

Terahertz-Driven Parametric Excitation of Raman-Active Phonons in LaAlO_3

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Achieving parametric excitation in an oscillating physical system involves periodically adjusting one of its parameters to modulate the oscillator's natural frequency. This phenomenon has been observed in numerous systems within physics and engineering, profoundly transforming modern science and technology. Despite rapid progress, the parametric control of collective excitations, such as phonons, remains a challenge while promising to generate novel and intriguing effects in a largely unexplored field. Here, we investigate the terahertz (THz) field-induced dynamics of Raman-active phonons in the perovskite structure of LaAlO_3 . Utilizing intense THz pulses, we demonstrate a novel mechanism of parametric phonon excitation marked by substantial subharmonic components. Theoretical analysis can successfully capture the hallmarks of the observed phenomena in a physical scenario with the THz field inducing a parametric coupling between the Raman mode and pairs of acoustic phonon excitations.

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The perovskite family of oxides (ABO_3) shows a wide range of functionalities due to the strong interplay between spin, orbital, and lattice interactions. This is, in particular, reflected in the different structural instabilities found in these systems. An example is the oxygen octahedra tilting and distortions, which are associated with structural phase transition, in particular, with the cooling from the undistorted, high-temperature cubic phase. The idea to control structural instabilities by a coherent drive of phonons in the material was exploited in the last decades using ultrashort laser pulses, and the emergence of new phases [1–7] or the control of existing orders [8–15] was observed. In most of these studies, the change in the material properties was primarily linked to the externally driven displacement of Raman modes, and in particular, the ones responsible for a distortion of the equilibrium crystal structure. It is therefore natural to expect that when a Raman phonon is driven by an external excitation, a larger displacement would lead to a

more significant change in the material properties. So far, the direct excitation of Raman-active modes in solids has been achieved via low-efficient, second-order processes that rely on the scattering of light, such as impulsive stimulated Raman scattering (ISRS) [16] and sum frequency generation (SFG) [17]. In principle, in noncentrosymmetric materials, a more efficient, first-order process that couples an external electric field to the electric dipole of the phonon is also allowed. However, in centrosymmetric materials, Raman phonons are not dipole active, so that, even if they play a crucial role in determining the material properties, their excitation is limited by the low efficiency of the SFG and ISRS. A possibility to indirectly access a Raman mode in centrosymmetric materials is offered by infrared resonant Raman scattering (IRRS) [18] or by ionic Raman scattering (IRS) [19–22]. In both cases, the resonant excitation of IR-active phonons mediates the Raman scattering process through the nonlinear lattice polarizability or through the coupling of phonons by the anharmonic lattice potential (IRRS and IRS, respectively). In their study, Neugebauer *et al.* [23] suggested that IRS can strongly dominate ISRS in exciting the lower energy Raman phonon (antiferrodistortive E_g) in the insulating perovskite (LaAlO_3). It should be pointed out that the efficiency of the IRS is strictly related to the magnitude of anharmonicity of the lattice potential and to the presence of

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an IR-active phonon strongly coupled to the target Raman-active one. Such a coupling relies on selection rules that are hardly achievable in a general compound, so that, even if IRS is an efficient complementary excitation path to the SFG and ISRS, it suffers from a lack of generality. In this context, exploring new directions for the excitation of Raman-active phonons in centrosymmetric materials would open new directions for the control of the structural phases and related macroscopic properties.

In this Letter, we present evidence of a yet unexplored mechanism for coherent parametric excitation of a low-energy Raman-active phonon in centrosymmetric lanthanum aluminate, LaAlO_3 , which makes use of an intense THz electric field. We demonstrate that an intense terahertz pulse with a central frequency between 1 and 2 THz not only coherently excites the Raman-active E_g phonon at 1.08 THz via ISRS and SFG, but also generates significant subharmonic spectral components, which are distinct signatures of an underlying parametric-driving mechanism. This mechanism is based on the absorption of photons through an optical transition involving a pair of acoustic vibrational modes. The interaction between the Raman-active and acoustic modes results in a parametric excitation of the Raman-active phonon, leading to the emergence of dynamical components at subharmonic frequencies.

LaAlO_3 is a versatile material: as a single bulk crystal, it is commonly used as a substrate for the epitaxial growth of other perovskite oxides, and as a thin film, it constitutes a building block for heterostructures and devices based on functional interfaces [24]. It is a wide gap insulator ($E_{\text{gap}} = 6.1$ eV), with a perovskite structure that undergoes a cubic $R\bar{3}c$ to rhombohedral $\text{Pm}\bar{3}m$ phase transition at $T = 813$ K, associated with the rotation of the AlO_6 octahedra in antiphase about the pseudocubic [111] axis [25]. At the same temperature, the material becomes ferroelastic [26,27], where elastic properties and low-energy optical vibrations are intimately related [28–31]. For our experiments, we use a double-sided polished LaAlO_3 [100] bulk crystal with a thickness of 0.5 mm. The material's nonlinear optical response is measured in a pump-probe experimental configuration, where the pump used is either a near-infrared pulse at 1300 nm or a broadband single-cycle terahertz pulse. In Fig. 1, we show the experimental configuration. The near-infrared radiation is generated via optical parametric amplification of a 40-fs-long pulse centered at 800 nm produced by a 1 kHz Ti: Sapphire amplifier. The pulse duration of the near-infrared pump is 60 fs. Broadband (0.5–3 THz) single-cycle terahertz radiation is generated by optical rectification of the same near-infrared laser pulse in the organic crystal OH1 [32]. The terahertz pulses are focused on the sample using parabolic mirrors to a maximum field amplitude of 700 kV/cm. We use a pair of wire-grid polarizers to control the amplitude of the THz field for conducting field-dependent measurements. The electro-optically sampled

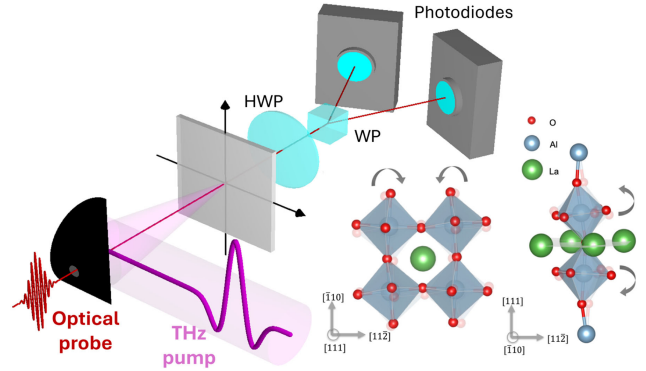


FIG. 1. Sketches of the experiment and of LaAlO_3 unit cell and phonon dynamics. Left: Schematic of the pump-probe setup used in this study. HWP = half-wave plate; WP = Wollaston prism. Bottom right: Crystal structure of LaAlO_3 in the rhombohedral phase and atomic displacements associated with the E_g mode.

THz electric field is shown in gray in Figs. 2(a) and 2(b) in time and frequency domains, respectively. The probe pulses are produced by the same 1 kHz amplifier used to generate the pump radiation. The geometry of the pump-probe setup is shown in Fig. 1. A half-wave plate and a Wollaston prism are used to implement a balanced detection scheme with two photodiodes.

The measurements are performed at 8 K, the temperature at which the material is centrosymmetric with a rhombohedral unit cell. Figure 2 shows the signal arising from the polarization rotation of the probe beam transmitted through the sample after its excitation. Pump and probe polarizations are orthogonal to each other. To facilitate the comparison between near-infrared and THz excitations, the sample response is normalized to the incident pump fluence. A long-lasting oscillation is visible in Fig. 2(a), which illustrates the pump-probe time domain data for both pump wavelengths. Figure 2(b) plots the fast Fourier transform (FFT) of the same time-domain signal. The FFT data are dominated by a narrow peak, whose central frequency of 1.08 THz is comparable to one of the lower-frequency Raman-active modes of LaAlO_3 , i.e., the antiferrodistortive phonon mode with E_g symmetry [33]. As illustrated in Fig. 1(bottom right), the atomic displacements of the mode are associated with the out-of-phase oxygen octahedra rotation about the pseudocubic axes [111].

The data presented in Fig. 2 demonstrate that both the 1300 nm and the broadband terahertz pumps are capable of exciting the E_g mode in the material. When excited with a broadband terahertz field, the lifetime of the material's response is one order of magnitude longer, and it assumes a nonmonotonic decay. Moreover, additional low-frequency spectral components are visible: a quasicontinuum below 1 THz with a broad peak around 0.8 THz and a well-defined peak at 0.3 THz. These lower-frequency contributions are not observed when driving the system with the 1300 nm pump. They are too high in frequency to be

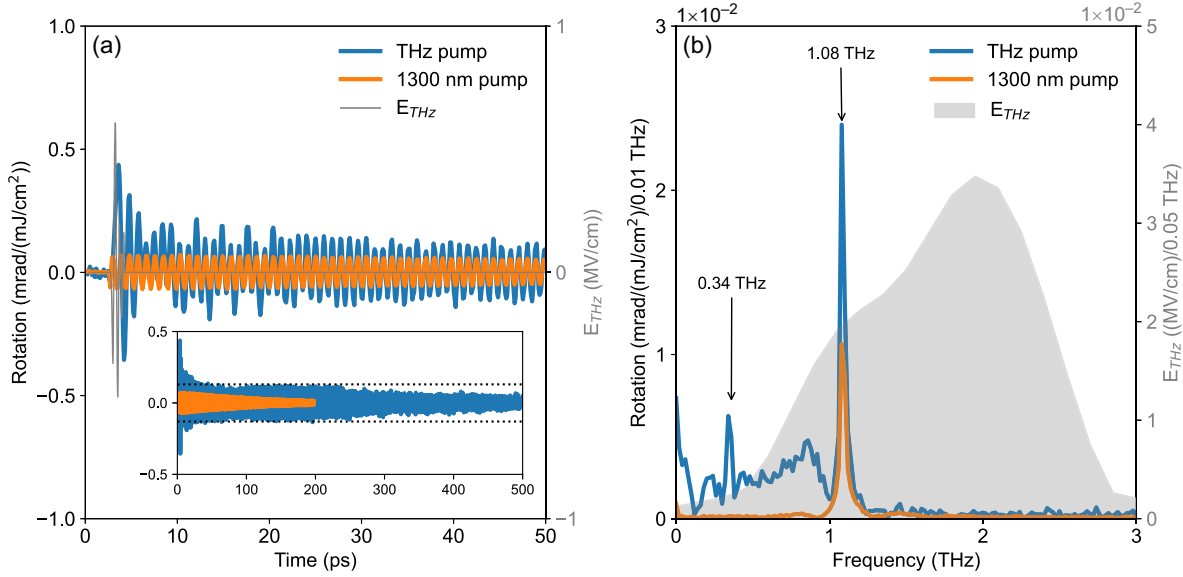


FIG. 2. Experimentally detected polarization rotation in LaAlO_3 following near-infrared or THz pumps. Polarization rotation of a transmitted 800 nm probe through a LaAlO_3 crystal following the excitation by a near-infrared (1300 nm) (orange curve) or a broadband THz excitation (blue curve) in (a) time and (b) frequency domains. Measurements are performed at 8 K. The FFT is evaluated in the range of 0–50 ps. The sample response is normalized by the pump fluence. Inset in panel (a): time-domain dynamics at longer timescales.

ascribed to Brillouin scattering (i.e., direct excitation of acoustic modes) and are too low in frequency to be van Hove singularities of the acoustic modes, which would occur at 3 THz [34]. It is worth noting that in a recent study, the broad peak at 0.8 THz was assigned to spurious effects resulting from the propagation of THz and optical probe pulses in birefringent twin domains [35,36]. On the other hand, another recent investigation, monitoring the THz-driven structural dynamics of a LaAlO_3 thin film by time-resolved x-ray diffraction [37], revealed the presence of a 0.3 THz mode, ruling out any implication of propagation-related effect for this component. Its origin was ascribed to a THz-driven acoustic breathing mode of the film. To gain a deeper insight into these features and into the symmetry of the underlying excitation mechanism, we measured the THz response of the system as a function of the probe polarizations (Fig. S4 in the Supplemental Material [38]). The evidence, shown in the Supplemental Material [38], indicates a similar response function symmetry (i.e., Raman tensor) for both the E_g mode and the low-frequency contributions.

At high temperature, i.e., 120 K (Fig. S5), only the optical pump is still able to excite the E_g mode at 1.08 THz, the response of the THz pump showing a slow background due to the terahertz Kerr effect. This indicates that ISRS and SFG cannot be the only mechanisms responsible for the THz-driven E_g excitation observed at 8 K, and an additional mechanism, more efficient at low T, should be assumed. Finally, the fluence dependence of the THz and optically driven E_g amplitude, presented in detail in Fig. S6 in the Supplemental Material [38], exhibits a linear trend,

whereas the 0.34 THz mode shows a departure from linearity, displaying an additional quadratic behavior. Please note that we ruled out any effect due to local heating induced by the pump intensity, as shown in Sec. II of the Supplemental Material [38]. Building on the above-presented experimental evidence, we now identify the potential excitation pathway responsible for the emergence of both the E_g and the low-frequency peak in the THz-driven sample's response. First, we note that even if the E_g mode is excited by Raman processes in the THz range (i.e., ISRS and SFG), these mechanisms alone are not sufficient to generate the observed low-frequency response. Particularly, the experimental observation of a well-defined sharp peak at 0.3 THz, together with the absence of optical phonons and electronic excitations below 1 THz [33,39], points toward an additional physical scenario involving THz-field-driven parametric excitation of the Raman-active phonon mode. Such a scenario can be captured by a phenomenological model, wherein light triggers two acoustic phonon modes (Q_{ac}) that are subsequently converted into Raman-active phonon modes (Q_R). Our assumption is justified by the improper ferroelastic properties of LaAlO_3 , implying the acoustic branch and the low-energy optical branch (the E_g phonon) mixing at finite momentum, as shown in Fig. S7. This mixing leads to a strong coupling between the acoustic and E_g phonon modes first observed in [40] and extensively discussed in [41]. Moreover, the large energy separation between the optical E_g mode and the next higher-energy optical phonon (A_g at 4.7 THz [42]) makes Raman down-conversion into acoustic phonons the dominant scattering channel for the E_g mode.

Focusing, for simplicity, on a single acoustic branch, the relevant symmetry-allowed terms in the expansion of the potential are

$$\begin{aligned}
 V(Q_R, Q_{ac}) &= \frac{\Omega_R^2}{2} Q_R^2 + \sum_{\mathbf{q}} \frac{\Omega_{ac}(\mathbf{q})^2}{2} Q_{ac}(-\mathbf{q}) Q_{ac}(\mathbf{q}) \\
 &\quad - R Q_R E_{\text{ext}}^2(t) + \frac{1}{2} \sum_{\mathbf{q}} Z_{ac}(\mathbf{q}) Q_{ac}(-\mathbf{q}) Q_{ac}(\mathbf{q}) E_{\text{ext}}^2(t) \\
 &\quad + \frac{1}{4} \sum_{\mathbf{q}} d(\mathbf{q}) Q_{ac}(-\mathbf{q}) Q_{ac}(\mathbf{q}) Q_R^2, \quad (1)
 \end{aligned}$$

The first row of Eq. (1) describes the Raman-active and acoustic phonons oscillating at their respective frequencies, Ω_R and Ω_{ac} . The second row accounts for the ISRS-SFG excitation mechanism of the Raman-active phonon, mediated by the Raman tensor R [42], and the simultaneous excitation of two acoustic phonons with opposite momenta by light—allowed by symmetry and momentum conservation—mediated by the effective charge Z_{ac} . The last row contains the anharmonic term responsible for acousto-optic conversion, controlled by the coupling constant d . We emphasize that, while the Raman-active phonon couples directly to light close to the Γ point, the full momentum dependence of both the acoustic phonons and their couplings to the Raman-active mode and to the external field is retained. As a final remark on Eq. (1), symmetry-allowed terms of the form $Q_{ac}^2 Q_R$ and $Q_{ac} E_{\text{ext}}^2$ are neglected, as they cannot account for the generation of subharmonic components.

According to Eq. (1), light directly excites the Raman-active phonon via ISRS and SFG while simultaneously generating two acoustic phonons from the same branch, which in turn, drive the system parametrically through the acousto-optic conversion mechanism sketched in Fig. 3. Indeed, by integrating out the acoustic modes (see Supplemental Material [38]), one finds that the time evolution of the Raman-active phonon is given by $Q_R(t) = R \int dt' x(t-t') E_{\text{ext}}^2(t')$, where $x(t)$ evolves as

$$\ddot{x}(t) + \Omega_R^2 [1 + f(t)] x(t) = 0, \quad (2)$$

with initial conditions $x(0) = 0$ and $\dot{x}(0) = 1$. Equation (2) describes a harmonic oscillator with a time-dependent modulation of the frequency $f(t) = (1/\Omega_R)^2 \int d\omega / \sqrt{2\pi} e^{-i\omega t} K(\omega) E_{\text{ext}}^2(\omega)$, which results from the convolution of the nonlinear kernel $K(\omega) = \sum_{\mathbf{q}} C(\mathbf{q}) / [4\Omega_{ac}^2(\mathbf{q}) - (\omega + i0^+)^2]$, describing two acoustic phonons with momenta $\mathbf{q}$ and $-\mathbf{q}$, where $C(\mathbf{q})$ selects the dominant phonon contributions, with the squared pump field. In the following, we distinguish between narrow-band and broadband pump fields.

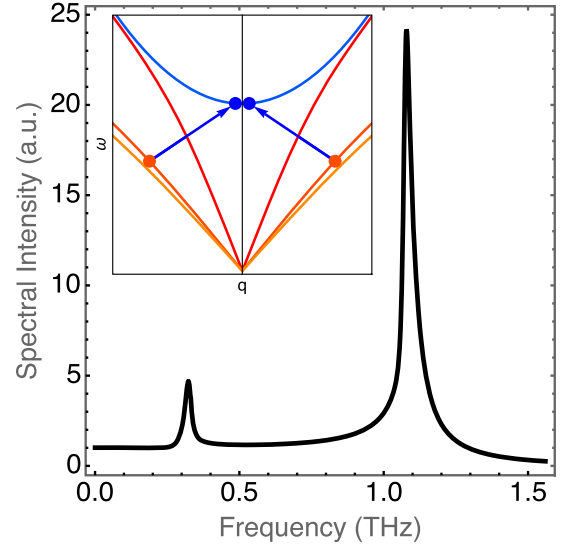


FIG. 3. Theoretical modeling of parametric phonon dynamics in LaAlO_3 . Spectral intensity of the Raman amplitude $|Q_R|$ (solid black line) as a function of frequency. Inset shows the phonon dispersions: the Raman-active optical branch (blue) and three acoustic branches (shades of red). Up-conversion of two acoustic phonons (red dots) into a Raman-active phonon (blue dot) is illustrated.

For a narrow-band monochromatic pump field with frequency Ω_{pump} , $f(t) = \alpha \cos(\tilde{\omega}t)$, where $\tilde{\omega} \equiv 2\Omega_{\text{pump}}$, and Eq. (2) reduces to a Mathieu equation. When $K(2\Omega_{\text{pump}}) \neq 0$, α is finite, and the Raman-active mode undergoes a nontrivial periodic motion that can be analyzed in the frequency domain through the Fourier transform $x(\omega) = \int dt e^{i\omega t} x(t)$. $x(\omega)$ exhibits a prominent spectral component at the Raman characteristic frequency accompanied by smaller components above or below Ω_R . The emergence of subharmonic components below the characteristic frequency is a unique fingerprint of the parametric driving mechanism [43]. To gain deeper insight into these features, we recall that a Mathieu equation with a modulating frequency $\tilde{\omega}$ defines a Floquet dynamical system with a characteristic period $T \equiv 2\pi/\tilde{\omega}$ [43]: this allows for stable solutions that can be expanded in Fourier series as

$$x(t) = \sum_n c_n e^{i(\Omega_R + n\tilde{\omega})t}, \quad (3)$$

where a nonzero coefficient c_n indicates a spectral peak at $\omega = \Omega_R + n\tilde{\omega}$. The solution of the Mathieu equation obtained by substituting Eq. (3) is discussed in detail in the Supplemental Material [38]. Here, we note that in the nonresonant case, when $\Omega_{\text{pump}} \neq \Omega_R$, two main regimes can be distinguished, depending on whether $\tilde{\omega}$ lies above or below $2\Omega_R$. When $\tilde{\omega} > 2\Omega_R$, spectral components appear only at $\omega = \Omega_R$ or higher frequencies. In contrast, when $\tilde{\omega} < 2\Omega_R$, nonzero spectral components appear below $\omega = \Omega_R$; for instance, the Fourier component

corresponding to $n = -1$ occurs at $\omega = |\tilde{\omega} - \Omega_R|$, which is a subharmonic of the main peak, since $|\tilde{\omega} - \Omega_R| < \Omega_R$. This analysis is general and applies whenever Eq. (2) exhibits a Mathieu-like form.

In the case of a broadband pump, such as that used in our experiment, the convolution is dominated by the characteristic frequency of the microscopic process, namely, $\tilde{\omega}(\mathbf{q}) = 2\Omega_{ac}(\mathbf{q})$ for a specific momentum $\mathbf{q}$. Hence, $f(t) = \sqrt{2/\pi}/\Omega_R^2 \sum_{\mathbf{q}} C(\mathbf{q}) E_{\text{ext}}^2(\tilde{\omega}(\mathbf{q})) \sin(\tilde{\omega}(\mathbf{q})t)$, with $C(\mathbf{q})$ and $E_{\text{ext}}^2(\tilde{\omega}(\mathbf{q}))$ filtering the relevant momenta. In particular, a THz field activates smaller momenta for which $\Omega_{ac}(\mathbf{q}) < \Omega_R$, and hence $\tilde{\omega}(\mathbf{q}) < 2\Omega_R$, leading to a nontrivial subharmonic spectrum. A precise comparison between theory and experiment, particularly for the broadband THz field illustrated in Fig. 2, requires the full computation of $f(t)$, which in turn necessitates accounting for the momentum dependence of Z_{ac} and d , whose computation is beyond the scope of the present work. Nevertheless, the experimental observation of a subharmonic peak at ~ 0.3 THz supports the occurrence of a Mathieu-like modulation at a single frequency. Such a modulation is microscopically offered by pairs of acoustic phonons with a specific frequency that parametrically drive the optical phonon. It is worth noting that the proposed scenario is supported by the calculated LaAlO_3 phonon dispersion shown in Fig. S7. Remarkably, the acousto-optic coupling is particularly enhanced between the Raman phonon and the upper acoustic branch along the Γ -X and the Γ -Z direction, within the energy range 1.5–4.0 THz (i.e., 50–133 cm^{-1} and corresponding momenta), where the two branches hybridize. A pair of acoustic phonons from the upper branch, with energy and momentum within this range, can subsequently scatter into two pairs of lower-branch acoustic phonons, with individual frequencies Ω_1 and Ω_2 that are dictated by energy and momentum conservation. Through this scattering channel, each of these pairs then parametrically drives the Raman-active phonon. Assuming an isotropic acoustic-phonon dispersion in momentum space, with sound velocities $c_L \sim 10 \text{ km} \cdot \text{s}^{-1}$ and $c_T \sim 6 \text{ km} \cdot \text{s}^{-1}$ for the longitudinal and two degenerate transverse branches [44], respectively, we consider all possible scattering processes, as detailed in the Supplemental Material [38]. From this analysis, we find that the resulting Ω_1 and Ω_2 values are distributed around an average frequency $\bar{\Omega} \sim 0.7$ THz. Accordingly, we expand $f(t)$ around $\tilde{\omega}(\mathbf{q}) = 2\bar{\Omega}$, so that Eq. (2) reduces, at leading order, to

$$\ddot{x}(t) + \Omega_R^2 [1 + \alpha \sin(2\bar{\Omega}t)]x(t) = 0, \quad (4)$$

where $\alpha = \sqrt{2/\pi} C_0 E_{\text{ext}}^2(2\bar{\Omega})/\Omega_R^2$, with C_0 a constant. Equation (4) is solved by substituting Eq. (3), with $\alpha = 0.2$ and an additional damping term $2\gamma\dot{x}(t)$, where $\gamma = 0.01$ is evaluated from the decay of oscillations at $\omega = \Omega_R$ in the

time domain. In order to compare the theoretical predictions with the experiment, we show the spectral amplitude $|Q_R(\omega)| \equiv |x(\omega)E_{\text{ext}}^2(\omega)|$ of the Raman-active phonon as a function of ω in Fig. 3. Beyond the main peak at Ω_R , associated with a finite Fourier component c_0 , Eq. (2) predicts, without any fine-tuning, the emergence of a subharmonic peak, associated with a finite c_{-1} , oscillating at $\omega_{-1} \equiv 2\bar{\Omega} - \Omega_R \sim 0.32$ THz, in very good agreement with the experimental findings. To further validate the consistency of our model with the observations, we report theoretical estimates for the pump-field dependence of the subharmonic component, which are discussed in detail in the Supplemental Material [38]. While c_0 is independent of the pump field, implying a quadratic pump-field dependence of the main peak (i.e., linear in the pump fluence), the subharmonic coefficient scales as $c_{-1} \sim \alpha$, with α proportional to the squared pump field. Therefore, the Raman amplitude at the subharmonic frequency scales as $|Q_R(\omega_{-1})| \sim |E_{\text{ext}}|^4$. This quartic behavior (i.e., quadratic in the fluence) is confirmed by the experimental pump-fluence dependence of the 0.34 THz mode amplitude shown in Fig. S6(a) of the Supplemental Material [38].

In conclusion, we have presented evidence for the occurrence of phononic parametric driving of a Raman-active mode through the coupling of intense terahertz pulses with acoustic waves. The resulting form of acoustic-optical interaction can explain a physical scenario in which the Raman mode is coherently excited and accompanied by the creation of dynamical components at subharmonic frequencies. Specifically, the ability to induce oscillating modes at frequencies lower than that of the Raman mode is a defining characteristic of the observed phenomenon. It is worth noticing that in the present work, we focused on the description of the origin of the subharmonic feature. The physical origin of long lifetime and nonmonotonous decay of the THz-driven response will be the topic of a future study focused on strain-assisted long living states, as recently suggested by [41]. The implications of our findings are generally vast and carry broad significance for the ultrafast, condensed-matter, and materials-science communities, stimulating further exploration of THz-driven control strategies for phonon-based functionalities. We would like to emphasise that LaAlO_3 , being a widely used substrate and an essential building block for oxide heterostructures, offers a natural platform to exploit the mechanism introduced here in a nonlocal fashion. In particular, the parametrically excited phonon mode could be coupled to other quasiparticle excitations in an overlying film or at an interface, enabling THz control pathways that are not accessible through conventional driving schemes.

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Data availability—The data that support the findings in the work are publicly available [45].

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Correction: The data availability statement has been corrected and the source for the data has been inserted as Ref. [45].