Generation and coherent control of terahertz acoustic phonons in superlattices of perovskite oxides

Chi-Yuan Yang, Ping-Chun Wu, Ying-Hao Chu and Kung-Hsuan Lin

Institute of Physics, Academia Sinica, Taipei 11529, Taiwan
Department of Materials Science and Engineering, National Chiao Tung University, Hsinchu 30010, Taiwan
Department of Physics, National Cheng Kung University, Tainan 70101, Taiwan

E-mail: linkh@sinica.edu.tw

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Abstract

We utilized transmission-type pump–probe technique to investigate coherent acoustic phonons in the superlattices of perovskite oxides such as SrIrO$_3$/SrTiO$_3$ and SrRuO$_3$/SrTiO$_3$. Because the films in the superlattices are of high-quality and their thicknesses are only several monolayers, quasi-monochromatic acoustic phonons with THz frequency have been achieved. By investigating the propagation of coherent acoustic phonons in the superlattices with different epitaxial periods, the phonon mean free path of SrTiO$_3$ were studied in the frequency range between 0.5 THz and 1 THz. We further demonstrated coherent control to amplify or cease the THz coherent acoustic phonon oscillations in the superlattices. By controlling the delay of two pulses for shining the superlattices, the amplitude and phase of the THz coherent acoustic phonons were manipulated. According to the measurements of time-domain Brillouin scatterings, we found the optoacoustic conversion efficiency of SrIrO$_3$ outperforms that of SrRuO$_3$ for generating acoustic phonons.

1. Introduction

Perovskite oxide materials have attracted much attention recently and have become emerging materials which have potential to new applications. One of the interesting discoveries was the conducting interface between two insulating complex oxides that triggered a series studies of interfacial anomalies in numerous fields such as conducting interface, superconducting to photo-material interaction, and multiferroicity, etc [1–5]. The exotic phenomena could originate from the displacement of atoms at the interfaces of heterojunctions, and could be controlled by phonon engineering. For investigating coherent phonons in perovskite oxides, metal thin films could be candidates of phonon transducers, which convert optical energy into acoustic phonons through the electron–phonon couplings [6–9]. But the measured frequency of acoustic phonons is typically below 100 GHz. Sub-THz to THz acoustic phonons and their coherent control have been demonstrated in semiconductor heterostructures, such as GaAs/AlAs and InGaN/GaN [10–15]. However, these semiconductor-based phonon transducers cannot deliver acoustic phonons into the perovskite oxide materials because these two material systems cannot be joined with high quality.

SrRuO$_3$ (SRO) has metallic properties and high absorption in the visible range [16]. Because it is a perovskite oxide material, it can be served as a phonon transducer [17] to deliver acoustic phonons into another perovskite oxide materials. Recently, SRO-based phonon transducers were used to study the acoustic properties of the representative material in perovskite oxide system, SrTiO$_3$ (STO), such as acoustic attenuations [18–20] and domain walls coupled with phonons [21]. Nevertheless, the frequency of acoustic phonon studies in perovskite oxide materials was still within GHz range to date. The acoustic properties in THz regime are important, for example, to heat transfer in nanometer scale. Ballistic phonon transport plays an important role in nanoscaled thermal conduction, and phonon mean free path (MFP) in THz
regime is a key property for investigation [22–25]. A THz perovskite-oxide-based phonon transducer is thus helpful to study ballistic thermal conduction in perovskite oxides.

Two key factors should meet the criteria of generating coherent acoustic phonons (CAPs) in THz regime. First, the driving force should be on the order of sub-picoseconds. Second, the wavelength of acoustic phonons is only several nanometers for THz regime. Spatial modulation of the generated strain profile should achieve length scales of nanometers. In this work, two types of superlattices SRO/STO and SIO/STO were fabricated for generating CAPs up to THz regime. It was investigated that the build-up time of stress through electron–phonon couplings was ~0.5 ps after electrons were generated in the SRO by femtosecond pulses [17]. SRO/STO superlattice was thus investigated in this work since the driving force was expected capable of achieving THz response in SRO. On the other hand, the superlattice structure of SRO and STO serves the spatial modulation of strain for quasi-monochromatic CAPs. In addition to SRO/STO superlattice, the superlattice of SrIrO3 (SIO) and STO was also studied. We demonstrated the generation efficiency of THz CAPs in the emerging material SIO/STO superlattice is better than that in the SRO/STO superlattice.

2. Methods

The high-quality superlattices were fabricated on the STO (001) substrate by pulse laser deposition with atomically control over thickness and interfaces [26]. Four superlattice structures were tailored as follows: (a) 5 pairs of 6 nm SIO/6 nm STO [5-SIO(6)/STO(6)], (b) 5 pairs of 6 nm SRO/6 nm STO [5-SRO(6)/STO(6)], (c) 10 pairs of 3 nm SIO/5 nm STO [10-SIO(3)/STO(5)], and (d) 10 pairs of 3 nm SRO/5 nm STO [10-SRO(3)/STO(5)]. All the superlattices were capped with 40 nm-thick STO thin films. Degenerate transmission-type pump–probe measurements were employed to investigate the CAPs in the superlattice of perovskite oxides. The central frequency of the pump/probe pulses from a Ti:sapphire laser was 800 nm and the repetition rate was 80 MHz. The fluences of pump and probe were ~63 and 40 mJ cm−2, respectively. The duration of the cross-correlation of pump/probe pulses was ~550 fs.

The schematics of figure 1 show the propagation of CAPs in the samples and how they can be monitored with transmission-type pump–probe measurements. Because the bandgap of STO (3.2 eV) is much higher than the photon energy of the optical pulses (1.55 eV), it does not absorb light. In contrast, SRO or SIO are absorbing layers in the superlattices. After optical pulses excite electrons in the superlattices of SRO/STO or SIO/STO, the longitudinal thermal stress in the SRO or SIO layers serves as the driving force to launch longitudinal CAPs propagating in counter direction out of plane of the sample. Figures 1(b)–(e) depict the simulated evolution of strain distribution (in red color) at a few time points. The acoustic reflection at the interfaces of SRO/STO or SIO/STO is neglected in the simulation because the acoustic impedances of different perovskite oxide layers do not differ seriously. The amplitude of the photo-induced strain is assumed to decay exponentially since the intensity of the optical pump beam decays exponentially from the surface of the cap layer to the substrate. The strain pulses toward the substrate leave the superlattice region and propagate into the STO substrate (figures 1(b)–(c)). Meanwhile, the strain pulses toward the cap layer will be reflected back at the surface of the cap layer (figures 1(b)–(c)). Since the acoustic impedance of air is much smaller than that of STO, the polarity of the reflected strain pulses is reversed. Following, the reflected strain pulses will go through the superlattice region again and propagate into the substrate (figures 1(d)–(e)).

When the strain pulses propagate though the absorbing layers such as SRO or SIO, they modulate the absorption, resulting in transmission changes of the optical probe pulses. The transient transmission changes ΔT(t) of the probe beam due to the temporal evolution of strain distribution S(z,t) can be expressed as follows [27]:

$$\Delta T(t) = \int_{-\infty}^{\infty} S(z,t)F(z)dz.$$  (1)

F(z) is the sensitivity function, which is associated with the strain-induced optical change of the probe beam through SRO or SIO. The blue color in figures 1(b)–(e) depicts the profiles of sensitivity functions. The sensitivity is dominated by the region of absorbing layers such as SRO or SIO because the absorption of STO at our probe wavelength is ignorable. And the exponential decay from the surface side to the substrate side reveals the penetration depth of the optical probe beam. Figure 1(f) shows the calculated results of transient transmission changes of the probe pulses. While the strain pulses in counter directions leaving the superlattice region as shown in figure 1(b), the strain pulses crossing the non-zero sensitivity lead to oscillation signals as described in equation (1). This time slot is designated as ‘generation’ as labeled in figure 1(f). And the time point T1 corresponds to the snapshot of strain evolution in figure 1(b). When the strain pulses toward the surface of the sample are echoed back into the superlattice region again as shown in
Figure 1. (a) Schematics of the sample structure. (b)–(e) Simulated evolution of the strain profiles in red color at different time points ($T_1\sim T_4$). The blue colors represent the sensitivity functions. (f) The calculated transient transmission change of the probe pulse due to the strain pulses traveling through the superlattice. Different time points in (b)–(e) are labeled accordingly.

Figure 1(d), oscillation signals appear again. This time slot of oscillations is designated as ‘echo’ as labeled in figure 1(f). The oscillations sustain until the echoed strain pulses completely leaving the superlattice region.

3. Results and discussions

3.1. Generation and propagation of coherent acoustic phonons

Figure 2 shows the transient transmission changes of the probe pulses for the superlattice samples (a) 5-SIO(6)/STO(6), (b) 5-SRO(6)/STO(6), (c) 10-SIO(3)/STO(5), (d) 10-SRO(3)/STO(5). For all the samples, we found oscillations on the relatively slow background curves, which were attributed to the relaxation of carriers in SRO or SIO. Figure 3 shows the CAP-induced transmission changes \( \Delta T(t) \), which were extracted from the curves in figure 2. Details of extraction procedures are discussed in the supplementary materials (https://stacks.iop.org/NJP/23/053009/mmedia). The oscillation frequencies of the CAPs are 0.50, 0.63, 0.75, and 0.97 THz for 5-SIO(6)/STO(6), 5-SRO(6)/STO(6), 10-SIO(3)/STO(5), and 10-SRO(3)/STO(5), respectively. The oscillation period \( \tau = d_1/v_1 + d_2/v_2 \), where \( d_1 \) and \( v_1 \) denote the thickness and the longitudinal acoustic velocity of the material in the superlattice. For 5-SIO(6)/STO(6) and 5-SRO(6)/STO(6) as an example, the structure is the same. The oscillation period of 5-SIO(6)/STO(6) \([0.50 \text{ THz}^{-1} = 2.00 \text{ ps}]\) is longer than that of 5-SRO(6)/STO(6) \([0.63 \text{ THz}^{-1} = 1.59 \text{ ps}]\) since \( v_{\text{SIO}} = 5.96 \text{ nm ps}^{-1} \) is slower than \( v_{\text{SRO}} = 6.31 \text{ nm ps}^{-1} \) [28, 29]. Similar situation is observed for 10-SIO(3)/STO(5) \([0.75 \text{ THz}^{-1} = 1.33 \text{ ps}]\) and 10-SRO(3)/STO(5) \([0.97 \text{ THz}^{-1} = 1.03 \text{ ps}]\).

The time slots ‘generation’ and ‘echo’ are also labeled in figure 3. For example, the oscillation period of 10-SRO(3)/STO(5) is \( \sim 1 \text{ ps} \), corresponding to the transit time of a layer of SRO and a layer of STO. As shown in black curve of figure 3, it takes \( \sim 10 \text{ ps} \) for strain pulses completely leaving the 10 pairs of SRO/STO. On the other hand, the thickness of the STO cap layer is 40 nm, and the round trip time of the strain pulses is 10.1 ps with \( v_{\text{STO}} = 7.9 \text{ nm/ps} \). Therefore, the time slot of echo starts at \( \sim 10 \text{ ps} \).
Figure 2. The transient transmission changes of the probe pulses for the superlattice samples (a) 5-SIO(6)/STO(6), (b) 5-SRO(6)/STO(6), (c) 10-SIO(3)/STO(5), (d) 10-SRO(3)/STO(5).

Figure 3. The extracted signals due to the CAPs from figure 2. The levels of the traces were arbitrarily shifted. The oscillation frequencies of the traces from top to bottom are 0.5, 0.63, 0.75, 0.97 THz, respectively.

and its duration is ∼20 ps for the echoed strain pulses entering and completely leaving the superlattice region.

3.2. Analysis of phonon mean free paths
Before we elaborate the method of analysis to quantitatively acquire the phonon MFP, we first present simulation results that how the phonon MFP affects the signals by including phonon attenuations. Although the CAPs propagate through the different layers of SRO (or SIO) and STO, we used a parameter ‘effective MFP’ to simplify the calculation model instead of using individual MFP for each layer. The amplitude of strain pulses is assumed to decay following the equation $\exp[-\alpha z]$, where $\alpha$ is the effective attenuation coefficient. And the effective MFP is defined as $1/\alpha$. Figure 4 shows the calculated results for
Figure 4. (a) The calculated transient transmission change of the probe pulse due to the strain pulses traveling through the superlattice 10-SRO(3)/STO(5) with no phonon attenuation (in black lines) and with phonon attenuation (in red lines). (b) The solid lines and dashed lines represent the corresponding Fourier spectra of the generation and echo parts of the traces in (a) with corresponding colors. (c) The black dotted line reveals the calculated $A_{E/G}$ as a function of MFP. The red arrows indicate that the MFP can be obtained from $A_{E/G}$.

CAP oscillations in the superlattice of 10-SRO(3)/STO(5). The black line in figure 4(a) reveals the transient transmission when the phonon attenuation is ignored. When the phonon MFP is introduced into the simulation as shown in the red line of figure 4(a), the oscillation amplitude gradually attenuates compared with that of traces without considering attenuation. In order to quantitatively investigate the relation between the amplitude of the oscillations and the phonon MFP, the time slots of generation (0–10 ps) and echo (10–30 ps) in the traces in figure 4(a) are fast Fourier transformed (FFT) as shown in figure 4(b). For the case without attenuation (in black lines), the peak spectral amplitude of the echo is slightly lower than that of the generation. In contrast, for the case with attenuation (in red lines), the peak spectral amplitude
Figure 5. The Fourier spectra of CAP oscillations of (a) 10-SRO(3)/STO(5), (b) 10-SIO(3)/STO(5), (c) 5-SRO(6)/STO(6), (d) 5-SIO(6)/STO(6) from the time slots of generation (in red dots) and echo (in black dots).

of the echo is significantly lower than that of the generation. As a result, the ratio of the spectral amplitudes for echo and generation ($A_{EG}$) at the peak frequency can be utilized to quantitatively reflect the effect of phonon MFP. Figure 4(c) shows the calculated $A_{EG}$ as a function of phonon MFPs. $A_{EG}$ increases with increasing MFPs until it asymptotically approaches to a loss-free value. With this calculated relation curve between $A_{EG}$ and MFP, one can use the experimentally obtained $A_{EG}$ to acquire the effective phonon MFP of the media. For example, if one experimentally obtained $A_{EG}$ of 0.5, it corresponds to a MFP of 175 nm as shown in figure 4(c). We therefore analyzed $A_{EG}$ of the traces for the four samples to acquire the effective phonon MFP with this method.

Figure 5 shows the FFT spectra of CAP oscillations from the four traces in figure 3. The red dots and black dots reveal the spectra of generation and echo, respectively. In figure 5(a), for example, $A_{EG} = 0.2$ can be obtained at the peak frequency 0.97 THz for the sample 10-SRO(3)/STO(5). As aforementioned, the phonon MFP at 0.97 THz can thus be acquired as 73.5 nm by locating the obtained $A_{EG}$ of 0.2 following the calculated curve as shown in figures 4(c) or 6(a). Figure S2 in the supplementary materials shows the good agreement between the experimental trace and the simulation trace in time domain, and confirms the validity of this method of obtaining MFP from $A_{EG}$.

In figure 6(a), several data points are from the results of different measured spots on the sample 10-SRO(3)/STO(5). And for each trace, we also used two different methods to retrieve the CAP oscillation signals. Detailed discussions can be found in the supplementary materials. Discrepancy of $A_{EG}$ values reflects the inhomogeneity of the sample, which will be discussed later in this section. According to the results from figure 6(a), the effective MFP of the sample 10-SRO(3)/STO(5) at 0.97 THz was obtained as 79 ± 11 nm. Similarly, we analyzed the $A_{EG}$ values for 10-SIO(3)/STO(5) at 0.75 THz (figure 5(b)), 5-SRO(6)/STO(6) at 0.63 THz (figure 5(c)), and 5-SIO(6)/STO(6) at 0.50 THz (figure 5(d)), respectively. By using the calculated curves as shown in figures 6(b)–(d), the effective MFPs were obtained as 70 ± 7 nm at 0.75 THz, 117 ± 23 nm at 0.63 THz, and 209 ± 61 nm at 0.50 THz, respectively.

These effective MFP values primarily reflect the MFP of STO since the CAPs propagate in the STO for ~80% of the analyzing duration for the four samples. According to our estimation, 80% of the aforementioned effective MFP values indicate the low bound estimates of the MFP of STO. Detailed discussions can be referred to the supplementary materials. One should note that the obtained effective MFPs do not completely reflect the propagation loss of STO. They include another mechanisms of losses. For example, the interface roughness of the superlattice will result in a scattering loss. In addition, the
variation in the thickness of the STO cap layer or the surface roughness of the STO cap layer will also lead
to a scattering loss. Therefore, the obtained MFP should be shorter than the intrinsic MFP of STO. In this
work, we argue that the backward scattering loss from the interfaces of superlattices would be ignorable.
The acoustic impedances of STO, SRO, and SIO are 40.6, 41.0, and 47.8 Gg m$^{-2}$ s$^{-1}$, respectively. Since the
acoustic impedance of SRO or SIO is close to STO, the reflectivity between them are smaller than 1%. The
backward scattering due to the interfaces especially for SRO/STO interfaces should be thus negligible. But
the forward scattering at the interfaces due to the velocity mismatch might not be neglected. In addition,
the backward scattering loss due to the surface roughness should be significant because the reflectivity at
the STO/air interface is almost 100%. And for such a high frequency up to THz regime, the acoustic wavelength
is only tens of nanometers. Even though the roughness of the STO surface is $\sim$ 1 nm, it could still result in
significant diffusive scattering and reduce the amplitude of the echoed CAP oscillations, leading to lowered
$A_{EG}$ and effective MFP we measured.

We tried to correct the effect of scattering loss from the STO surface by using a model [30, 31] to
estimate the specular probability. Ideally, the specular reflectivity ($R_{sp}$) of the STO/air interface is close to 1.
However, if the acoustic phonons are reflected by a rough surface, the reduction of specular reflectivity will
lead to reduced amplitude of the echo signals and $A_{EG}$. The contribution of roughness loss to the measured
$A_{EG}$ ($A_{EG, measured}$) might be removed by using the relation $A_{EG,corrected} = A_{EG, measured}/R_{sp}$. By using the
corrected $A_{EG}$ ($A_{EG,corrected}$) and the calculated curves as shown in figure 6, the obtained MFP should
increase and be closer to the intrinsic MFP of STO. The specular reflectivity can be represented as [30, 31]

$$R_{sp}(\omega) = \exp \left(-2k^2h^2\right) \cdot \left\{ 1 + \int_{0}^{\infty} \cos(kz) \frac{J_1(\omega)}{z} \left[ \exp \left(-k^2h^2(C(z)-1)\right) - 1 \right] \mathrm{d}z \right\},$$

where $h$ is the root-mean-square (rms) of the surface height, $k$ is the wave number, $J_1$ is the Bessel function
of the first kind, and $C(z)$ is the surface correlation function. We have examined the STO surfaces of the
samples by using atomic force microscopy (AFM). The representative AFM results are shown in figure S3 in
the supplementary materials. From the AFM results, the correlation lengths (>50 nm) of the four samples
are far longer than the acoustic wavelength between 8 and 12 nm. The surface correlation function
Figure 7. The measured MFP values of STO from the literature and our work. The theoretical curve is shown in dashed line. The inset highlights our work in the frequency range between 0.5 THz and 1 THz.

\[ C(z) = 1 \] was thus used to simplify equation (2) as

\[ R_{\text{sp}}(\omega) = \exp \left( -2k^2 h^2 \right). \] (3)

We have analyzed the AFM images from several locations for each sample, and the ranges of rms surface roughness were 0.54–1.26, 0.9–2.69, 0.39–1.04, and 0.63–1.84 nm for the samples 10-SRO(3)/STO(5), 10-SIO(3)/STO(5), 5-SRO(6)/STO(6), and 5-SIO(6)/STO(6). Figure S4 in the supplementary materials reveals the frequency dependence of specular reflectivity when the averaged values 0.84, 1.10, 0.73, and 0.81 nm were used for \( h \) in equation (3) for the samples 10-SRO(3)/STO(5), 10-SIO(3)/STO(5), 5-SRO(6)/STO(6), and 5-SIO(6)/STO(6). After correction with specular reflectivity, the corrected MFP values were obtained as 192, 172, 204, 355 nm for 0.97, 0.75, 0.63, 0.50 THz, respectively. The inset of figure 7 summarizes the uncorrected (in green triangles) and corrected (in blue triangles) MFPs with error ranges. As mentioned previously, the error ranges of the uncorrected MFPs (shown by green lines in the inset of figure 7) were partially attributed to the inhomogeneity of the samples. According to the AFM results, the inhomogeneity most likely comes from different roughness values for different measured spots. The error ranges for corrected data (in blue lines) are larger than that of uncorrected data because statistical ranges of the surface roughness rms values were further considered into correction.

Figure 7 includes the measured MFP values of STO from the literature [18–20, 32–34] and the theoretical curve [34] of Akhiezer’s model (in red dashed line). The results of corrected MFPs in our work seem to still follow the Akhiezer’s model with \( 1/\omega^2 \) dependence in the frequency range between 0.5 and 1 THz. This frequency dependence of MFP is similar to that of GaAs and Si around 1 THz based on first-principle calculations [35]. From the present experimental results, it is challenging to accurately obtain the MFP of STO. Although the model [30, 31] we used to correct the surface scattering loss is qualitatively right, our experimental condition did not meet the criterion of far-field in this model. Our suggested MFPs of STO (in blue triangles of figure 7) also exhibit large uncertainty. Nevertheless, we have still demonstrated the feasibility to study MFP at THz regime by using superlattices of perovskite oxides. And the measured MFP (in green triangles of figure 7) can be treated as the low bound estimates of the intrinsic MFP of STO.

One possible way to improve the correction of surface scattering loss and to increase the measured accuracy of MFP is to measure the surface height distribution of the sample in situ where the optical spot shined. Such surface height distribution can be utilized to correct the delay times of echoes and \( A_{\text{EG}} \) due to the rough surface [36]. The other way is to conduct a modified experiment to avoid surface scattering loss. For example, two sets of superlattices can be used for measurements as demonstrated in InGaN/GaN superlattices recently [24].

### 3.3. Coherent control of coherent acoustic phonons

The CAPs in the superlattice can be coherently manipulated by another optical pulse, called control pulse. Assume the pump pulse initiates CAP oscillations in the superlattice following \( A_p \cos \left( 2\pi ft \right) \). And a time-delayed control pulse generates CAP oscillations \( A_c \cos \left( 2\pi f(t + \phi) \right) \), where \( \phi = 2\pi \tau f \). If the delayed time \( \tau \) is in phase (with \( \phi = 2\pi \tau f \)), the CAP oscillations will be coherently amplified. On the other hand, an
Figure 8. (a) Coherent control of the 0.75 THz CAP oscillations. The black curve shows the CAP oscillations generated by the pump pulse only. The oscillations are amplified (in red curve) or ceased (in blue curve) when the control pulse is introduced in phase or out of phase to the generated oscillations. (b) The amplitudes and (c) phases at 0.75 THz in the Fourier spectra of the CAP oscillations with different delayed times of the control pulse.

out-of-phase $\tau$ (with $\phi = \pi + 2n\pi$) will result in destructive interference. Manipulating amplitudes and phases of acoustic phonons has been experimentally demonstrated in GaAs/AlAs [13] and InGaN/GaN [14, 15] multiple quantum wells. Similar results should be anticipated in the superlattices of perovskite oxides in this work. To further examine the validity of the model in equation (1) for perovskite oxide superlattices, we simulated coherent control of CAP oscillations and quantitatively compared the results with the experimental data. We experimentally demonstrated coherent control of the 0.75 THz CAP oscillations in 10-SIO(3)/STO(5). We have fine controlled the optical fluences of the pump and control pulses that they could individually generate the same amplitude of CAP oscillations. The black curve in figure 8(a) reveals the CAP oscillations induced by the pump pulse only. By introducing the control pulse at delayed time of 0.5 oscillation period ($\sim 0.67$ ps), the CAP oscillations almost cease as shown in blue curve. When the time-delay of the control pulse is 1 oscillation period ($\sim 1.33$ ps) to the pump pulse, the amplitude of the CAP oscillations is enhanced as shown in red curve. We analyzed each 0.75 THz CAP oscillation trace with different delayed times between the pump and control pulses by Fourier transform. The blue dots in figures 8(b) and (c) indicate the amplitudes and phases at 0.75 THz in the Fourier spectra for different delayed times of control. While the timing of the control pulse varies, not only the amplitude but also the phase of the CAP oscillations are manipulated. For simulation, in addition to the two counter-propagating strain pulses generated by the pump pulse similar to figure 1, another propagating strain pulses with the same profiles are created by the control pulse with different time delays. The transient transmission changes $\Delta T(t)$ of the probe beam due to the coherently controlled CAPs were calculated following equation (1) and were Fourier transformed to acquire the resulting FFT amplitudes and phases of CAP oscillations. The red lines in figures 8(b) and (c) show the simulation results. In addition, the black dashed lines show the fitting results from a sinusoidal model with $A_p \cos (2\pi ft) + A_c \cos [2\pi f (t + \tau)]$. The simulation results based on the model in equation (1) reveal a better fit and overall agree well with the experimental data, which confirms the validity of our model to simulate the engineering of CAP oscillations in the superlattices of perovskite oxides.
3.4. Comparison of optoacoustic conversion efficiency between SRO and SIO

In figure 2, we found that the contrast of the CAP oscillations to the background curves for the SIO/STO superlattice is much better than that for SRO/STO superlattices. However, these observations could not lead to the conclusion that the optoacoustic efficiency of SIO for phonon generation is better than that of SRO. The CAP oscillations are both associated with the strain and the sensitivity function as described in equation (1). Both the efficiency of generation and detection of the strain pulses affect the amplitude of the CAP oscillation. If one would compare the generation efficiency of SIO and SRO, the strain pulses should be measured with the same mechanism of detection. Therefore, two additional samples were fabricated and measured to address this issue. One is 6 nm-thick SIO single-layer sandwiched by the 40 nm-thick cap layer STO and the STO substrate. The other is to replace the SIO with the 6 nm-thick SRO layer. We measured the Brillouin scattering in time domain when the strain pulse propagates in the STO substrate, similar to the previous report [20]. Detailed experimental setup of the collinear and reflection type pump–probe measurements can be found elsewhere [37]. The pump/probe fluences were \( \sim 110 \text{ mJ cm}^{-2} \). And the duration of the cross-correlation was \( \sim 1 \text{ ps} \). Figure 9 shows the time-domain Brillouin oscillations from the SIO and SRO samples, and the oscillation frequencies are both 47 GHz. The magnitude of the Brillouin oscillations for SIO and SRO samples are \( 1.53 \times 10^{-4} \) and \( 1.72 \times 10^{-4} \), respectively. With the sample pump fluence, the amplitudes of the generated strain pulses from SIO and SRO are roughly equivalent. However, the absorbed optical energy of SRO is \( \sim 1.6 \) times larger than that of SIO [38]. The efficiency of generating phonons in SIO is thus \( \sim 1.4 \) times better than that in SRO.

4. Conclusions

In summary, we demonstrated that CAPs can be generated and detected in the superlattices of SRO/STO and SIO/STO with acoustic frequency up to 1 THz. We found the optoacoustic conversion efficiency of SIO outperforms that of SRO for generating acoustic phonons. A model was used to account for the transmission of light modulated by the propagating CAPs in the superlattices. And we analyzed the CAP oscillations in the transient transmission traces and developed a method to experimentally estimate the phonon MFP of STO between 0.5 THz and 1 THz. We further demonstrated coherent control to manipulate the amplitude and phase of the THz CAP oscillations. The first demonstration of generating and manipulating CAPs in THz regime should open the gate to experimental studies of THz acoustic properties and phonon engineering in perovskite oxides.

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Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

ORCID iDs

Kung-Hsuan Lin https://orcid.org/0000-0003-2731-5400

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