

Dynamical Breaking of Inversion Symmetry, Strong Second Harmonic Generation, and Nonequilibrium Ferroelectricity with Nonlinear Phonons

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(Received 24 October 2025; revised 12 March 2026; accepted 9 July 2026; published 4 September 2026)

We show how crystalline inversion symmetry can be dynamically broken by optical phonons with generic, hardening Kerr-like nonlinearities. The symmetry-broken state is reached through a parametric instability that can be accessed by driving close to half the phonon frequency. The system then settles to a steady state with inversion symmetry breaking phonon trajectories and strong second harmonic generation. The time averaged positions of the atoms are displaced relative to equilibrium, resulting in a ferroelectric rectification of the driving signal. For circularly polarized phonons, complex Lissajous-like trajectories, resulting in structured magnetic fields within a unit cell, can be achieved.

DOI: [10.1103/lwpl-brrr](https://doi.org/10.1103/lwpl-brrr)

Introduction—The nonequilibrium behavior of condensed matter systems currently attracts considerable attention due to its potential for on-demand control over materials [1–3]. Out-of-equilibrium phonons are of particular interest as lattice distortions have an immediate impact on the electronic properties of solids [4,5]. Nonlinear lattice dynamics can be exploited to enhance and manipulate superconductivity [6–10], magnetism [11–14], and other states of matter [15–17], including ferroelectricity [18,19]. Other effects predicted for driven, nonlinear collective modes include nontrivial steady states [16,17,20–24], chaos [25], and parametric amplification [26–28].

Here, we show how IR-active phonons oscillating in anharmonic interatomic potentials can be driven in unconventional ways to break lattice symmetries and generate symmetry forbidden responses. Inversion symmetry is known to prohibit second harmonic generation (SHG) and rectification [29]. We demonstrate that, for a generic, hardening Kerr-like nonlinearity, this rule can be circumvented by a tailored driving protocol, resulting in a symmetry-breaking steady state with strong phononic SHG and rectification. We stress that the phonon potential is a single well, and that the effects appear in the absence of any metastable equilibrium ferroelectric states.

A static displacement of atoms from their equilibrium positions is generated, leading to a constant-in-time dipolar electric field. This behavior is analogous to that of an

equilibrium ferroelectric, where—below a critical temperature—dipolar fields form due to spontaneous, symmetry-breaking lattice deformations. In our case, the dipolar response is triggered by a dynamical breaking of inversion symmetry, and we argue that the predicted effect can be used to create an on-demand ferroelectric. In particular, we show that the nonequilibrium ferroelectric response exhibits hysteretic behavior, which underlines that the effect is not merely a nonlinear piezoelectric response [30] but instead is a symmetry-breaking out-of-equilibrium phase. We show that this phase is robust to noise and is accessible at reasonable field strengths.

We then explore the above effects in a system of circularly polarized phonons [31], which are sometimes referred to as chiral phonons [32] and have been used to manipulate magnetic states. Here, the strong SHG leads to Lissajous-like, non-inversion-symmetric phonon trajectories. Such trajectories create structured magnetic fields influencing the dynamics of electrons and spins on a microscopic level [33–35] and provide an interesting way of engineering electronic behavior [36].

Symmetry-breaking instability and second harmonic generation—To show how the symmetry breaking instability emerges, we first focus on a single IR-active phonon mode $Q_x(t)$. For a generic, inversion symmetric system, the dynamics of $Q_x(t)$ is governed by the Hamiltonian

$$H = \frac{1}{2}P_x^2 + \frac{\Omega_0^2}{2}Q_x^2 + \frac{\beta}{4}Q_x^4 + V_{\text{l-m}}. \quad (1)$$

Here, Q_x is given in units of $\text{\AA}\sqrt{u}$, where u is the atomic mass unit, Ω_0 is the resonance frequency of the phonon, and $\beta > 0$ controls the strength of the nonlinearity. Notice that, for positive β , unlike for $\beta < 0$, the lattice potential has no metastable minima at $Q_x \neq 0$. This underlines the truly

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dynamical nature of the symmetry breaking described in the following. Finally, V_{l-m} is a the dipolar coupling between the phonon and an electromagnetic field:

$$V_{l-m} = -\mathbf{E} \cdot \mathbf{p}_x, \quad (2)$$

where the electric dipole moment of the phonon is given by $\mathbf{p}_x = \mathbf{Z}_x Q_x$, with effective charge $\mathbf{Z}_x$. We assume $\mathbf{Z}_x \propto \hat{\mathbf{e}}_x$. Then, for circularly polarized light, the phonon equation of motion reads as

$$\ddot{Q}_x + 2\gamma\dot{Q}_x + \Omega_0^2 Q_x + \beta Q_x^3 = Z_x E_x \cos \omega t, \quad (3)$$

where we include a damping term with damping rate γ .

It is often assumed that the third order nonlinearity of Eq. (3) does not generate even harmonics of the driving frequency ω . Indeed, shifting the time coordinate according to $t \rightarrow t + (\pi/\omega)$ changes the sign of the right side, which seems to enforce the symmetry $Q_x[t + (\pi/\omega)] = -Q_x(t)$, excluding any response at frequencies $2n\omega$, where $n \in \mathbb{N}$, including zero. We now show that this symmetry, stemming from the inversion symmetry of the Hamiltonian (1), can be dynamically broken.

It is useful to divide $Q_x(t)$ into parts composed of odd and even harmonics:

$$Q_x(t) = Q_{x,\text{odd}}(t) + Q_{x,\text{even}}(t), \quad (4)$$

which are, respectively, antisymmetric and symmetric under a time translation by half the oscillation period of the electromagnetic field:

$$Q_{x,\text{even/odd}}\left(t + \frac{\pi}{\omega}\right) = \pm Q_{x,\text{even/odd}}(t). \quad (5)$$

We now show how even harmonics ($Q_{i,\text{even}}$) can be created via a parametric instability. Let us first study the onset of this instability. Using the decomposition of Eq. (4), we can separate Eq. (D1) into coupled equations for $Q_{x,\text{odd}}$ and $Q_{x,\text{even}}$. We use that, at the onset of the instability, $Q_{i,\text{even}}$ will be very small, such that $|Q_{i,\text{even}}| \ll |Q_{i,\text{odd}}|$. Then, the equation for the odd part, neglecting contributions stemming from $Q_{i,\text{even}}$, reads as

$$\ddot{Q}_{x,\text{odd}} + 2\gamma\dot{Q}_{x,\text{odd}} + \Omega_0^2 Q_{x,\text{odd}} + \alpha Q_{x,\text{odd}}^3 = Z_x E_x \cos(\omega t). \quad (6)$$

For our purposes it is sufficient to approximate the response $Q_{x,\text{odd}}$ with the fundamental harmonic and write

$$Q_{x,\text{odd}} \approx F_{x,1}(E_x) \cos(\omega t + \varphi_x). \quad (7)$$

$F_{x,1}(E_x)$ is then found by inverting the amplitude equation

$$F_{x,1}^2 \left[4\gamma^2 \omega^2 + \left((\omega^2 - \Omega_0^2) - \frac{3}{4} \beta F_{x,1}^2 \right)^2 \right] = Z_x^2 E_x^2. \quad (8)$$

For the even component $Q_{x,\text{even}}$, we find the Mathieu equation,

$$\ddot{Q}_{x,\text{even}} + 2\gamma\dot{Q}_{x,\text{even}} + \tilde{\Omega}_0^2(E_x)[1 + h(E_x) \cos(2\omega t + 2\varphi_x)]Q_{x,\text{even}} = 0, \quad (9)$$

where $\tilde{\Omega}_0(E_x)$ is an effective, amplitude dependent resonance frequency [see Fig. 1(a)] given by

$$\tilde{\Omega}_0(E_x) = \Omega_0 \sqrt{1 + \frac{3\beta}{2\Omega_0^2} F_{x,1}^2(E_x)} \quad (10)$$

and $h(E_x) = 3\alpha F_{x,1}^2(E_x) / [2\tilde{\Omega}_0^2(E_x)]$. We note that Eq. (9) contains the phase φ_x not present in the canonical definition (see, e.g., Ref. [37]). We use Eq. (7) to approximate $Q_{x,\text{odd}}^2$. It is then the constant-in-time part of $Q_{x,\text{odd}}^2$ that modifies the resonance frequency of the mode and leads to a blueshift, while the oscillating part of $Q_{x,\text{odd}}^2$ acts as a parametric driving for $Q_{x,\text{even}}$. The Mathieu equation (D2) is known to exhibit parametric instabilities for $\tilde{\Omega}_0(E_x) = n\omega$, with n being a positive integer [38]. However, $Q_{x,\text{even}}$ has to obey Eq. (5), which excludes the $n = 1$ resonance. The $n = 2$ resonance, however, is allowed and leads to the symmetry-breaking instability that we want to study. Here $\tilde{\Omega}_0(E_x) = 2\omega$, such that, for driving slightly above half the original resonance frequency of Ω_0 , we expect a response at $\tilde{\Omega}_0(E_x)$ —i.e., we expect strong SHG.

As is typical for parametric resonances, the instability occurs in a small frequency window where for $\Delta = 2\omega - \tilde{\Omega}_0$ holds (see, e.g., Ref. [37], p. 394 [39])

$$\begin{aligned} \frac{\tilde{\Omega}_0}{24} \left[3 \left(\frac{4\gamma^2}{\tilde{\Omega}_0^2} - \sqrt{h^4 - \frac{64\gamma^2}{\tilde{\Omega}_0^2}} \right) + 2h^2 \right] &< \Delta \\ &< \frac{1}{24} \tilde{\Omega}_0 \left[3 \left(\frac{4\gamma^2}{\tilde{\Omega}_0^2} + \sqrt{h^4 - \frac{64\gamma^2}{\tilde{\Omega}_0^2}} \right) + 2h^2 \right]. \end{aligned} \quad (11)$$

Here, and in what follows, we omit writing out the dependence of $F_{x,1}$, $\tilde{\Omega}_0$, h , and Δ explicitly. The blueshift and the instability window are shown and compared to the results of a numerical simulation in Fig. 1(a) (see Appendix A for a note on the initial conditions used). The full Eq. (3) was used for the simulation.

To overcome damping effects, a minimal driving amplitude is required. This threshold amplitude $E_{x,*}$ can be calculated by setting $\Delta = 0$. To leading order in γ/Ω , we find $h(E_{x,*}) = \sqrt{8\gamma/\Omega_0}$ and invert for E_x using Eq. (8). For

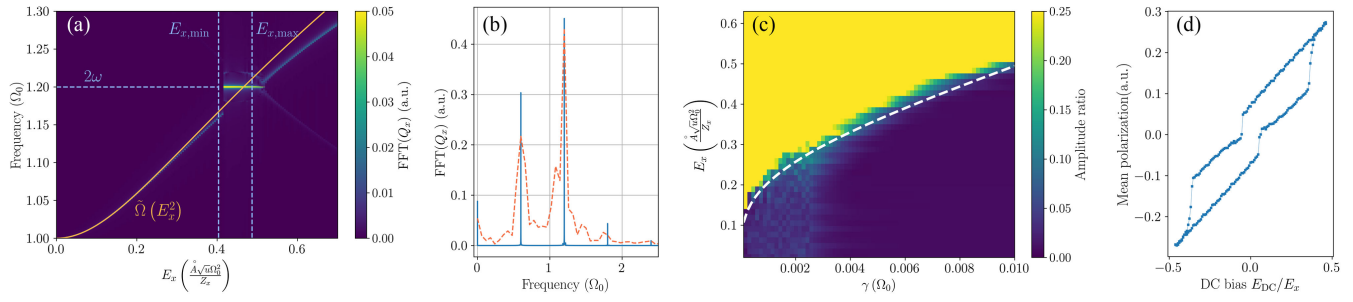


FIG. 1. (a) Phonons [Eq. (18)] driven with a linearly polarized electric field $E_x \cos(\omega t)$ oscillating at frequency $\omega = 0.6\Omega_0$. The effective phonon frequency $\tilde{\Omega}_0(E_x)$, given in Eq. (10), exhibits a blueshift as E_x is increased. Around $\tilde{\Omega}_0(E_x) \approx 2\omega$, the system enters the symmetry breaking state with strong second harmonic generation. In the symmetry-breaking regime, the resonance curve $\tilde{\Omega}_0(E_x)$ is interrupted. A small damping of $\gamma = 0.001$ was chosen to have a large instability interval for clarity. (b) Spectrum of $Q_x(t)$ in the symmetry breaking state. Dashed lines show the spectrum when the system is driven by a short pulse (see Appendix A). (c) Amplitude ratio of second and first harmonics across the symmetry breaking transition for different damping γ . The dashed line indicate the result of Eq. (12). (d) Hysteresis of the mean polarization $p_x = Z_x F_{x,0} = (2\pi/\omega) \int_0^{2\pi/\omega} dt Q_x(t)$ when a static bias field E_{dc} is swept through positive and negative values in a triangle-wave-like manner.

small damping, the threshold electric field amplitude is then given by

$$E_{x,*} \approx \frac{\sqrt{3}\Omega_0^3}{2^{3/4}\sqrt{\beta}Z_x} \left(\frac{\gamma}{\Omega_0}\right)^{1/4}. \quad (12)$$

Notice that Ω_0 is the bare resonance frequency, but the blueshift has been taken into account in the derivation of Eq. (12). As can be expected, $E_{x,*}$ is lowered by a strong nonlinearity β and increased by a larger γ . The result of Eq. (12) is confirmed by numerical simulations as shown in Fig. 1(c). The threshold fields for different (bare) phonon resonance frequencies Ω_0 are given in Table I. Since $E_{x,*} \sim \Omega_0^3$ holds, much stronger amplitudes are required at higher frequencies. The amplitudes are, however, in an experimentally accessible range.

Symmetry breaking steady state and pulsed excitation—The above analysis demonstrates that the usual solution, consisting of a strong response at the fundamental harmonic and perturbative corrections at odd harmonics, is unstable if driving frequency and strength are tuned such that the condition of Eq. (11) at $2\omega \approx \tilde{\Omega}_0(E_x)$ is fulfilled. This instability is associated with strong second harmonic generation due to the parametric resonance in Eq. (D2). Investigating the dynamics of the nonlinear phonon mode in this regime, we find that the system converges to a stable steady state which breaks the inversion symmetry of the system. Besides a strong second harmonic, this symmetry-breaking is characterized by a relatively large dc offset. The spectrum of Q_x in the steady state, obtained by solving Eq. (3) numerically, is shown in Fig. 1(b). We note in passing that the instability and steady state studied here have been described in nonlinear systems literature, although the only extensive study, to our knowledge, is presented in Ref. [41].

For low frequency phonons the instability is accessible already at relatively small driving fields (see Table I), which might be produced by continuous wave lasers. However, reaching the symmetry breaking with pulsed signals is also possible. We performed a series of numerical experiments using the same parameters as in the continuous wave case and find that short pulses with as few as five oscillation cycles (see Appendix A) are sufficient to observe both second harmonic generation and a dc offset. However, flat-top pulses must be used because, due to the nonlinear blueshift, the frequency and amplitude of the drive must be tuned together. The dashed lines in Fig. 1(b) show the response spectrum obtained for a flat-top pulse of five optical cycles. The finite duration of the pulse introduces spectral broadening; however, the SHG and the dc offset are clearly visible.

Before extending our results to a system of chiral phonons, we investigate how the dc component of the steady state at $2\omega \approx \tilde{\Omega}_0(E_x)$ leads to a nonequilibrium ferroelectric response and show how auxiliary phonon modes can be exploited to trigger the symmetry breaking instability resonantly.

Nonequilibrium ferroelectricity, hysteresis, stability to noise—We now show that the symmetry-breaking steady state outlined above necessarily implies the presence of a static displacement of the atoms from their equilibrium positions. To see this, we average Eq. (3) over one period of the drive [38]. Writing $Q_x(t) = \sum_a F_{x,a} \cos(a\omega t + \varphi_{x,a})$

TABLE I. Critical driving fields $E_{x,*}$ according to Eq. (12) for the parameters $\gamma/\Omega = 0.05$, $Z_x = e/\sqrt{u}$, and $\beta = 1 \text{ eV u}^{-2} \text{ Å}^{-4}$ [40].

$\Omega_0/2\pi$	1 THz	3 THz	5 THz
$E_{x,*}$	$1.5 \times 10^6 \text{ V/m}$	$4.1 \times 10^7 \text{ V/m}$	$1.9 \times 10^8 \text{ V/m}$

and truncating the series at $a = 2$, we find

$$\omega_0^2 F_{x,0} + \beta \left[\frac{3}{2} (F_{x,1}^2 + F_{x,2}^2) F_{x,0} + \frac{3}{4} F_{x,1}^2 F_{x,2} \cos(2\varphi_{x,1} - \varphi_{x,2}) + F_{x,0}^3 \right]. \quad (13)$$

This equation has one nontrivial, real solution for $F_{x,0}$. To leading order in $F_{x,1}$ and $F_{x,2}$, it reads as

$$F_{x,0} = -\frac{3\beta}{4\omega_0^2} F_{x,1}^2 F_{x,2} \cos(2\varphi_{x,1} - \varphi_{x,2}), \quad (14)$$

showing that any response at the second harmonic is accompanied by a dc offset. Being third order in the first and second harmonic amplitudes, we expect the dc offset to be smaller in magnitude; it can, however, still be sizable [see Fig. 1(b)].

We conclude that although the symmetry breaking instability is triggered by an oscillating driving field, inversion symmetry is statically broken. This results in constant-in-time electric fields produced by the dipoles $\mathbf{p} = \mathbf{Z}_x F_{x,0}$, where $\mathbf{Z}_x$ is the effective electric charge of the phonon mode in question. The driving signal is thus rectified and triggers a ferroelectriclike response.

This dynamically triggered response shares significant similarities with equilibrium ferroelectrics, showing that it can be used as a road to on-demand ferroelectricity. To substantiate this claim, we demonstrate that hysteresis can be observed when the phonon mode is in the symmetry-breaking state. To this end we add a static component E_{dc} to the driving field in Eq. (3) and sweep it through positive and negative values in a triangle-wave-like manner. We numerically integrate Eq. (3) while slowly changing E_{dc} . Figure 1(d) shows the mean polarization,

$$p_x = Z_x F_{x,0} = (2\pi/\omega) \int_0^{2\pi/\omega} dt Q_x(t), \quad (15)$$

which exhibits a hysteresis. Interestingly, the hysteresis curve is pinched—also a feature known from equilibrium ferroelectrics [42].

Finally we want to investigate the sensitivity of the symmetry-breaking steady state to noise. First, a simple estimation can be made. In the steady state, we find $\sqrt{\langle Q^2 \rangle} \sim 1 \text{ Å} \sqrt{u}$ (see, e.g., Fig. 3). We can expect that the steady state breaks down when, roughly, $\Omega_0^2 \langle Q^2 \rangle \approx k_B T$, yielding a threshold temperature of $\sim 430 \text{ K}$ for the 3 THz mode or $\sim 1200 \text{ K}$ for the 5 THz mode. To substantiate this estimate, we perform a simulation of the equation of motion (3) with an additional Langevin term $A\eta(t)$, where $\eta(t)$ is normally distributed noise that is uncorrelated on physically relevant timescales. We indeed find that the steady state collapses above a threshold noise

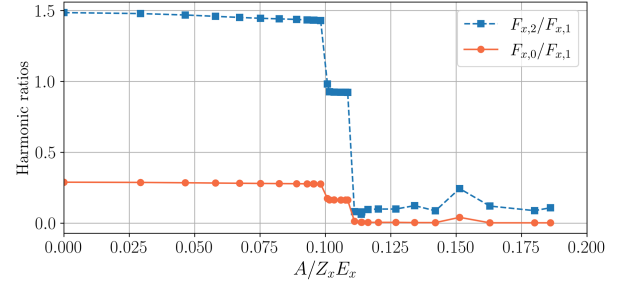


FIG. 2. Collapse of the symmetry-breaking steady state under the influence of noise. In the case of thermal noise and $\Omega_0/2\pi = 3 \text{ THz}$, the critical noise strength $A \approx 0.1 Z_x E_x$ corresponds to a temperature of $\sim 700 \text{ K}$. The same parameters as in Fig. 1 were used.

amplitude (see Fig. 2 and Appendix B). Interestingly, the collapse seems to proceed in steps—an effect that could be related to the pinched hysteresis of Fig. 1(d). Using the parameters $\gamma = 0.1$, $\Omega_0/2\pi = 3 \text{ THz}$, and $E_{x,*} = 4.1 \times 10^7 \text{ V/m}$, we find that this critical noise amplitude corresponds to a temperature of $\sim 700 \text{ K}$. We thus conclude that the symmetry breaking state is reasonably robust to perturbations by noise in general and thermal effects in particular.

Finally, we note that a macroscopic dipole generated by the symmetry breaking will act as a depolarizing field that can influence the dynamics of the system [43,44]. We leave effects like domain formation for future study and simply note here that the demagnetizing field can be screened [43], restoring the dynamics of Eq. (3).

Exploiting auxiliary IR- and Raman-active modes— Auxiliary modes can be used to further simplify reaching the symmetry-breaking steady state. IR-active phonons with frequencies ω_A close to $\Omega_0/2$ can be exploited to resonantly enhance the otherwise off-resonant driving: consider a phonon mode Q_A with a coupling term

$$H_{qQ} = \lambda Q_x Q_A. \quad (16)$$

This coupling preserves the original inversion symmetry of the system and leads to λQ_A taking over the role of the electric field in Eq. (D1). The mode q_A can be pumped resonantly, such that the energy of the drive is stored over $\sim \omega_A/\gamma_A$ cycles, resulting in a large amplitude oscillation of Q_A . Because of the nonlinear blueshift of $\tilde{\Omega}_0$ [Eq. (10)], the frequency of the auxiliary phonon ω_A can be adjusted by choosing a suitable driving amplitude, such that it exactly hits $\omega_A = \tilde{\Omega}_0/2$.

Raman-active modes can be used in a similar way. Here the lowest order, inversion symmetric coupling Hamiltonian is given by

$$H = A Q_R Q_x^2. \quad (17)$$

Q_R is an auxiliary Raman mode. As shown in Appendix C, including the coupling of Eq. (17) again leads to an

equation of motion for Q_x that can be mapped onto the original Eq. (D1). Thus Raman-active modes can, too, be used to drive the system into the symmetry-breaking state. This might be advantageous since driving these modes allows for greater optical penetration depths compared to IR-active modes.

Nonlinear, circularly polarized phonons—We now investigate the effects of dynamical symmetry breaking on circularly polarized phonons. While sometimes such phonons are referred to as chiral [32], this terminology has been questioned [31]. We discuss both degenerate circularly polarized optical phonons [32,45,46] and phonons with split frequencies for right- and left-handed motion [47–50].

A simple model for nonlinear, degenerate, circularly polarized phonons is given by the Hamiltonian [32]

$$H = \frac{P_x^2}{2} + \frac{P_y^2}{2} + \frac{\Omega_0^2}{2} (Q_x^2 + Q_y^2) + \frac{\beta}{4} (Q_x^2 + Q_y^2)^2 + V_{\text{l-m}}. \quad (18)$$

Here, Q_x and Q_y are the coordinates of two orthogonal phonon modes. The light-matter coupling $V_{\text{l-m}}$ is given by

$$V_{\text{l-m}} = -\mathbf{E} \cdot (\mathbf{p}_x + \mathbf{p}_y), \quad (19)$$

where $\mathbf{p}_n = \mathbf{Z}_n Q_n$ are the electric dipole moments of the phonon components, with effective charges $\mathbf{Z}_n$. For simplicity, we assume $\mathbf{Z}_n \propto \hat{\mathbf{e}}_n$. The equations of motion for elliptically polarized light are given in Appendix D, where we added damping at rate γ .

Solving the equations of motion (D1) numerically, we observe that the instability intervals are larger for elliptical polarization with $E_x \neq E_y$. A stability analysis for the system following Ref. [38], for which we refer to Appendix D, rationalizes this observation.

The result of a numerical simulation of the phonon trajectory in the symmetry-breaking state is shown in Fig. 3 for driving with elliptically polarized light where $E_y = 0.25E_x$. The resulting Lissajous trajectory breaks

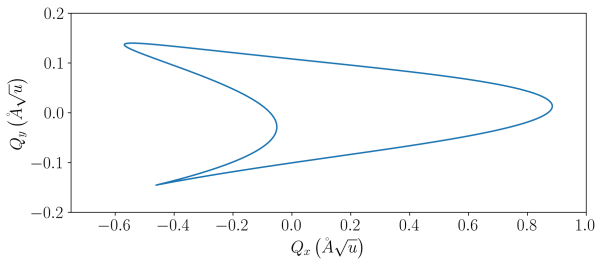


FIG. 3. Lissajous trajectory of phonon coordinates $Q_x(t)$ and $Q_y(t)$ when driven into the symmetry-breaking state using elliptically polarized light with $\mathbf{E} = E_x[\cos(\omega t), 0.25 \sin(\omega t)]$. The inversion symmetry of Eq. (18) is broken dynamically.

the inversion symmetry of the Hamiltonian (18) due to the large second harmonic component of $Q_x(t)$.

We note that, beyond the instability at $\omega \approx \Omega_0/2$, higher order, inversion symmetry breaking instabilities at frequencies $\omega \approx \Omega_0/n$, where n is an even number, can be induced. These generate more complex Lissajous figures; however, the required threshold driving amplitudes grow according to $E_{x,*} \sim \gamma^{1/2n}$ [38].

Finally, we investigate dynamical symmetry breaking for nondegenerate circularly polarized phonons [47] (also called “false chiral phonons” [31]). Such phonons can appear in systems with broken time-reversal symmetry [51] and have been observed in monolayer TMDs [48]. A toy model with split frequencies for phonons of opposite chiralities is obtained by substituting $P_i \rightarrow P_i - \kappa A_i$ in the Hamiltonian of Eq. (18) [52]. Here $\mathbf{A} = B_{\text{eff}}[-Q_y, Q_x, 0]$ takes the role of an effective magnetic vector potential. To linear order in κ , the above substitution is equivalent to adding the term $\kappa \mathbf{B}_{\text{eff}} \cdot \mathbf{L}$ to the Hamiltonian (18), where $\mathbf{L} = (Q_x P_y - Q_y P_x) \hat{\mathbf{e}}_z$ is the phonon angular momentum and $\mathbf{B}_{\text{eff}} = [0, 0, B_{\text{eff}}]$. Solving the linearized equations of motion, we find the phonon eigenfrequencies

$$\Omega_{0,\pm} = \Omega_0 \pm \kappa B_{\text{eff}}, \quad (20)$$

where the $\pm$ -signs correspond to right- and left-handed motion, respectively.

We find that the symmetry-breaking instability described above can also be achieved with nondegenerate chiral phonons and present these results in Fig. 4.

Conclusion—We have described a new, inversion-symmetry-breaking steady state for driven nonlinear phonons. This state is characterized by strong second harmonic generation and by the emergence of ferroelectriclike behavior. These responses—forbidden by the inversion symmetry of the underlying lattice—can serve as sharp experimental signatures of the physics presented in this Letter.

Ferroelectrics above the transition [53–55] are natural platforms for realizing the effects. The softening phonon modes responsible for the transition will have low frequencies and strong anharmonicity [56–58], resulting in low threshold powers $E_{x,*}$ as shown in Eq. (12).

The symmetry-breaking state can be induced with flattop pulses as short as five optical cycles and at reachable field strengths (Table I). Beyond possible applications for second harmonic generation, rectification, and driven, on-demand ferroelectricity, the study of interactions of phonons in the newly described state with electrons and collective modes offers a route to novel out-of-equilibrium states.

Acknowledgments—We acknowledge comments by the anonymous referee of one of our previous publications [59], which, eventually, led to the research reported here, as

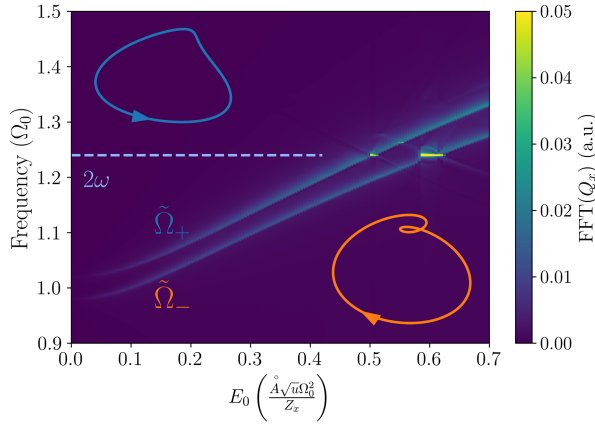


FIG. 4. Symmetry breaking with nondegenerate chiral phonons [see Eq. (20)]. The phonon frequencies are split according to Eq. (20): $\Omega_{\pm}$ corresponds to right- and left-handed motion. The two modes are accessed with light of opposite polarization. To excite the Ω_{+} mode, we use $\mathbf{E} = E_0[-(1 - \delta) \cos \omega t, \sin \omega t, 0]$, and for the Ω_{-} mode, we use $\mathbf{E} = E_0[\cos \omega t, (1 - \delta) \sin \omega t, 0]$, with $\delta = 0.25$ and $\omega = 0.62\Omega$. As for degenerate chiral phonons, a slight detuning from circularity δ is necessary in order to trigger the instability at half the resonance frequency [see Eq. (D5)]. The figure combines the results of two runs, in which the two chiralities were simulated separately. The resonance frequencies exhibit a driving amplitude dependent blueshift such that the instability occurs at different powers for the two chiralities.

well as useful discussions with Mark Rudner. We also thank Jonas F. Karcher for his motivating remarks. This project received funding from the Horizon Europe Marie Skłodowska-Curie Action program under Grant Agreement No. 101155351.

Data availability—The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

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End Matter

Appendix A: Achieving symmetry breaking with pulsed excitations—We perform simulations to show that the symmetry-breaking steady state can be reached with short pulsed signals. We confine the oscillating electric field in Eq. (3) to a window of length $T_p = 5 \cdot 2\pi/\omega$. This mimics a flattop pulse (see Fig. 5) whose amplitude is constant throughout T_p . The constant amplitude is necessary because due to the blueshift of $\tilde{\Omega}_0(E_x)$ both the amplitude and frequency of the drive have to be matched with each other (see Fig. 1). Figure 1 shows the flattop pulse and the response of the phonon mode $Q_x(t)$.

A note on initial conditions and symmetry breaking: We started our simulation runs at $t = 0$ and used the forcing term $E_x(t) = E_{0,x} \cos(\omega t)$; effectively this corresponds to $E_x(t) \sim \Theta(t) \cos(\omega t)$. The response of Eq. (3) to this forcing necessarily consists of particular solution oscillating at ω and persisting for $t \rightarrow \infty$ and a homogeneous solution oscillating at $\tilde{\Omega}_0$ that fixes the boundary conditions at $t = 0$ and decays due to damping. The homogeneous solution has both finite Q_{even} and Q_{odd} components since $\tilde{\Omega}_0$ is not necessarily a multiple of ω . It thus initializes the parametric instability in Eq. (9), which gives $Q_{\text{even}}(t) = 0$ for $Q_{\text{even}}(0) = 0$ and $\dot{Q}_{\text{even}}(0) = 0$, acting as an initial forcing for Q_{even} and determining the sign of the symmetry breaking.

Appendix B: Noise robustness—In our noise simulations, $\eta(t)$ is a normally distributed random function on a time grid with time points t_i separated by intervals $\Delta t = 5 \times 10^{-3}/\Omega_0$. For simplicity, we use cubic splines to interpolate $\eta(t)$ between the t_i and use a standard numerical integrator (SciPy's `odeint`). This does not influence the physical results since $\Delta t \ll 2\pi/\Omega_0$. The

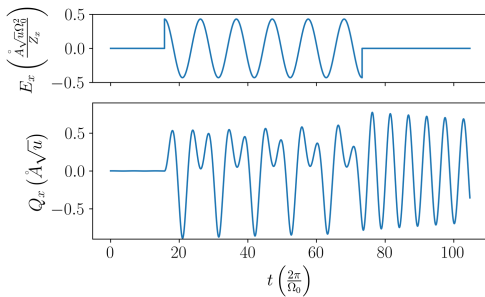


FIG. 5. Reaching the symmetry-breaking state is possible with a short flattop pulse of only five optical cycles. The upper plot shows the driving electric field, and the lower plot shows the response with visible second harmonic and dc offset components. When the pulse ends, the system switches to unforced oscillations at frequency Ω_0 . The spectrum of $Q_x(t)$ is shown in Fig. 1 of the main text.

symmetry-breaking steady state is stable up to a threshold noise amplitude A_* .

To relate the noise amplitude A_* to a temperature, we calculate the internal energy of the oscillator at finite temperatures via $U = -\partial_{\beta} \log Z$:

$$\begin{aligned} U &= -\frac{1}{2\pi\hbar} \frac{\partial}{\partial(1/k_B T)} \log \int dq \int dp e^{-\frac{H(p,q)}{k_B T}} \\ &= \frac{1}{8} \left(\frac{\Omega^4 K_{\frac{5}{4}} \left(\frac{\Omega^4}{8\beta k_B T} \right)}{\beta K_{\frac{3}{4}} \left(\frac{\Omega^4}{8\beta k_B T} \right)} - \frac{\Omega^4}{\beta} + 2k_B T \right) \\ &\approx k_B T - \frac{3\beta(k_B T)^2}{4\Omega^4} - \mathcal{O}(\beta^2). \end{aligned}$$

Assuming a phonon resonance frequency of 3 THz and extracting the internal energy U corresponding to noise of amplitude A_* (in absence of driving), we estimate that A_* corresponds to a temperature of about 700 K.

Appendix C: Pumping via Raman-active modes—In centrosymmetric materials, Raman- and IR-active modes are mutually exclusive. They can be, however, nonlinearly coupled to each other via the coupling Hamiltonian of Eq. (17), where Q_R is the Raman-active mode and Q_x is the IR-active mode. The equations of motion for the coupled system can then be written as

$$\ddot{Q}_x + 2\gamma\dot{Q}_x + \Omega_0^2 Q_x + \beta Q_x^3 = 2A Q_x Q_R + Z_x E_0 \cos(\omega t), \quad (\text{C1})$$

$$\begin{aligned} \ddot{Q}_R + 2\gamma\dot{Q}_R + \Omega_R^2 Q_R + \beta_{R,2} Q_R^2 + \beta_{R,3} Q_R^3 \\ = A Q_x^2 + \frac{1}{2} \chi_R E_0^2 [1 + \cos(2\omega t)]. \end{aligned} \quad (\text{C2})$$

We assume that the drive is in resonance with the Raman-active mode, such that $2\omega \approx \Omega_R$. Furthermore, we assume that Ω_0 is distinct from Ω_R , such that the driving is off resonant with Ω_0 and the direct, off-resonant driving of Q_x via Z_x is negligible. We furthermore are interested in instabilities of Q_x induced by the Raman mode. At the instability onset, which we want to study here, Q_x will be small, and we can neglect higher order terms in this variable. The simplified system of equations reads as

$$\ddot{Q}_x + 2\gamma\dot{Q}_x + \Omega_0^2 Q_x - 2A Q_x Q_R = 0, \quad (\text{C3})$$

$$\begin{aligned} \ddot{Q}_R + 2\gamma\dot{Q}_R + \Omega_R^2 Q_R + \beta_{R,2} Q_R^2 + \beta_{R,3} Q_R^3 \\ = \frac{1}{2} \chi_R E_0^2 [1 + \cos(2\omega t)]. \end{aligned} \quad (\text{C4})$$

Inverting C4, we find that, neglecting higher harmonics,

$$Q_R \approx F_{R,0}(E_0) + F_{R,2}(E_0) \cos(2\omega t), \quad (\text{C5})$$

where $F_{R,0}$ and $F_{R,2}$ are the amplitudes of dc and second harmonic (with respect to the driving frequency) components, respectively. Inserting this into C3 gives

$$\ddot{Q}_x + 2\gamma\dot{Q}_x + \tilde{\Omega}_0(E_0)[1 - h(E_0) \cos(2\omega t)]Q_x = 0. \quad (\text{C6})$$

Here $\tilde{\Omega}_0(E_0) = \Omega_0^2 + F_{R,0}(E_0)$ is the effective, blueshifted resonance frequency of the IR-active mode and $h(E_0) = 2AF_{R,2}(E_0)$. This equation maps onto Eq. (D2) of the main text. This shows that a Raman-active mode can be used to trigger the symmetry-breaking instability of the IR-active mode. The frequency of the resonant Raman-active mode $\Omega_R \approx 2\omega$ has to be reasonably close to Ω_0 . However, some detuning is possible because the effective resonance frequency $\tilde{\Omega}_0(E_0)$ experiences a blueshift. This blueshift can be exploited to adjust the effective resonance frequency by adjusting the driving amplitude E_0 .

Similarly, an auxiliary IR-active mode with a frequency close to half the resonance Ω_0 can be used instead of the Raman-active mode. An advantage of using an auxiliary (Raman- or IR-active mode) is that the mode will store the energy supplied by the drive over many cycles. Thus the threshold amplitude for the instability might be easier to reach.

Appendix D: Collective instability of the x and y modes—The equations of motion (with the addition of a damping term) following from Eq. (18) read as

$$\begin{aligned} \ddot{Q}_x + 2\gamma\dot{Q}_x + \Omega_0^2 Q_x + \beta Q_x(Q_x^2 + Q_y^2) &= Z_x E_x \cos \omega t, \\ \ddot{Q}_y + 2\gamma\dot{Q}_y + \Omega_0^2 Q_y + \beta Q_y(Q_x^2 + Q_y^2) &= Z_y E_y \sin \omega t. \end{aligned} \quad (\text{D1})$$

We first derive the two-component analog of Eq. (9), which is given by

$$\begin{aligned} \ddot{Q}_{x/y,\text{even}} + 2\gamma\dot{Q}_{x/y,\text{even}} \\ + \left[\tilde{\Omega}_{x/y}^2 \pm \frac{\alpha}{2} \left(3F_{x/y,1}^2 - F_{y/x,1}^2 \right) \cos(2\omega t) \right] Q_{x/y,\text{even}} \\ + \alpha F_{x/y,1} F_{y/x,1} \sin(2\omega t) Q_{y/x,\text{even}} = 0, \end{aligned} \quad (\text{D2})$$

with $\tilde{\Omega}_{x/y}^2 = \Omega_0^2 + \alpha(3F_{x/y}^2 + F_{y/x}^2)/2$, where F_y is defined analogously to F_x in Eq. (7). As above, we expect parametric resonances near $2\omega = \tilde{\Omega}_{x/y}$, where $\tilde{\Omega}_x \neq \tilde{\Omega}_y$ for $F_x \neq F_y$. For now, let us choose the case $2\omega = \tilde{\Omega}_x$, such that the instability occurs for the $Q_x(t)$ component.

The oscillating terms in Eq. (D2) couple harmonics with frequencies $2\omega, 4\omega, \dots$ and dc terms. For the stability

analysis, we therefore choose the ansatz

$$\begin{aligned} Q_{i,\text{even}} &= a_{i,1} \sin(2\omega t) + a_{i,2} \sin(4\omega t) + b_{i,0} \\ &+ b_{i,1} \cos(2\omega t) + b_{i,2} \cos(4\omega t). \end{aligned} \quad (\text{D3})$$

Furthermore, we neglect γ for the duration of this analysis. While γ determines the instability threshold amplitudes of the electromagnetic fields [see Eq. (12)], its effects become less important for driving amplitudes above the threshold, i.e., for any driving amplitude, γ can be always chosen small enough that our analysis is accurate. We again search the instability window for the detuning $\Delta = 2\omega - \tilde{\Omega}_x$, such that the mode amplitudes $a_{i,n}$ and $b_{i,n}$ grow exponentially for $\Delta_{\min} < \Delta < \Delta_{\max}$. At $\Delta = \Delta_{\max/\min}$, the amplitudes will be constant. The boundaries of the instability interval $\Delta_{\max/\min}$ are then found by inserting the ansatz (D3) into Eq. (D2) and assuming that $a_{i,n}$ and $b_{i,n}$ are indeed constant [38]. After a lengthy calculation, in which we compare the coefficients of different harmonics after inserting the ansatz (D3) into Eq. (D2), we find that, to fourth order in $F_{x,1}$ and $F_{y,1}$,

$$\begin{aligned} \Delta_{\max} - \Delta_{\min} &= F_{y,1}^4 \left(\frac{\beta^2}{16\Omega_0^3} + \frac{287\beta^4 F_{x,1}^4}{576\Omega_0^7} + \frac{47\beta^3 F_{x,1}^2}{192\Omega_0^5} \right) \\ &+ F_{y,1}^2 \left(\frac{77\beta^3 F_{x,1}^4}{192\Omega_0^5} - \frac{5\beta^2 F_{x,1}^2}{8\Omega_0^3} \right) + \frac{9\beta^2 F_{x,1}^4}{16\Omega_0^3}. \end{aligned} \quad (\text{D4})$$

The full result is too long to be quoted here but is easily found using computer algebra. Equation (D4) is valid for small F_x and F_y , i.e., for small driving amplitudes. It is interesting to study the behavior of Δ close to $F_{y,1} = F_{x,1}$, i.e., for nearly perfect circular polarization. Writing $F_{y,1} = F_{x,1} + F_e$, we find

$$\Delta_{\max} - \Delta_{\min} = -F_e^3 \left(\frac{9\Omega_0}{8F_{x,1}^3} + \frac{\beta^2 F_{x,1}}{\Omega_0^3} + \frac{51\beta}{16F_{x,1}\Omega_0} \right). \quad (\text{D5})$$

Notice that for $F_{y,1} \geq F_{x,1}$ (we choose both amplitudes positive without loss of generality), we have $\Delta_{\max} \leq \Delta_{\min}$, which indicates that the system is stable. For $F_{y,1} > F_{x,1}$, the Q_x and Q_y components switch places, and the instability occurs for $2\omega = \tilde{\Omega}_y$. The analysis for this case is completely analogous with $F_{x,1}$ and $F_{y,1}$, as well as Ω_x and Ω_y interchanged. We therefore conclude that, in general, the instability occurs either for the Q_x or Q_y component, depending on whether $F_{x,1}$ or $F_{y,1}$ is larger. For $E_x = E_y$, resulting in $F_x = F_y$, we find $\Delta_{\min} = \Delta_{\max}$ and the instability cannot be reached [60].