Photonic time-crystalline behaviour mediated by phonon squeezing in $\text{Ta}_2\text{NiSe}_5$

Manipulating materials in time and space through optical pumping paves the way for new phenomena and functionalities. A notable example in this field is photonic time crystals, defined as materials whose effective dielectric properties oscillate periodically in time, in analogy to photonic crystals whose effective dielectric properties oscillate periodically in space (Photonic time crystals should not be confused with discrete time crystals that refer to spontaneous symmetry-breaking in time). Photonic time-crystals have emerged as a promising platform for terahertz (THz) amplification of reflectivity in a narrow frequency band. The theory is supported by experimental results on $\text{Ta}_2\text{NiSe}_5$ where photoexcitation with short pulses leads to enhanced THz reflectivity with the predicted features. We explain the key mechanism leading to THz amplification in terms of a simplified electron-phonon Hamiltonian motivated by ab-initio DFT calculations. Our theory suggests that the pumped $\text{Ta}_2\text{NiSe}_5$ is a gain medium, demonstrating that squeezed phonon noise may be used to create THz amplifiers in THz communication applications.
Previous analysis has demonstrated the existence of four types of drive-induced features in reflectivity, depending on the relative strength of parametric driving and dissipation. Edge-like features occur from interference between different Floquet components of the transmission channels when dissipation is strong compared to the oscillation amplitude. In the opposite regime, strong amplification of reflectivity occurs when the parametric drive is not compensated by dissipation and the material exhibits a lasing instability. In fact, such reflectivity features can serve as reporters of a lasing instability, indicating that the effective Floquet medium can be used as a gain medium in a laser.

In this article, we provide a theory that exemplifies that THz noise of phonons can be used to create a THz photonic time crystal in the correlated material Ta2NiSe5. In particular, we are able to explain pump-induced THz amplification of reflectivity in Ta2NiSe5 that is observed experimentally.

Ta2NiSe5 attracted considerable attention in the literature due to its potential as an excitonic insulator candidate. This material has an insulating gap below the critical temperature which grows to be 160 meV at low temperatures. The pump-probe experiment demonstrates that an optical excitation at frequencies above the electronic gap induces reflectivity amplification which is maximal at 4.7 THz; a frequency that corresponds to an IR-active B3u phonon mode seen previously in infrared measurements. This turns out to be a surprising result given that the high-frequency pump cannot directly excite the collective THz modes. Instead, we show phenomenologically and using ab initio calculations that this dramatic down conversion is the consequence of creating a photonic time crystal through phonon squeezing.

The multi-step process described in this article, converting high-frequency pumping to THz amplification of reflectivity, is schematically illustrated in Fig. 1a and is outlined as follows: (a) The pump excites electrons from the valence bands to the conduction bands through direct dipole transitions. (b) The photoexcited electrons exhibit an unusual quadratic coupling to the 4.7 THz phonons (see Results):

\[ H_{\text{el-ph}} = \sum_k g_k n_{\text{el}} Q_k^2, \]  

where \( Q \) is the IR-active phonon displacement, \( n_{\text{el}} \) is the photoexcited electron occupation, and \( g_k \) is the effective electron-phonon coupling. The pump is non-resonant with the IR-active THz phonons and thus does not directly initiate the enhanced reflectivity dynamics. Instead, IR-active phonon pair generation occurs via a Raman process caused by pump-induced changes in the electronic occupation. The result is that even though the expectation value of the phonon displacement is zero, \( \langle Q \rangle = 0 \), the fluctuations are squeezed and coherently oscillate at twice the phonon frequency, \( \langle Q^2(t) \rangle = \langle Q^2 \rangle_0 + A \cos(2\omega_{\text{ph}} t) \). (c) The squeezed phonon oscillations and phonon nonlinearities create a Floquet material/photonic time crystal oscillating at 2\( \omega_{\text{ph}} = 9.4 \) THz. Parametric resonances due to the oscillating field occur primarily around \( \omega_{\text{ph}} \), parametrically amplifying the reflectivity as depicted in Fig. 1b.

The parametric amplification of reflectivity in the photonic time crystal created by the squeezing phonon oscillations can be understood as follows. The oscillations of phonon fluctuations, parametrically drive the rest of the material through nonlinear interactions, which can be schematically represented as:

\[ H_{\text{amp}} = X(t) a_{\text{ph}}^\dagger a_{\text{ph}} + h.c., \]  

driving from the phonon oscillations can create pairs of photons at the signal and idler frequency once stimulated by the probe pulse. This enhances the reflectivity and also scatters counter-propagating light oscillating at the idler frequency.

In this article, we provide a theory that exemplifies that THz noise of phonons can be used to create a THz photonic time crystal in the correlated material Ta2NiSe5. In particular, we are able to explain pump-induced THz amplification of reflectivity in Ta2NiSe5 that is observed experimentally.
4.7 THz is consistent with the creation of a photonic time crystal whose orthorhombic phase. This indicates that the THz parametric amplification and the order parameter hints at the possibility that the creation of a photonic time crystal can even be used as a novel method to track the magnitude of the electron-phonon coupling and the IR activity of the phonon:

\[
H_{\text{el.-ph}} = Q \sum_k \Delta_k (\hat{c}^\dagger_{1k} \hat{c}_{2k} + \hat{c}^\dagger_{2k} \hat{c}_{1k}).
\]

where \( Q \) is a zero momentum IR phonon coordinate, \( \hat{c}_1 \) and \( \hat{c}_2 \) the annihilation operators of the two electronic bands and \( \Delta_k \) the electron-phonon coupling matrix element as a function of momentum \( k \). Using a Schrieffer-Wolff transformation, we “integrate out” the linear electron-phonon coupling, which is non-resonant due to the different energy scales between the IR-phonon and the electronic transition, to reveal the resonant non-linear coupling between the electron occupation number and the phonon squeezing operator, \( Q^2 \).

In the Methods, we show that this procedure leads to the effective coupling:

\[
H_{\text{el.-ph,eff}} = \sum_k \frac{\Delta_k^2}{\Delta} (n_{1k} - n_{2k}) Q^2.
\]

where \( \Delta_k = E_{1k} - E_{2k} \) is the energy difference between the electronic states and \( n_{ik} = \hat{c}^\dagger_{ik} \hat{c}_{ik} \) the number operator. The above equation applies to phonons that are coupled to independent pairs of electronic bands. If three or more bands are simultaneously coupled to a specific phonon the effective electron-phonon Hamiltonian is more complicated but with similar qualitative features, such as the coupling of electron density to the square of the phonon coordinate.

### Results

**Phonon squeezing initiated by photoexcited electrons**

**Electron-phonon coupling.** In this section, we develop a microscopic theory of the coupling of electronic bands to IR active phonons. From symmetry arguments, the IR phonon coordinate is odd under inversion, and cannot couple linearly to the electron density which is even under inversion. However, it can couple linearly to electronic dipole transitions which are odd under inversion. For two electronic bands with an allowed dipole transition, this coupling takes the form:

\[
H_{\text{el.-ph}} = Q \sum_k \Delta_k (\hat{c}^\dagger_{1k} \hat{c}_{2k} + \hat{c}^\dagger_{2k} \hat{c}_{1k}).
\]

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**Phonon squeezing.** Once electrons have been photoexcited, the finite number of optically excited electrons quenches the frequency of the phonon:

\[
H_{\text{el.-ph,eff}} = \frac{Mf(t)Q^2}{2},
\]

where \( M \) is the mass of the phonon. In equation (5), the parametric driving \( f(t) \) comes from the photoexcited distribution of electrons that is strongly coupled to the phonon:

\[
f(t) = \sum_k \frac{2\Delta_k^2}{M^2 \omega_{ph}^2} (n_{1k}(t) - \langle n_{2k}(t) \rangle).
\]

The photoexcited electron dynamics are fast and can be approximated as a delta function in time. More generally we can approximate the photoexcited distribution as having a characteristic lifetime, \( f(t) = f(t)^{\text{eff}} \exp(-t/\tau_{\text{eff}}) \), but this does not change our conclusions.

The above electron-induced drive describes a Raman process that does not excite the phonon directly (i.e. \( Q = 0 \)). However, the squeezing operator starts oscillating at a frequency equal to twice the phonon frequency, as shown in the Methods:

\[
\left( \omega_{ph}^2 + 2Q^2 \right) Q^2 = -2f(t)Q^2 \theta',
\]

where \( \langle Q^2 \rangle_0 \) is the equilibrium fluctuations and \( \omega_{ph} \) is phonon frequency at zero momentum which is renormalized by the coulomb force, \( \omega_{ph}^2 = \omega_{ph,0}^2 + \frac{\lambda^2}{C_0^2} \).
To show that this phenomenon is related to squeezing, we expand $Q^2$ in terms of creation and annihilation operators, using
\[
Q^2(t) = \frac{1}{2M_{\text{ph}}} \left( a'(t)a'(t) + a(t)a(t) + a'(t)a(t) + a(t)a'(t) \right).
\]
Expectation values of $a(t)a(t)$ do not oscillate rapidly while the anomalous pairs $a'(t)a'(t)$ and $a(t)a(t)$ oscillate at twice the phonon frequency. As a result, a state with phonon fluctuations, $Q^2$, that oscillates at twice the phonon frequency implies the existence of a condensate of phonon pairs $a'(t)a'(t) + a(t)a(t) = 0$.

**Parametric amplification of reflectivity in pumped Ta$_2$NiSe$_5$**

Equilibrium reflectivity. The reflectivity of a material is captured by the frequency-dependent refractive index appearing in the Maxwell equations:
\[
\left( \frac{n(\omega)^2 \omega^2}{c^2} - k^2 \right) E = 0
\]
where $E$ is the electric field and all information about phonons and other IR active modes is encoded in $n(\omega)$. We assume that the probe corresponds to an electromagnetic wave reflected from the sample at normal incidence. The propagation direction is along the $b$-axis of the crystal, which we refer to as the $y$-direction, whereas the electric field points in the $a$-direction which we refer to as the $x$-axis. The refractive index can be directly extracted experimentally from the complex reflection coefficient at normal incidence in an equilibrium system:
\[
n(\omega) = 1 - \frac{r(\omega)}{1 + r(\omega)}.
\]

**Eigenstates in the driven state.** Once we have obtained the refractive index from the equilibrium reflectivity, we model the Floquet material as experiencing a parametric drive oscillating at frequency, $\omega_d$, which mixes signal and idler frequencies. Solutions of coupled light-matter equations are given by Floquet-type eigenmodes:
\[
E(t) = e^{i\omega_d t} \left( E_s e^{-i\omega_s t} + E_{id} e^{i\omega_{id} t} \right),
\]
where $\omega_s$ is the frequency of the incoming probe and $\omega_{id} = \omega_d - \omega_s$. Such mixing corresponds to degenerate perturbation theory in Floquet systems and the idler component is the nearest Floquet band contribution to $\omega_s$ which is responsible for parametric instabilities. The oscillating mode is included phenomenologically through a time-periodic contribution to the electric permittivity:
\[
\delta e(t) = 2A_{\text{drive}} \cos(\omega_d t).
\]
Using the ansatz in equation (11), the equations of motion in the Floquet state for the different oscillating components of the electric field become:
\[
\frac{n(\omega)^2 \omega^2}{c^2} E_s + A_{id} E_{id} = 0
\]
\[
\frac{n(\omega_{id})^2 \omega_{id}^2}{c^2} E_{id} + A_s E_s = 0.
\]
To compute the reflectivity at normal incidence, we first find the allowed $k$ values for a given $\omega_d$. Due to the coupling of signal and idler components, two such $k$ values exist, associated with two transmission channels, both of which oscillate at signal and idler frequencies. The transmission channels correspond to eigenvectors of the Floquet equations of motion inside the material,
\[
E_i = t_i E_0 e^{i\kappa y} (e^{-i\omega_s t} + a_i e^{i\omega_{id} t}),
\]
where $a_i$ is the relative amplitude of the signal and idler component in the eigenvector of wave-vector $k_i$. $t_i$ is the transmission coefficient of the $i$th channel and $E_0$ is the amplitude of the incoming field.

**Floquet-Fresnel equations.** In a reflection problem, the eigenvalue equation (13) enables the computation of the transmitted wavevectors as a function of a fixed frequency set by the incoming light. However, the answer is given in terms of $k_i^2$ rather than $k_i$, and the correct root is chosen such that the field vanishes at infinity, $\text{Im}(k_i) > 0$. The reflectivity is computed by solving the Floquet-Fresnel equations, matching electric and magnetic fields parallel to the surface both at frequency $\omega_s$ and at frequency $\omega_{id}$. Inside the material, we have the electric field:
\[
E_{\text{mat}} = -E_0 \sum_i t_i e^{i\kappa y} (e^{-i\omega_s t} + a_i e^{i\omega_{id} t}),
\]
and for vacuum, we have:
\[
E_{\text{vac}} = E_0 \left( e^{i\omega_{id}/\omega_s \kappa y} t_i e^{-i\omega_{id}/\omega_s \kappa y} + r_i e^{-i\omega_{id}/\omega_s \kappa y} \right) + E_0 e^{i\omega_{id} t} e^{i\omega_{id} \kappa y}.
\]
Using the homogeneous Maxwell equations, $\nabla \times E = -\partial B/\partial t$, to compute the magnetic field and matching boundary conditions at $y = 0$, we obtain the Fresnel equations for the driven system:
\[
1 + r_s = \frac{k_s}{\omega_s} t_1 + t_2,
\]
\[
1 - r_s = \frac{k_s}{\omega_s} t_1 + t_2
\]
\[
r_{id} = t_1 a_1 + t_2 a_2,
\]
\[
r_{id} = \frac{k_s}{\omega_{id}} t_1 a_1 + \frac{k_s}{\omega_{id}} t_2 a_2.
\]
To fit the data, we choose a drive at 9.4 THz motivated by the DFT calculations outlined in the ab initio calculations section. We then use equations (13) and (17) and fit the parameters $A_{\text{drive}}$ and $n(\omega)$ to the experimental data allowing for small changes in the static properties of the system such as a photoinduced conductivity stemming from photexicted charge carriers. The fitted parameters are given in the Methods section.

**Photonic time crystal from squeezing dynamics of phonons.** Material properties such as the electric permittivity, become time-periodic in the presence of oscillating fields through interactions. Here, we demonstrate how lattice potential anharmonicities lead to a time-periodic index of refraction:

We consider a phonon system with a $Q^4$ anharmonicity for the IR-phonon with a Hamiltonian:
\[
H_{\text{ph}} = \sum_k \left( Z E_k Q_k + M \left( \omega_{ph,0}^2 + f(t) \right) Q_k Q_k + \frac{\Pi_s \Pi_{id}}{M} + \omega_{id}^2 Q_{id} Q_{id} \right),
\]
where we explore how the squeezing oscillations of the phonon at zero momentum, $Q_{id}=0$, affects the propagation of light at finite $\{E\text{ph}, Q_k\}$. The equations of motion for the phonon given by the Hamiltonian in

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*Note: The text continues with further details and equations that are not fully transcribed here.*
To describe the dynamics of phonons, we include phenomenologically the damping term $\gamma$, which captures the decay of the phonon to other degrees of freedom. Using a Gaussian ansatz for the phonons, we can linearize the above equation as:

$$\left( \partial_t^2 + y \partial_t + \omega_{ph,0}^2 + \frac{1}{2} \left( Q_{g=0}^2 \right) \right) Q_k = \frac{Z}{M} E_k.$$  (19)

where the fluctuations $\langle Q^2 \rangle = \langle Q_{g=0}^2 \rangle + A \cos(2Q_{g=0} t)$. The contribution from $f(t)$ corresponds to the fast dynamics of the photoelectrons and therefore has a broad spectrum. As such, it does not lead to resonant dynamics and will at most contribute to an incoherent background. In contrast, squeezing oscillations occurring at a specific frequency can lead to parametric amplification that is narrowband. Hence, we drop the contribution of $f(t)$ and focus on the consequences of the squeezing mode oscillations. The phonon mode appears in the Maxwell equations as:

$$\left( \frac{1}{c^2} \partial_t^2 - k^2 \right) E_k = -Z \partial_t^2 Q_k.$$  (20)

To find the effective signal idler mixing presented in equation (13), we expand the equations of motion in signal and idler contributions:

$$Q_k = Q_s e^{-i\omega_l t} + Q_id e^{i\omega_l t}.$$  (22)

Equation (20) becomes:

$$\begin{bmatrix} Q_s \\ Q_id \end{bmatrix} = \begin{bmatrix} \frac{Z}{M} \omega_l^2 + i \gamma \omega_l - \omega_{ph,0}^2 \\ 0 \end{bmatrix} \begin{bmatrix} E_s \\ E_id \end{bmatrix} + \begin{bmatrix} 2 \omega_{ph,0}^2 \\ 0 \end{bmatrix} \left( \begin{bmatrix} \omega_l^2 + i \gamma \omega_l - \omega_{ph,0}^2 \\ \omega_{ph,0}^2 + i \gamma \omega_{ph,0} - \omega_{ph,0}^2 \end{bmatrix} \begin{bmatrix} E_s \\ E_id \end{bmatrix} \right).$$  (23)

Substituting equation (23) in Maxwells equation we find the equations of motion for the signal and idler component of the electric field to be:

$$\left( \frac{n_{eq}^2 (\omega_s)}{c^2} - k^2 \right) E_s + A_{drive,s} (\omega_s, \omega_id) E_id = 0,$$  (24a)

$$\left( \frac{n_{eq}^2 (\omega_id)}{c^2} - k^2 \right) E_id + A_{drive,id} (\omega_s, \omega_id) E_id = 0$$  (24b)

where the signal and idler driving amplitudes, $A_{drive,s}$ and $A_{drive,id}$ are given by:

$$A_{drive,s} = -\frac{Z^2 / M u A \omega_s^2}{2 \left( \omega_s^2 + i \gamma \omega_s - \omega_{ph,0}^2 \right) \left( \omega_id^2 + i \gamma \omega_id - \omega_{ph,0}^2 \right)},$$  (25a)

$$A_{drive,id} = -\frac{Z^2 / M u A \omega_id}{2 \left( \omega_s^2 + i \gamma \omega_s - \omega_{ph,0}^2 \right) \left( \omega_id^2 + i \gamma \omega_id - \omega_{ph,0}^2 \right)}.$$  (25b)

The above analysis presents an illustration of how non-linearities and squeezing dynamics lead to a Photonic time crystal. In this specific model, we find that the signal idler coupling is proportional to the IR activity of the mode, $Z$, the phonon anharmonicity, $\Delta_k$, and the amplitude of the oscillations. $A_{drive} = Z u A$. The precise strength of the coupling between signal and idler modes will depend on the choice of non-linearity. As a result, in this article, we take an agnostic view and instead fit the experimental data to the signal and idler coupling in equation (13).

**Ab initio calculations: the 4.7 THz IR-Phonon**

The above discussion is generic and in principle applies to every IR phonon inside a given material. In this section, we use DFT calculations to determine which phonons make the dominant contribution to the parametric amplification of reflectivity in Ta$_2$NiSe$_5$. We start our discussion by identifying which valence and conduction bands are involved in photo-absorption by evaluating the optical dipole transition matrix elements. In Fig. 2(a), we plot the momentum-resolved optical contribution, for field polarization along the $a$-axis, of each band to dipole-allowed transitions within the experimentally relevant energy window between 0.33 eV and 0.9 eV set by the pump parameters. For a given valence(conduction) band and k-point, the optical contribution is defined by summing the square of the dipole transition matrix elements associated with the transition. We find that it is the first three conduction bands that are predominantly excited by the pump.

Turning our attention to phonons, we use ab initio calculations to identify all IR active phonons. In particular, we find a number of phonons in the low-temperature monoclinic phase between 4 and 5 THz presented in Supplementary Table 1 in Supplementary Note 1. In Fig. 2(b) we plot the IR activity of phonons as a function of frequency and identify the 4.7 THz phonon which, as shown below, turns out to be the most strongly electron-coupled phonon.

We compute the electron-phonon interaction between IR active phonons and electrons. This is to identify the phonons that dominate the interaction with the photo-excited electronic bands. To accomplish this we use the method of frozen phonons. In this approach, the electronic bands are recalculated with the lattice shifted along a phonon eigendisplacement. To quantify the coupling strength of a specific phonon to a particular electronic band we integrate the energy changes of the band in the presence of the phonon over the Brillouin zone as outlined in Supplementary Note 1. We find that in the vicinity of 4.5 THz, relevant to the experimental observables, the 4.7 THz mode has roughly an order of magnitude larger electron-phonon coupling strength compared to the nearby phonons. In particular, as shown in Fig. 2c, the 2nd and 3rd conduction bands (in this paper we number the conduction bands from lowest to highest in energy) are significantly renormalised in the presence of the 4.7 THz phonon. Since the phonon is mostly coupled to two electronic bands, the frozen phonon calculations are consistent with equation (4) leading to the two bands shifting in energy by an equal and opposite amount given by:

$$E_{k,2}^{\Delta \omega,0} = E_{k,1}^{\Delta \omega,0} = -\left( E_{k,2}^{\Delta \omega,0} - E_{k,1}^{\Delta \omega,0} \right),$$  (26)

where $E_{k,2}$ and $E_{k,3}$ is energy of conduction bands two and three at momentum $k$, and $\Delta_k$ is the energy difference of the two bands in equilibrium. Therefore, in this case, calculating the energy shifts as a function of momentum in the frozen phonon approximation allows for direct computation of the electron-phonon interaction.

To summarize, we use ab initio calculations to identify the electron bands excited by pumping and to establish the IR-active phonons in Ta$_2$NiSe$_5$. Subsequently, frozen phonon calculations allowed for the identification of the phonon with the dominant electron-phonon coupling to the excited electronic bands. This leads to the identification of the 4.7 THz mode as responsible for the parametric
amplification observed in experiments through the mechanism outlined in previous subsections. We note that we do not exclude the possibility of subdominant amplification in reflectivity spectra arising from other phonons at different frequencies. In Supplementary Note 1, we provide more details of our DFT analysis of IR-phonons in Ta$_2$NiSe$_5$, the frozen phonon calculations, and the calculation of optical matrix elements.

Therefore, the DFT calculation provides strong support for the minimal electron-phonon model and uncovers the dominant electronic bands and IR phonons involved in the nonlinear process. The other ingredients needed to extract the amplification amplitude, i.e., phonon nonlinearities, phonon dissipation, and the dynamics of the photoexcited electrons responsible for phonon squeezing, were included phenomenologically as described in the photoinduced reflectivity fitting section.

**Connection to the order parameter.** Finally, we discuss the effects of the phase transition on the phonon-squeezing process. In Fig. 2, we compute the electron-phonon interaction of the 4.7 THz phonon in the low temperature monoclinic phase while for the high temperature orthorhombic phase we compute the electron-phonon interaction of the phonon adiabatically connected to the 4.7 THz phonon eigenstate (for details on the identification of the adiabatically connected phonon see Supplementary Note 1 and Supplementary Fig. 1). The electron-phonon interaction effectively disappears in the high-temperature orthorhombic phase while for the high temperature monoclinic phase is strongly coupled to the electronic bands 2 and 3 which are highlighted in the figure. In the high-temperature orthorhombic phase, the effective electron-phonon coupling is reduced by an order of magnitude. This indicates that the electron-phonon interaction is very sensitive to the order parameter.

**Discussion**

We have investigated the microscopic mechanism of amplification of THz optical reflectivity in Ta$_2$NiSe$_5$ arising from high-frequency optical pumping. We showed that strong electron-phonon coupling opens new pathways toward realizing THz parametric amplification through high-frequency pumping. Ab initio calculations highlight the importance of the 4.7 THz IR-active phonon which is strongly coupled to electrons allowing for the amplification to manifest in the THz reflect spectrum. This parametric process is attributed to the phonon squeezing phenomenon which transiently drives the material into a photonic time crystal.

Our theory indicates that choosing which electronic band to photoexcite selects the IR phonon that is most strongly coupled to that electronic band. As a result, we can use different pumping frequencies in the same material to tune the frequency of the THz parametric amplification through mode-selective phonon squeezing.

Finally, we showed that the electron-phonon coupling is strongly dependent on the order parameter and becomes suppressed in the high-temperature orthorhombic phase. This suggests that time crystalline behaviours in Ta$_2$NiSe$_5$ may be used as a new probe to order parameter dynamics.

**Methods**

The theoretical fit of the parametric reflectivity amplification in pumped Ta$_2$NiSe$_5$

As mentioned in the main text in equation (10), in principle the complex refractive index can be directly computed by the complex reflectivity amplitude, $r(\omega)$. However, small phase errors upon experimental
extraction of \( r(\omega) \) could lead to the unphysical behaviour of the reflectivity. To overcome this complication, we fit the data by assuming that for an insulator like \( \text{Ta}_2\text{NiSe}_5 \) the refractive index is real. As a result, we can instead use the absolute value of \( |r| = \sqrt{R} \) and express the refractive index as:

\[
n(\omega) = \frac{1 + \sqrt{R}}{1 - \sqrt{R}}.
\]  

(27)

Upon parametric resonance and solving equation (13) together with the Floquet-Fresnel boundary conditions in equation (17), assuming a purely real spectrum with no dissipation leads to highly divergent behavior on parametric resonance. As a result, we put back transient dissipation by including an imaginary component to the refractive index. In the driven case we argue that this is physical since \( n_{\text{driven}}(\omega)^2 = n^2(\omega) + i \delta n_{\text{driven}}/\omega \) and dissipation can arise through a transient contribution to the conductivity by electrons excited across the gap of the insulator. To fit the data we choose a parametric drive at \( \omega_d = 9.4 \text{THz} \), drive amplitude \( A_{\text{drive}} = 7.5 \text{THz} \), overall constant renormalization of the refractive, \( n^2_{\text{drive}} = n^2(\omega) + \delta n^2 \), with \( \delta n^2 = 0.1 \text{THz} \) and an overall Gaussian broadening function with a standard deviation of 0.1 THz. In particular, \( \omega_d = 9.4 \text{THz} \) was set to match the squeezing fluctuations of the 4.7 THz mode, which we derived from DFT as being the most strongly coupled mode to the electrons in that frequency region. The fitted \( A_{\text{drive}} \) corresponds to a small time-dependent change in permittivity compared to the equilibrium permittivity \( \varepsilon = \sqrt{\varepsilon_0} - 0.01 - 0.1 \).

We note in the present work as mentioned earlier in this section, it is not possible to experimentally estimate the imaginary part of the refractive index, accounting for dissipation. This leads to an ambiguity in \( A_{\text{drive}} \) since the presence of more dissipation would need a larger drive amplitude to achieve the same amplification. We found the best fit by fitting both \( A_{\text{drive}} \) and dissipation together. Careful determination of the equilibrium complex reflectivity would allow us to better constrain this parameter from the photoinduced reflectivity. 

**Experimental details**

For the experimental data, a state-of-the-art reflection-based optical pump - THz probe spectroscopy system was used. In the pump-probe setup, the sample was excited with 45-fs 0.5 eV (2.4 µm) pulses generated from a Ti: sapphire laser system having 1 kHz repetition rate, 3 mJ pulse energy, and 800 nm central frequency, via a Light Conversion TOPAS-C optical parametric amplifier. Broadband THz probe beams generated by a 2-color air-based laser plasma scheme were employed.

For the detection, the electro-optic sampling method was employed using a GaP sampling crystal which enabled spectral access to a broad energy window of 0.4 to 7.5 THz. A 1 MHz bandwidth Newport 2307 balanced photodetector was utilized to detect the signal. For the acquisition of the data, a NI PXIe-5122 data acquisition system (DAQ) with a 100 MHz maximum sampling rate and 100 MHz bandwidth was used.

Subsequently, the THz reflectivity of the \( \text{Ta}_2\text{NiSe}_5 \) was obtained using a gold mirror as the reference. We measured \( E_{\text{gold}}(t) \) and \( E_{\text{sample}}(t) \), which are the reflections of the incident THz probe field from the reference and sample, respectively. Here, \( t \) is the electro-optic gate time. Fast-Fourier transforming the time-domain signals yield frequency domain responses \( E_{\text{gold}}(\omega) \) and \( E_{\text{sample}}(\omega) \). The equilibrium reflectivity was computed as \( R = |E_{\text{sample}}(\omega)/E_{\text{gold}}(\omega)|^2 \). The pump-induced transient change in the reflected probe electric field was measured as \( \Delta R(t) \) which can be written as \( \Delta R(t) = E_{\text{sample, pumpe}}(t)/E_{\text{sample, unpumpe}}(t) \). Using the pump-probe signal \( \Delta E(t) \), the photoinduced reflectivity \( R + \Delta R \) can be calculated as \( R + \Delta R = |E_{\text{sample}}(\omega) + \Delta E(\omega)/E_{\text{gold}}(\omega)|^2 \).

Further, pump-induced change in reflectivity \( \Delta R \) was computed by subtracting the equilibrium reflectivity spectrum \( R \) (pump off) from photoinduced reflectivity spectrum \( R + \Delta R \) (pump on). As such, the \( \Delta R/\Delta \) spectrum as shown in Fig. 1(c) essentially denotes the enhancement in THz reflectivity upon photoexcitation.

**Electron-phonon interaction**

The Hamiltonian of two electronic bands coupled by an allowed direct dipole transition that is, in turn, coupled to a phonon is given by:

\[
H_{\text{fi}} = \frac{\Delta}{2} \left( c_1^\dagger c_2 + c_2^\dagger c_1 \right) + \lambda \left( c_1^\dagger c_2 + c_2^\dagger c_1 \right) Q.
\]  

(28)

where \( \Delta = E_1 - E_2 \) is the difference in energy between the two bands, \( \lambda \) is the coupling constant coming from the dipole-dipole interaction in the dipole gauge, \( Q \) is the phonon coordinate and \( |c_1, c_2\rangle \) are the annihilation operators of the two-electron bands. The IR-phonon quadratic Hamiltonian is given by:

\[
H_{\text{ph}} = Z E_Q + M a_\text{ph,0}^2 \frac{Q^2}{2} + \frac{\Pi^2}{2M}
\]  

(29)

where \( \Pi \) is the conjugate momentum of the phonon coordinate \( Q \), \( Z \) is the effective coupling to the electromagnetic field, \( E, \) and \( \omega_{\text{ph,0}} \) is the phonon frequency. Since the transition itself is not resonant with the phonon mode, we decouple the linear electron-phonon interaction perturbatively using a Schrieffer-Wolff transformation. This generates an interaction between the electron-hole pair density and the fluctuations of the phonon field which can be a resonant process. To perform this transformation, it is convenient to note that bilinear combinations of \( |c_1^\dagger c_2, c_2^\dagger c_1\rangle \) appearing in the Hamiltonian, obey SU(2) commutation relations by making the following identification:

\[
S^x = \frac{c_1^\dagger c_2 + c_2^\dagger c_1}{2},
\]  

(30a)

\[
S^y = -i\frac{c_1^\dagger c_2 - c_2^\dagger c_1}{2},
\]  

(30b)

\[
S^z = \frac{c_1^\dagger c_1 - c_2^\dagger c_2}{2},
\]  

(30c)

where their commutators are \( [S^x, S^y] = iS^z \) and their cyclic permutations. In terms of the spin operators, we separate the Hamiltonian into a non-interacting and an interacting part:

\[
H_0 = \Delta S^x + M a_\text{ph,0}^2 \frac{Q^2}{2} + \frac{\Pi^2}{2M}
\]  

(31)

\[
V = 2AS^x Q
\]  

(32)

To remove the interacting part \( V \) to linear order in \( \lambda \), we consider a unitary transformation of the type:

\[
U = \exp(iA),
\]  

(33)

\[
A = aQS^x + b\Pi^x
\]  

(34)

The Schrieffer-Wolff expansion is given by:

\[
U H_{\text{total}} U^\dagger = H_0 + V - i[H_0, A] - i[V, A] - \frac{1}{2} \left[ [H_0, A], A \right].
\]  

(35)
The parameters $\alpha$ and $\beta$ are found such that:
\begin{equation}
V = i[H_0, A]
\end{equation}

Matching linear terms in $\Pi$ and $Q$, leads to the parameters:
\begin{equation}
\beta = \frac{\alpha}{M\Delta},
\end{equation}
\begin{equation}
-\Delta \alpha + 2l + M\omega_\text{ph,0}\beta = 0,
\end{equation}
\begin{equation}
\Rightarrow \alpha = -\frac{2l}{\Delta (1 - \frac{\omega_\text{ph,0}}{\Delta})},
\end{equation}

which confirms that this perturbation theory can be carried out as long as $\omega_\text{ph,0}$ is off-resonant with the transition energy $\Delta$. The effective electron-phonon interaction after the Schrieffer-Wolff transformation is given by:
\begin{equation}
H_{\text{eff}} = -i[V, A] - \frac{1}{2} [H_0, A] = -\frac{i}{2} [V, A],
\end{equation}
where the second term does not depend on the phonons. The residual electron-phonon interaction is thus given by:
\begin{equation}
H_{\text{el-phon}} = \frac{\lambda^2}{\Delta (1 - \frac{\omega_\text{ph,0}}{\Delta})} Q^2 (n_1 - n_2),
\end{equation}

where $n_1 = c_1^\dagger c_1$ is the occupation number of mode 1. This implies that if either mode 1 or mode 2 are photoexcited the phonon can be squeezed.

The linear term in the electric field, $\text{ZEQ}$, appearing in equation (29), is also transformed by the Schrieffer-Wolff transformation. It gives rise to a term $\beta\text{ZEQ}$, which provides a small renormalization of the dipole transition amplitude between the bands and does not affect our discussion.

The above result can be generalized to an arbitrary number of independent pairs of electronic states. For example, for two electronic states, the above result in the limit is generalized to:
\begin{equation}
H_{\text{el-phon}} = \sum_k \frac{\lambda_k^2}{\Delta_k} Q^2 (n_{1,k} - n_{2,k}),
\end{equation}
This expression is the one used in the main text.

Phonon squeezing
Using a Hartree-Fock type approximation on the effective electron-phonon Hamiltonian in equation (40), we derive an effective Hamiltonian for the phonon system only:
\begin{equation}
H_{\text{ph}} = \text{ZEQ} + M \left( \omega_\text{ph,0}^2 + f(t) \right) \frac{Q^2}{2} + \frac{\Pi^2}{2M}
\end{equation}

where $Z$ is the IR activity of the phonon mode, $\omega_\text{ph,0}$ the bare phonon frequency and the effective parametric drive, $f(t)$, is given by photoexcited electron density coupled to the phonon mode:
\begin{equation}
f(t) = \sum_k \frac{2\lambda_k^2}{M\Delta_k} (\langle n_{1,k} \rangle - \langle n_{2,k} \rangle),
\end{equation}

Due to the fast dynamics of electrons, $f(t)$ acts as an impulsive delta function like a parametric drive. Such a drive is not periodic but it can linearly excite phonon fluctuations $\langle Q^2 \rangle$ which will oscillate in time. To show this, we compute the equations of motion for fluctuations of the phonon field:
\begin{equation}
\partial_t \langle Q^2 \rangle = \frac{\langle (Q^2) \rangle}{M},
\end{equation}
\begin{equation}
\partial_t \langle Q^\dagger Q \rangle = -2M \left( \omega_\text{ph,0}^2 + f(t) \right) \langle Q^2 \rangle + 2 \langle \Pi^2 \rangle - 2Z\langle \text{ZEQ} \rangle,
\end{equation}
\begin{equation}
\partial_t \langle \Pi^2 \rangle = -M \omega_\text{ph,0}^2 \langle f(t) \rangle \langle Q^2 \rangle + Z\langle \text{ZEQ} \rangle + Z\langle \text{ZEQ} \rangle.
\end{equation}

At $k=0$, we can use Maxwell’s equations to remove the electric field dependence, $E = \frac{Z}{\varepsilon_0} Q$. Performing this substitution simplifies the equations of motion,
\begin{equation}
\partial_t \langle Q^2 \rangle = \frac{\langle (Q^2) \rangle}{M},
\end{equation}
\begin{equation}
\partial_t \langle Q^\dagger Q \rangle = -2M \omega_\text{ph,0}^2 \langle f(t) \rangle \langle Q^2 \rangle + 2 \langle \Pi^2 \rangle - 2Z\langle \text{ZEQ} \rangle,
\end{equation}
\begin{equation}
\partial_t \langle \Pi^2 \rangle = -M \omega_\text{ph,0}^2 \langle f(t) \rangle \langle Q^2 \rangle + Z\langle \text{ZEQ} \rangle + Z\langle \text{ZEQ} \rangle.
\end{equation}

where $\omega_\text{ph,0}^2 = \omega_\text{ph,0}^2 + \omega_\text{pl,phonon}^2$ is the frequency of the phonon at $k=0$ which differs from the bare frequency by the phonon plasma frequency given by $\omega_\text{pl,phonon}^2 = \frac{Z}{\varepsilon_0}$. Being perturbative in the drive $f(t)$, we expand the phonon fluctuations,
\begin{equation}
\langle Q^2 \rangle = \langle Q^2 \rangle_0 + \langle Q^2 \rangle_1,
\end{equation}
where $\langle Q^2 \rangle_0$ is the thermal expectation value and $\langle Q^2 \rangle_1 \propto f(t)$. To linear order in $f(t)$, the equations of motion imply, $\langle \Pi^2 \rangle_1/M = -M\omega_\text{ph,0}^2 \langle Q^2 \rangle_1 + C(f^2)$. Finally, combining equations in (44) we find that:
\begin{equation}
\partial_t^2 \langle Q^2 \rangle_1 = -2f(t) \langle Q^2 \rangle_0.
\end{equation}

This result shows that phonon fluctuations are linearly driven by photoexcitation and oscillate at twice the phonon frequency, $2\omega_\text{ph}$. These coherent oscillations of phonon fluctuations behave as a Raman mode.

In our treatment, we implicitly assume that only $k=0$ phonons are excited by photoelectrons. The contribution of finite momentum squeezing modes is beyond the scope of our work.

Data availability
All the data and supporting materials can be made available from the corresponding authors upon request. The source data for Fig. 1c, which represents the main result of the current paper, are provided in this paper. Correspondence should be addressed to Marios H. Michael or Sheikh Rubaiat Ul Haque. Source data are provided in this paper.

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**Author contributions**

E.D. and R.D.A. conceived the project together with M.H.M. and S.R.U.H. M.H.M. and E.D. developed the theory framework, analyzed the data, and performed numerical simulations. The optical pump - THz probe data was provided by S.R.U.H. and Y.Z. L.W., S.L., and A.R. performed the DFT analysis. All authors participated in the discussion and interpretations of the results. E.D. and R.D.A. supervised the project. M.H.M. and S.R.U. H. wrote the manuscript with input from all authors.

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