Phonon-Phonon Quantum Coherent Coupling in GaAs/AlAs Superlattice

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Abstract:
We report the observation of quantum coherent coupling at ambient temperature between a zone-center phonon at 330 GHz and two acoustic phonons in GaAs/AlAs superlattice. Fluence-dependent measurement with coherent phonon spectroscopy clearly revealed the transition from weak coupling to strong coupling regimes. After passing a threshold, the energy exchange rate between the driving and target phonons increased nonlinearly with pump laser fluences. Extrapolation with experimental data suggests that an extreme state may eventually be reached, where all three phonons share the same coherent state and become indistinguishable. This extreme state has some similarities to the “quantum entanglement” state between photons.

Keywords: quantum coherent coupling, coherent phonon spectroscopy, superlattice

Introduction
Quantum computing, which utilizes quantum mechanical phenomena that has no classical counterpart, may enable performing enormous computational tasks not possible with traditional computers. Coupling among quantum states forms the basis of quantum computing. When the coherent energy exchange rate between two quantum states exceeds their decaying rates to the background, this interaction is called quantum coherent coupling (QCC). So far, QCC has been observed in many different quantum systems, including photon-mechanical vibration [1-6], photon-electron [7], photon-exciton [8], and electronic energy transfer in complex biological and
chemical systems [9]. In the optical-mechanical system, reversible coherent energy exchange is observed between the optical field (photon) and the mechanical oscillator (phonon) [1-6]. In biological and chemical systems, QCC is found important even at ambient temperature [9]. For phonons, the quantized quasi-particle that involved in lattice vibrations and heat transfer, its quantum nature by far enjoys much less attention comparing with photon and electrons. QCC among phonons are more difficult to observe experimentally because: (1) Driving certain phonon modes coherently to the nonlinear region requires high-power and ultrashort laser pulses, which can easily damage the material itself. (2) In bulk materials, density of states of acoustic phonons near first Brillouin zone center, that usually have longer lifetime and are easily accessible via optical approaches, is very small, which limits the number of phonon-phonon coupling channels that QCC could potentially occur.

In 1966, Orbach predicted a phonon QCC regime where the driving phonon mode selects a particular channel to release its energy [10]. If the driving phonon is a zone center phonon, usually the target phonons would be acoustic phonons at half energy with opposite wavevectors, to satisfy the energy and momentum conservation. The coupling of this three-phonon system (one driving mode and two target modes) could be very strong such that all three modes share the same coherent state, meaning, it is not possible to differentiate them. The energy can flow back and forth freely between the driving and target modes. This process was termed “phonon breakdown” but has not been observed experimentally. Recently, Teitelbaum et al. [11] observed a parametric resonance down conversion from the photoexcited coherent A$_{1g}$ phonon into two acoustic phonons in single crystalline Bismuth. Ultrafast diffuse X-ray scattering recorded the amplitude build-up for a particular acoustic phonon mode when the A$_{1g}$ phonon decays monotonically [11,12]. This is the first time the coupling between specific phonon modes was observed experimentally. However, for the QCC observed in optic-mechanical cavity [1-6], bi-directional energy flow is expected, i.e. energy is coupled from optical mode to mechanical mode (forward process) and verse versa (backward process). For the case of Bismuth [11], only the forward energy transfer process, e.g. decaying of driving mode (A$_{1g}$) and the increasing amplitude of target mode was observed, not the backward process. One possible reason is the population of the driving phonon mode (A$_{1g}$), or the coupling strength between the driving and target modes, did not reached the threshold necessary for “phonon breakdown” [10].
In superlattice (SL) structures, the phonon dispersions are folded into a smaller first Brillouin zone, and more phonon modes fall into the region optically accessible (0 to $2k_{\text{probe}}$, the wave vector of probe laser). With coherent phonon spectroscopy (CPS), phonon modes up to 1.25 THz have been excited coherently in superlattices, such as GaAs/AlAs [13-15], GaN/InGaN[16,17], Bi$_2$Te$_3$/Sb$_2$Te$_3$ [18,19], InGaAs/GaAs [20], YBa$_2$Cu$_3$O$_7$/La$_{1/3}$Ca$_{2/3}$MnO$_3$ [21], and Si/SiGe [22] etc. Among them, GaAs/AlAs SL is an ideal system to study coherent coupling effect between different phonon modes, because the lattice mismatch between GaAs and AlAs is ~0.1% (5.653 Å for GaAs and 5.660 Å for AlAs) [23], and high quality interfaces can be fabricated with molecular beam epitaxy (MBE). Raman spectroscopy has revealed SL-related acoustic phonon modes up to 2.7 THz [24] and CPS detects coherent phonons up to 1.25 THz [25]. Furthermore, some intriguing phonon phenomena have been observed in GaAs/AlAs SL, including coherent thermal phonon transport [26,27], phonon localization [28] and coherent amplification of phonons [29,30]. In this study, we utilized CPS (degenerate pump and probe at 800nm) to excite high frequency zone center coherent phonons in GaAs/AlAs SL and study the coherent coupling among different phonon modes. At low pump fluences, we only observed a monotonic decay of coherent phonon at 330 GHz to the background. When the pump fluence reaches 53.3 $\mu$J/cm$^2$, shown clearly is the feature of quantum coherent phonon-phonon coupling: initial fast draining of energy from the driving phonon (330 GHz) to target phonon modes and then feeding back to the driving mode. When the fluence increases further, this energy exchange process keeps becoming faster and may eventually reach to an extreme state similar to “quantum entanglement”.

**Experiments**

As shown in Fig. 1a, the SL sample studied was a 30-period GaAs/AlAs SL (8 nm/8 nm, period $D =$16 nm) grown on GaAs (001) substrate with molecular beam epitaxy (MBE). With a mode-locked Ti: Sapphire femtosecond laser (Tsunami, Spectra Physics), degenerate pump-probe experiment was performed in a non-collinear reflection geometry at room temperature. Both pump and probe pulses have an 800 nm central wavelength, 35 fs (~258 fs incoming on the sample surface) pulse width (FWHM) and 76 MHz repetition rate. At this wavelength the pump photon energy (1.55 eV) lies above the bandgap of 8 nm GaAs (1.5 eV from photoluminescence spectra, Fig. S2), but below that of AlAs (3.03 eV)[23]. Therefore, the AlAs layer will appear to be transparent and only the GaAs layers will absorb photons and have photo-carriers excited. The excited carriers will
change the electronic distribution in GaAs, leading to mechanical stress via the deformation potential and generate strain [31-33]. Similar to the Raman spectra selection rules [34], coherent acoustic phonon (CAP) is generated through a pair of photons in the pump pulse due to its large spectrum and the coherence is transferred from the photon field to the phonon field. The absorption depth for GaAs at 800 nm is 743.2 nm [35], much larger than the SL film thickness. Pump and probe beams were focused onto the sample surface by a 10x objective lens, with spot sizes (diameter at the $1/e^2$ intensity level) of 13.38 $\mu$m and 6.69 $\mu$m respectively.

Results and Discussion

![Figure 1](image)

Figure 1 (a) Coherent phonon oscillations after subtracting the slowly varying background, measured at the pump fluence of 89 $\mu$J/cm$^2$. Inset: experiment schematics and the SL arrangement (green: GaAs, orange: AlAs). (b) Short-time Fourier Transform (STFT) for the experimental data presented in Fig. 1(b). A clear collapse and revival feature is observed in the FFT amplitude for multiple frequencies.

Fig. 1a shows the coherent phonon oscillations after removing the slowly varying background signal with a digital FFT filter (Raw data displayed in Fig. S5 in SM). The oscillations include high frequency components with modulated envelope. Short-time Fourier Transform (STFT) was used to analyze the data to reveal features in both frequency and time domain, as shown in Fig. 1b. The FFT amplitude clearly shows a multi-cycle collapse and revival feature with time for both the modes around 330 GHz and 280 GHz. For simplicity, we will only focus on the phonon mode around 330 GHz in the following discussion.
Figure 2  Time-resolved FFT amplitude around 330 GHz under different pump fluences: (a) 17.8 $\mu J/cm^2$; (b) 35.6 $\mu J/cm^2$; (c) 53.3 $\mu J/cm^2$; (d) 62.2 $\mu J/cm^2$; (e) 84.6 $\mu J/cm^2$; (f) 97.8 $\mu J/cm^2$. At low laser fluence, 400 times of scans are averaged to achieve good signal to noise ratio.

To further understand this phenomenon, we also conducted experiments at different pump fluences, as shown in Fig. 2. The data suggest three distinct stages. For the lowest two fluences, the FFT amplitudes only show monotonic decaying feature. For the highest two fluences, two isolated regions in time were observed, which suggest phonon amplitude decreases to a minimum value and then starts to increase again. At the two intermediate fluences, two separate regions start to appear but not yet fully isolated.

A natural question to ask is what the physical origin of the phenomenon observed here is. In 2004, a similar collapse and revival feature of $A_{1g}$ phonon amplitude was observed in single crystalline Bismuth and was attributed to the nonclassical states/quantum beats [36]. About 10 years later the same group redid the experiments and confirmed that the collapse and revival feature was actually an artificial effect from inhomogeneous excitation of the coherent phonons,
which occurred because the pump and probe beam sizes are the same [37,38]. In our experiment, the pump/probe size ratio is about 2, so we do not expect any artificial effect from inhomogeneous excitation. We did measure a bismuth thin film with our experimental setup, and only observed monotonic decay of phonon amplitude (Fig. S7). In GaAs/AlAs microcavity capped with thick distributed Bragg reflectors (DBR) made of Al\textsubscript{0.18}Ga\textsubscript{0.82}As/AlAs layers at two ends, a similar dip in the phonon amplitude was observed in the cavity modes (20 GHz and 60 GHz) around 900 ps [29]. This feature was explained as a result of the interference/coupling between the cavity modes and some leaking modes of similar frequency reflected back from the cap layer. Since we do not have the thick cap layers in our sample, there is no chance of having any leaking modes reflected back into the superlattice. Furthermore, the phenomena observed in Ref. [29] is independent of fluence, which differs from what we observed. The third possibility is Rabi Oscillation between two phonon modes. Rabi Oscillation is a quantum beats feature that originates from a strong coupling effect in any two-level quantum system, where the system can cyclically absorb and re-emit photons. However, experimentally, Rabi Oscillation usually displays a feature of $\sin^2\left(\frac{1}{2} \Omega t\right)$ that corresponds to the probability in any of the two states [39-43], where $\Omega$ is the Rabbi frequency. In Fig. 3 we plot time dependent FFT amplitude for 330 GHz at various fluences, the trends are far away from $\sin^2\left(\frac{1}{2} \Omega t\right)$. As a result, what observed here is a new phenomenon.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure3.png}
\caption{Time dependent FFT amplitude at the phonon frequency of 330 GHz at different pump fluences ($\mu J/cm^2$). Inset: phonon resonant parametric process, for which the zone center mode $\omega_1$ in the SL is coupled to two bulk acoustic modes with half the energy $\omega_0$ and opposite wavevector $\pm q$. $a$ is the lattice constant and $D$ is the period thickness for the SL.}
\end{figure}
Consistent with Fig. 2, the time-dependent FFT amplitudes shown in Fig. 3 also exhibit three distinct regions with pump fluence:

(1) At the lowest two fluences, 17.8 and 35.6 $\mu J/cm^2$, the decay of FFT amplitude could be fitted with a single exponential function (Fig. S10). The phonon dephasing time extracted for these two fluences are $50.6 \pm 5.9$ ps, and $47.7 \pm 4$ ps. This regime is called weak coupling region, where scattering with background (including photoexcited) electrons and incoherent phonons dominate. The phonon lifetime in this region stays almost constant and is governed by the density of background electrons and incoherent phonons.

(2) At the three medium fluences, 53.3-$71.1$ $\mu J/cm^2$, the initial FFT amplitudes firstly decay with a rate much faster than that of the weak coupling region, and bounce back to a higher value which peaks around 130 ps, and then decay again. It is noteworthy that for all these three fluences the experimental curves almost overlap. The observed phenomena could be explained well by a picture of strong coupling between the driving mode ($\omega_1$ as shown in the inset of Fig. 3) and two target modes ($\omega_0$), a pair of acoustic phonons with half of the energy ($2\omega_0 = \omega_1$) and opposite wave vectors ($q$ and $-q$). In the field of nonlinear optics, the resonant parametric down-conversion process converts one photon of higher energy into a pair of lower energy photons with equal energy, and resonant parametric up-conversion process will behave vice versa [43]. These two processes can happen simultaneously when the phase match condition is achieved. For phonons in this nonlinear regime, due to the large coupling strength between the driving and target modes, energy in the driving phonon mode flows into target modes very rapidly (forward process), exceeding that into the background electrons and phonons. At around 70 ps, the FFT amplitude of $\omega_1$ reaches a minimum point and starts to increase again, indicating an energy transfer from the target mode back to the driving mode (backward process). Very similar phenomena has been observed in the case of QCC between a mechanical oscillator and an optical cavity mode, e.g. multi-cycle oscillations between coherent optical and mechanical excitations [3]. The forward energy transfer process from the driving to target mode is similar to the one observed in Bismuth [11], where the backward process was not observed. We used a model describing coupled Harmonic Oscillators to simulate the coupling process between $\omega_0$ and $\omega_1$ (Section VII in supplemental material). To generate a feature observed in Fig.2 c~f, the coupling factor is estimated in the range of 2.94-3.67 (Fig. S11), which is much larger than $g_q = 0.7$ in the Bi [11]. Due to the limitation with optical probing, we were unable to monitor the time evolution of target
modes. The bulk acoustic modes along the in-plane direction that satisfy both energy and momentum conservation are the most possible candidates of target modes [44].

Fig. 4a plots the initial FFT amplitude ($A_{\text{initial}}$) and that of the second peak ($A_{\text{2nd}}$) with pump fluence. In the weak coupling region, $A_{\text{initial}}$ increases linearly with pump fluence, and there is no obvious second peak. In the strong coupling region, $A_{\text{initial}}$ starts to saturate, and the $A_{\text{2nd}}$ stays almost constant. The slope of the time-dependent FFT amplitude, $dA_{\text{FFT}}/dt$ shown in Fig. 3, could serve as an indicator of energy transfer rate between the driving and target modes. For simplicity, when extracting the slopes for the backward process, only the linear region is used (Fig. S10). At the three medium fluences, 53.3~71.1 $\mu J/cm^2$, the energy transfer rate of forward process (200~260 $ps^{-1}$) is almost twice of those of backward process (100~150 $ps^{-1}$), and do not show significant fluence dependence. Since the energy exchange rate between the driving and target modes far exceeds their decay rates into the bath (~0.02 $ps^{-1}$), the coupling in this region is “quantum coherent” in nature.

(3) When pump fluence further increases, 84.6~97.8 $\mu J/cm^2$, the minimum turning point in FFT amplitude occurs at earlier time (Fig. 3), which indicates faster energy transfer. In addition, $A_{\text{2nd}}$ increases very rapidly with pump fluences (Fig. 4a), even though the $A_{\text{initial}}$ stays almost constant, which indicates that the amount of energy returned from the target mode to the driving phonons becomes larger. Moreover, both the energy transfer rate of the forward and backward processes increase with pump fluences, while the backward process increases faster (Fig. 4b). At the fluence of 97.8 $\mu J/cm^2$, the backward energy transfer rate becomes almost the same as the forward process.

Fig. 4a also plots the ratio of $A_{\text{2nd}}/A_{\text{initial}}$ (red dots), which increases nonlinearly with pump fluence. It is plausible that if the pump fluence keeps increasing without damaging the sample, (which is not achievable with our current experiments), $A_{\text{2nd}}$ might eventually become equal to $A_{\text{initial}}$. To estimate at what fluence this extreme state might occur, we used a simple exponential function to fit the fluence dependent $A_{\text{2nd}}/A_{\text{initial}}$ ratio in Fig. 4a: $\frac{A_{\text{2nd}}}{A_{\text{initial}}} = R_0 + B e^{-\frac{F-53.3}{T}}$. Where $R_0$, $B$ and $T$ are the fitting factors. The pump fluence at which $\frac{A_{\text{2nd}}}{A_{\text{initial}}} \sim 1$, is estimated to be: 112.95$\mu J/cm^2$. At this fluence, extremely long lifetimes of both driving and target modes are expected since the energy flows freely among them without obvious decay. Moreover, if the minimum turning point keeps moving to earlier time, it might reach a state that the energy
exchange rate between the driving and target modes becomes extremely fast (imagine the curves in Fig.3 for forward and backward process are vertical lines). In addition to the possible extremely long lifetimes for both driving and target modes, this extreme state describes a state very similar to “quantum entanglement”, where almost all energy is confined among these phonon modes and energy exchange is so fast that it is not possible to distinguish them.

![Image](image_url)

**Figure 4** (a) FFT amplitudes at various pump fluence (blue), as well as the ratio between the second peak and initial amplitude (red); (b) Energy coupling rate between the driving and target modes for both forward and backward processes, estimated with the gradient extracted from Fig. 3.

**Discussion and Conclusion**

In conclusion, we have used coherent phonon spectroscopy to study power-dependent coherent phonon dynamics in a GaAs/AlAs 8 nm/ 8 nm superlattice structure. The revolution of FFT amplitude of the center mode at 330 GHz ($\omega_1$) extracted from STFT method has shown a collapse and revival signature. We propose there is quantum coherent coupling between $\omega_1$ and two bulk acoustic phonon modes ($2\omega_0 = \omega_1$) along in-plane direction. This work suggests an exciting opportunity to use high quality SL structures to manipulate coherent phonon propagation in the nonlinear region and to obtain quantum control of large wave vector acoustic phonons which are difficult to directly excite via driving certain zone-center modes. Extrapolation of experimental data suggests an extreme QCC state where all three phonons share the same coherent state and become indistinguishable. In our current experimental system, it is impossible to reach the proposed extreme state without sample damage. Nevertheless, it is possible to apply electrical...
bias[45] to further enhance the coupling constant, which could open a pathway to achieve phonon entanglement.

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Supplemental Material: Phonon-phonon Quantum-coherent Coupling in GaAs/AlAs Superlattice

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I. Sample Information
II. Experimental Setup
III. Raw Data
IV. Bismuth Measurement
V. Saturation
VI. Fitting
VII. Harmonic Oscillator
I. Sample Information

Figure S1  (Left) Schematic SL structure (not to real scale); (Right) Sample information with XRD characterization. The $SL_{(0)}$, $SL_{(-1)}$ and $SL_{(+1)}$ peaks are shown for both SL8/8 and SL8/8 without AlAs Etching Release layer while the AlAs (001) peak only shows up in the SL8/8.

The sample studied is GaAs/AlAs SL grown with molecular beam epitaxy (MBE) by Nathaniel Sheehan from Dr. Seth Bank’s group. High quality SL structures will be grown on GaAs (001). As shown in Fig. S1, a 500 nm AlAs etching layer and a 150 nm GaAs buffer layer are grown on (001) GaAs wafer substrate for further transferring process. The SLs are prepared 8 nm/8 nm with 30 periods. The absorption depth for GaAs at 800 nm is 743.2 nm [1].

Characterization of the period thickness for GaAs/AlAs SL sample sets have been performed with XRD (see Fig. S1 right), where we can see the significant peaks for $SL_{(0)}$, $SL_{(-1)}$ and $SL_{(+1)}$. We have also used photoluminescence (PL) (see Fig. S2) to determine the bandgap for SL sample. Raman spectroscopy has been applied to reveal the folded acoustic phonons [2,3] in these semiconductor SLs. PL and low cut-off Raman spectroscopy are taken under the help with Jason in Dr. Lin’s lab. In Fig. S2, we can clearly see that with decreasing periodicity, the band gap is increasing, which can be well explained in quantum mechanics, i.e., the narrower the quantum well is, the higher the ground state is.
Figure S2  PL of SL8/8 structure and GaAs substrate: with thinner SL periods, PL has blue shifts (red line is for eye guidance of our 800 nm laser).

Low cut-off Raman spectra for the sample sets can be seen form Fig. S3(b), aiming to reveal the 2\(k\) doublets modes. The GaAs substrate shows no Raman peaks in the range of 20-100 cm\(^{-1}\) while the GaAs/AlAs sample sets all show Raman peaks in this range.

Figure S3  (a) Raman spectra for sample SL8/8 and GaAs substrate; (b) Calculated dispersion curve for LA phonons within the first mini-Brillouin zone using Eq. (S1). Marked frequencies are the 2\(k_{\text{probe}}\) modes.
From the elastic continuum model, the phonon dispersion relation in a superlattice structure with period thickness $D = d_A + d_B$ is given by [2,4-6]

$$\cos(qD) = \cos\left(\frac{\omega d_A}{v_A}\right) \cos\left(\frac{\omega d_B}{v_B}\right) - \frac{1 + \delta^2}{\delta} \sin\left(\frac{\omega d_A}{v_A}\right) \sin\left(\frac{\omega d_B}{v_B}\right)$$  \hspace{1cm} (S1)

where $\delta = \rho_A v_A / \rho_B v_B$, $\rho_A$ and $\rho_B$ are the corresponding mass density and $v_A$ and $v_B$ are the sound velocities for phonons along [001] for GaAs and AlAs respectively. Fig. S3(a) shows the Raman spectra for 8/8 and Fig. S3(b) shows the calculated LA dispersion curve for SL 8/8. By comparing the observed doublets with the computed doublets, we can see they match. To better match the experimentally observed doublets, further adjustment of the thickness for GaAs and AlAs will be needed.

II. Experimental Setup

With a mode-locked Ti: Sapphire femtosecond laser (Tsunami, Spectra Physics), one-color pump-probe experiment is performed in non-collinear reflection geometry at room temperature. Both pump and probe pulses have an 800 nm central wavelength, 258 fs pulse width (FWHM) and 76 MHz repetition rate. Pump and probe beams are focused onto the sample surface by a 10x objective lens, with spot sizes (diameter at the $1/e^2$ intensity level) of 13.38 $\mu$m and 6.69 $\mu$m respectively. An optical delay stage in the probe beam is used to control the delay time between the pump and the probe beams. The differential reflectivity $\Delta R/R$ is recorded by a Si detector (DET100, Thorlabs). A lock-in amplifier (model 7265, Singal recovery) with a chopper working at 2.7 kHz is used to acquire the data. The white light and the CCD camera are used to observe the sample surface. More averages are needed for detecting high frequency coherent phonons.
Figure S4  CPS system with oscillator: In the one-color pump-probe experiment, we use the polarizer to make the pump and the probe light perpendicular and use another polarizer in front of the detector to filter out the pump signal.

III.  Raw Data
Figure S5  Reflectivity change of SL8/8 at the pump fluence of 89 μJ/cm². Inset is the coherent phonon oscillations after subtracting the slowly varying background and times a factor of 100.

IV.  Bismuth Measurement
Figure S6  (a) Reflectivity change signal for Bismuth at the pump fluence of 15.4 $\text{mJ/cm}^2$; (b) Coherent phonon signal after subtracting the slowly varying background; (c) Short-time Fourier Transform (STFT) for the experimental data presented in (b).

V. Saturation
The electron signal is shown in Fig. S7 (a) and the peak values are plotted with power in (b). We can see the peak values are linearly increased with power under 15 $mW$, then starts to saturate at higher powers.

![Figure S7](image1.png)

Figure S7  (a) Fluence-dependent electron signal; (b) Peak value vs. incident power (the red line is a linear line for eye guidance).

Figure S8  Schematic of simplified band structure of SL 8/8 where the smaller band gap $E_g$ is for bulk GaAs (1.424 $eV$) and $E_g'$ for bulk AlAs (3.03 $eV$). The $n = 1$ is the first quantized energy level for the SL which is around 1.5 $eV$. The 1.55 $eV$ shown in the figure is our laser photon energy.

![Figure S8](image2.png)

From the PL for SL 8/8 in Fig. S2 (a), we can see the energy gap is around 1.5 $eV$ and our pump photon energy (1.55 $eV$) is slightly larger than it (see Fig. S8). The mechanism for light-matter interaction in the SL is the energy and coherence of photons will first transfer to the electrons and then the electrons will transfer the energy and coherence to the phonons in a short time (~hundreds of femtoseconds). SL first absorbs the incident photons and the excited electron population will increase linearly with photon number (incident power). Once the density of states (DOS) of electrons in the energy level $n = 1$ is fully occupied, no more photons can be absorbed, and the excited population of electrons saturates. Therefore, the energy transferred from electrons to phonons is also saturated.
VI. Fitting

![Figure S9](image)

Figure S9  Single Exponential Fitting of (a) 17.8 μJ/cm²; (b) 35.6 μJ/cm² and the extracted phonon dephasing time are 50.6 ± 5.9ps and 47.7 ± 4ps respectively.

![Figure S10](image)

Figure S10  Linear fit of linear region for forward and backward process at fluences of (a) 53.3μJ/cm²; (b) 71.1 μJ/cm² and the extracted the slopes.

VII. Harmonic Oscillator

The Hamiltonian of this system, describing by annihilation (creation) of a coherent zone center mode $\omega_1$, a pair of target modes ($\omega_0, \pm q$) are created (annihilated), is given by

$$
\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_{int}
$$

$$
\mathcal{H}_0 = \hbar \omega_1 \left( b_1^+ b_1 + \frac{1}{2} \right) + \sum_q \hbar \omega_q \left( b_q^+ b_q + \frac{1}{2} \right)
$$

(S2)
\( \mathcal{H}_{\text{int}} = \hbar \sum_q g_{01} (b_1 b_q^\dagger b_{-q}^\dagger + b_1^\dagger b_q b_{-q}) \)

Where \( \mathcal{H}_0 \) is the free phonon Hamiltonian for the zone center mode and target modes in the harmonic approximation and \( \mathcal{H}_{\text{int}} \) is the interaction Hamiltonian between the zone center mode and target modes. \( b_q^\dagger \) and \( b_q \) are the annihilation and creation operators for the phonon mode described by \((\omega_0, q)\). \( g_{01} \) is the coupling constant between \( \omega_1 \) and \( \omega_0 \). They can be written in terms of normal mode coordinate \( Q \) and momentum \( P \) as:

\[
\mathcal{H} = \frac{1}{2} (p_1^2 + \Omega^2 Q_1^2) + \frac{1}{2} (p_0^2 + \omega_0^2 Q_0^2) + g_{01} Q_1 Q_0
\]  

(S3)

where the first two terms describe free phonon Hamiltonian for the zone center mode and the target modes, while the third term describes the interaction Hamiltonian. Therefore, by applying the Heisenberg equations on Eq. (S3), we can obtain the equations of motion as:

\[
\ddot{Q}_1 + \gamma_1 \dot{Q}_1 + \omega_1^2 Q_1 = -g_{01} Q_0^2 + f(t) \tag{S4}
\]

\[
\ddot{Q}_0 + \gamma_0 \dot{Q}_0 + \omega_0^2 Q_0 = -2g_{01} Q_1 Q_0 \tag{S5}
\]

where \( f(t) \) is for the laser pulse field, which in our case is a Gaussian pulse with FWHM of 258 fs and \( \gamma_1 \) and \( \gamma_0 \) are the phenomenological damping constant for \( \omega_1 \) and \( \omega_0 \) respectively. We can treat the r.h.s. as the driver term in the mechanical oscillators.

Figure S11  Simulation of mean displacement for \( \omega_1 \) and \( \omega_0 \) in the strong coupling region with the coupling constant to be \( g_{01} = 0.1 \) (a) and \( g_{01} = 0.08 \) (b).

By solving the coupled equations (S4) and (S5) simultaneously with parameters \( \gamma_1 = 0.05 \text{ ps}^{-1} \) and \( \gamma_0 = 0.0125 \text{ ps}^{-1} \) which are based on the theoretical three-phonon scattering trend
yielding a $1/\omega^2$ frequency dependence [7], in addition with $g_{01} = 0.1$ and $g_{01} = 0.08$, we can produce the mean displacement of $\omega_1$ and mean square displacement for $\omega_0$. Figure S11 (a) & (b) plot the mean displacement of $\omega_1$ and the square root of mean square displacement $\omega_0$ in comparison. We can estimate the $g_{01}^* = g_{01}/\omega_0^2$ is in the range of 2.94-3.67 where the dip shows up between 51 $ps$-74 $ps$, which is larger than $g_q = 0.7$ in the Bi [8]. Further detailed fitting processes are needed to determine the exact coupling constant.

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