstable at critical value of the fast mode amplitude \(Q_{IR,max} = \Omega_1 \sqrt{2/(2g)}\). This can be achieved at sufficiently smaller driving force amplitudes provided sweeping frequency pulse is used. We show corresponding results of numerical calculations in Fig. 6(c). Fig. 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. Recently there has been a lot of interest in effective models arising from periodic driving, and the suggested method can be useful for this area of research as well. The proposed method can be useful also for recent proposals and experiments on driven orthorombic perovskites (like ErFeO$_3$, see [28, 29]), where three-linear phonon coupling is realized: two high-frequency infrared-active modes are coupled to the third, Raman mode.

To conclude, we demonstrate that drastic improvement in efficiency of excitation of nonlinear phonons can be achieved using 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 low-frequency modes, and corresponding phase transitions. 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, including induced ferroelectricity in perovskites, induced structural transitions in LCO cuprates, and excitation of orthorombic perovskites.

We are grateful to A.I.Neishtadt and A.Cavalleri for insightful discussions. The work was supported by NWO via Spinoza prize and by European Research Council (ERC) Advanced Grant No. 338957 FEMTO/NANO.
Supplemental Materials: Efficient excitation of nonlinear phonons via chirped mid-infrared pulses: induced structural phase transitions

A. Nonlinear part of the potential energy surface for KTaO₃

The nonlinear part of the potential energy surface is:

\[
V^{nl}(Q_{hx}, Q_{1z}, Q_{1x}) = \sum_{k=2}^{6} \left(a_{2k}(Q_{1z}^{2k} + Q_{hx}^{2k}) + n_1 Q_{1z}^{2} Q_{hx}^{2} + n_2 Q_{1z}^{6} Q_{hx}^{2} + n_3 Q_{1z}^{4} Q_{hx}^{4} \right)
\]

Values of the main coefficients are:

<table>
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<th>Term</th>
<th>KTaO₃ Coefficient</th>
<th>Term</th>
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<td>(g)</td>
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<td>(c_4)</td>
<td>(Q_{1R}^4)</td>
</tr>
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</table>

B. Strong resonant driving: chaotic dynamics

FIG. S1: Resonant driving, the same as in Fig. [2], but with a larger driving amplitude. From left to right: \(Q_{1z}, Q_{1z}, Q_{hx}\) modes, and \(E(t)\) pulse. \(E_0 = 60\) MV cm\(^{-1}\). Amplitude of driving is so large that dynamics become chaotic, nevertheless the mode \(Q_{1z}\) is unexcited.
C. Two coupled modes with periodic driving

Although the linear frequency of the $Q_{1x}$ mode is much smaller than that of $Q_{hz}$, strong coupling between them causes these modes to oscillate synchronously. One can therefore introduce an effective system combining these modes together and averaging over the driving frequency. The third mode, $Q_{1z}$ can be neglected until it becomes excited due to creation of the effective double-well potential. Consider firstly a conservative system, without dissipation.

The reduced two-mode Hamiltonian is (we denote $Q_1 \equiv Q_{1x}, Q_h \equiv Q_{hz}$)

$$H = \frac{P_h^2}{2} + \frac{P_{Q_h}^2}{2} + \Omega_1^2 Q_1^2 + \Omega_h^2 Q_h^2 + a_4 Q_1^4 + c_4 Q_h^4 +$$

$$+ t_1 Q_1^3 Q_h + t_3 Q_1^3 Q_h^3 + t_3 Q_1 Q_h^3 + \sum_{k=1}^{5} u_k Q_1^{2-k} Q_h^k +$$

$$- Q_h F_0 \sin \Omega t$$  \hspace{1cm} (S2)

Moreover, we neglect the $u-$terms in the analytical considerations below, as values of $t-$coefficients are considerably higher.

Introducing radial coordinates ($I_h, \phi_h, I_1, \phi_1$)

$$Q_{hx} = \sqrt{\frac{2I_h}{\Omega_h}} \sin \phi_h, \quad P_h = \sqrt{2I_h \Omega_h} \cos \phi_h,$$

$$Q_{1x} = \sqrt{\frac{2I_1}{\Omega_1}} \sin \phi_1, \quad P_1 = \sqrt{2I_1 \Omega_1} \cos \phi_1,$$  \hspace{1cm} (S3)

we then switch to rotating phases $\gamma_h - \Omega t, \gamma_1 - \Omega t$ by means of a generating function

$$W = \rho_h (\phi_h - \Phi) + \rho_1 (\phi_1 - \Phi), \Phi \equiv \Omega t$$  \hspace{1cm} (S4)

Averaging the resulting Hamiltonian over the explicit time dependence, we get as an effective reduced two-mode Hamiltonian

$$H = \rho_h (\Omega_h - \Omega) + \rho_1 (\Omega_1 - \Omega) + \frac{3}{2} c_4 \rho_h^2 \Omega_h^2 + \frac{3}{2} a_4 \rho_1^2 \Omega_1^2 +$$

$$+ \frac{F_0}{2} \sqrt{\frac{2\rho_h}{\Omega_h}} \sin \gamma_h + \frac{3}{2} t_1 \left( \frac{\rho_h}{\Omega_h} \right)^{3/2} \left( \frac{\rho_h}{\Omega_h} \right)^{1/2} \cos (\gamma_1 - \gamma_h)$$

$$+ \frac{3}{2} t_3 \left( \frac{\rho_h}{\Omega_1} \right)^{1/2} \left( \frac{\rho_h}{\Omega_h} \right)^{3/2} \cos (\gamma_1 - \gamma_h) +$$

$$+ \frac{1}{2} t_3 \left( \frac{\rho_h}{\Omega_1} \right)^{1/2} \left( \frac{\rho_h}{\Omega_h} \right)^{3/2} (2 + \cos 2(\gamma_1 - \gamma_h)) + u-term + ...$$  \hspace{1cm} (S5)

We then return to cartesian coordinates via $x_k = \sqrt{\frac{\Omega_k}{\Omega_h}} \cos \phi_k$, $y_k = \sqrt{2\rho_h \Omega_k} \sin \phi_k$, and search for equilibria of the resulted two-mode system.

We get two coupled algebraic equations for coordinates of the equilibrium:

$$\frac{3}{2} c_4 X_h^3 + X_h \left( \Omega_h^2 (1 - x) + \frac{3}{4} t_2 X_1^2 \right) + \frac{9}{8} t_3 X_1 X_h^2$$

$$- \frac{3}{8} t_1 X_1^2 = \frac{F_0}{2},$$

$$X_1 \left( \Omega_1^2 (1 - x \Omega_h \Omega_1) + \frac{3}{4} t_2 X_h^2 \right) + \frac{9}{8} t_1 X_1^2 X_h$$

$$+ \frac{3}{2} a_4 X_1^3 - \frac{3}{8} t_3 X_h^3 = 0$$  \hspace{1cm} (S6)

As amplitude of $X_h$ grows beyond certain critical value, deviation of $X_1$ from 0 becomes considerable. Then, dynamics of the system becomes complicated and the first mode stops to absorb the energy from the drive. Note
FIG. S2: Steady states of the mode $X_h$ of the driven reduced two-mode system (S6) without dissipation as a function of frequency $x \equiv \Omega / \Omega_{h_x}$ at several values of the driving force amplitude $E_0$. From bottom to top: $E_0 = 5, 10, 15, 20, 25, 30$ MeV cm$^{-1}$. (b) Steady values of amplitude of the mode $Q_{1x}$ (i.e., $X_1$) as a function of $X_h, x$. White areas correspond to highest values of $X_1$.

that the steady state value of $X_h$ in Eq. (S6) is a function of $F_0$ and depends on $X_1$, while $X_1$ is determined from the second equation which do not depends on $F_0$. So we could depict a value of $X_1$ as a density plot on the plane ($\Omega, X_h$): see Fig. (S2b). It is clearly seen that as one tries to increase steady amplitude $X_h$, above certain curve on ($\Omega, X_h$) plane the mode $Q_{1x}$ becomes excited. Importantly, detuning from the exact resonance to higher frequencies allows to reach higher values of $X_h$ without considerable excitation of the $Q_{1x}$ mode. This important qualitative result remains valid in the full system. When we take into account dissipation and the remaining nonlinear coupling terms, the critical curve goes higher, and it become possible to reach sufficiently high values of $X_h$ (again, shifting from the exact resonant to the higher frequencies allows higher values of $X_h$).